Radioisotopes in Hydrology

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ERRATA

Pp. 25 resp. 255: The paper "Appareil pour la mesure des vitesses relatives des eaux souterraines par la méthode de dilution ponctuelle" by J. Guizerix et al. should appear on page 255 in place of the paper "Les mesures de débits effectuées en France à l'aide de traceurs radioactifs par la méthode d'intégration" by J. Guizerix et al. which would appear on page 25.

It should be noted, however, that the discussions appearing after these papers are correctly placed and should be taken as referring to the rearranged papers and not to those they follow at present.

p. 200 Line 2 of caption A to Fig. 3 should read as follows:
The product of Carrizozo rainfall and tritium is indicated by the solid line (right scale).
RADIOISOTOPES IN HYDROLOGY
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RADIOISOTOPES IN HYDROLOGY

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The increasing emphasis on the development of water resources poses problems which are of interest to all countries, both developing and advanced, where the demand for water is continuously rising. There is no doubt that greater efforts must be made to evaluate, control and develop water resources using all scientific means available and during recent years increasing attention has been directed to the supplementation of hydrological methods with radioisotope techniques. These techniques have already been applied to a number of problems and their potential usefulness demonstrated.

Radioisotopes can be used for stream discharge measurements with an accuracy as good as that obtainable with conventional methods. They are also finding increasing application in the measurement of groundwater direction and velocity, the study of suspected interconnections between different sources of water, and the investigation of mixing processes in rivers and lakes. Radioisotope techniques have been used in different parts of the world for studying the transport of silt in rivers and harbours. Present research is directed towards making these investigations on a quantitative basis which, if successful, would be of great importance in the design of hydraulic structures.

The method of finding out the age of groundwater by measuring its natural tritium content can be applied to the determination of the recharge rate of groundwater bodies, so enabling a more rational use of the groundwater reserves without fear of overexploitation. Current research is aimed at using carbon-14 for groundwater-dating to extend the age measurable by tritium.

A Symposium on the use of radioisotopes in hydrology was organized by the Agency and held in March 1963 in Tokyo in co-operation with the Japanese Government, for whose material and other assistance and generous hospitality the Agency wishes to record its grateful appreciation. The Symposium was attended by about 100 participants from 14 countries and 5 international organizations.

The 27 papers and the discussions which form the proceedings of the Symposium illustrate that already radioisotope techniques are a valuable adjunct to non-radioactive methods. In some cases they are the method of choice and in others they are even the only method available. It is hoped that this publication will contribute to a more rapid development and use of these particular applications of radioisotopes.
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A COMPARISON OF RADIOISOTOPE METHODS FOR RIVER FLOW MEASUREMENT

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Abstract — Résumé — Аннотация — Resumen

A COMPARISON OF RADIOISOTOPE METHODS FOR RIVER FLOW MEASUREMENT. The Dilution, Continuous Sample and Total Count Methods have been used in a comparative study of radiotracer methods of river flow measurement in the Aylburton Brook in Gloucestershire and the Usway Burn and Alwin Rivers in Northumberland. These are three rivers of differing geological character with flow rates varying between 2.5 l/s and 3 m$^3$/s.

In all tracer methods of flow measurement the distance between the measuring point and the point at which the tracer is introduced should be great enough for complete lateral mixing to have occurred. On the other hand, it should not be so great that longitudinal dispersion is excessive or that tracer is lost by adsorption on the river bed. Dispersion depends on the hydraulic characteristics of the stream and is intrinsic to the method. Adsorption depends on the choice of tracer and on the geological nature of the river bed. From this latter point of view $^3$H is ideal as it is not adsorbed; but it is difficult to measure, and its progress in the river cannot be followed directly. Other tracers are more convenient but they may be adsorbed.

The degree of adsorption of $^{24}$Na, $^{82}$Br and $^{32}$P in the form of NaHCO$_3$, NH$_4$Br and NaH$_2$PO$_4$ has been studied by comparison of measurements at several points downstream of a common injection point and by introduction of tritiated water (HTO) simultaneously with each of the three tracers.

It has been shown that $^{32}$P, which was expected to undergo a high degree of chemisorption, is valueless as a tracer for river flow measurement when used without carrier although there is a significant decrease in error when it is used with carrier.

$^{24}$Na and $^{82}$Br show no evidence of adsorption at distances much greater than that required to give lateral mixing although at a distance of over 660 m in the slowest-moving river there is some evidence of Na removal.

Some observations of the degree of lateral and longitudinal dispersion which affect the general application of tracer techniques to river flow measurement will be presented.

It is concluded that all three methods can be satisfactorily used. The Continuous Sample Method is ultimately preferred as it gives the highest accuracy for the smallest quantity of tracer.

COMPARAISON DE MÉTHODES RADIOISOTOPIQUES DE MÉSURE DU DÉBIT DES COURS D'EAU. Aux fins d'une étude comparative des méthodes radioisotopiques de mesure de débit fluvial, on a employé les méthodes de dilution, d'échantillonnage continu et de comptage total sur l'Aylburton (Gloucestershire), l'Usway et l'Alwin (Northumberland). Ces trois cours d'eau présentent des particularités géologiques différentes et leur débit varie entre 2,5 l et 3 m$^3$/s.

Dans toutes les méthodes de mesure de débit faisant appel à l'emploi d'indicateurs, il est souhaitable que la distance entre le point où la mesure est effectuée et celui où l'indicateur est introduit soit suffisamment grande pour permettre un brassage latéral complet. D'autre part, cette distance doit être suffisamment réduite pour que la dispersion longitudinale ne soit pas excessive ou que l'indicateur ne se perde pas par adsorption sur le lit. Le mécanisme de dispersion est lié aux caractéristiques hydrologiques du cours d'eau et à la méthode utilisée. L'adsorption est fonction du choix de l'indicateur et de la nature géologique du lit de la rivière. Le tritium est idéal à cet égard car il n'est pas adsorbé, mais il est difficile à mesurer et on ne peut pas suivre directement son mouvement dans la rivière. D'autres indicateurs conviennent mieux, mais ils peuvent être adsorbés.

On a étudié le degré d'adsorption de $^{24}$Na, $^{82}$Br et $^{32}$P sous forme de NaHCO$_3$, NH$_4$Br et NaH$_2$PO$_4$, en comparant les mesures faites en plusieurs points situés en aval d'un point unique d'injection et en introduisant de l'eau tritiée (HTO) en même temps que chacun des trois indicateurs.
Il s'est révélé que \(^{32}\)P est sujet à une forte chimiosorption, comme on le supposait, et n'est pas employé avec un entraîneur; mais avec un entraîneur, on observe une amélioration marquée de l'exactitude des résultats. \(^{32}\)P et \(^{82}\)Br n'ont accusé aucune adsorption à des distances bien supérieures à celles qui sont nécessaires pour assurer une brassage latéral; dans la rivière la plus lente, toutefois, on a noté, à plus de 660 m du point d'injection, une certaine diminution de \(^{32}\)Na.

Le mémoire contient quelques observations sur la dispersion latérale et longitudinale qui, à une certaine degré, influent sur l'application générale des méthodes radioisotopiques à la mesure du débit des cours d'eau. Les auteurs paraissent à la conclusion que les trois méthodes donnent des résultats satisfaisants. Ils préfèrent, en fin de compte, la méthode d'échantillonnage continue, car elle donne les résultats les plus exacts pour la quantité minimum d'indicateur.

СРАВНЕНИЕ РАДИОИЗОТОПНЫХ МЕТОДОВ ИЗМЕРЕНИЯ РЕЧНЫХ СТОКОВ. Методы разбавления, непрерывного отбора проб и полного счета применяются при сравнительном изучении радиоиндикаторных методов измерения стоков в реках Aylburton Brook в Глостершире и Usway Burn и Alvin Rivers в Нортумберленде. Это три реки с различными геологическими характеристиками и скоростями стоков, изменяющихся в диапазоне от 2,5 м³/с до 3 м³/сек.

При использовании любых индикаторных методов измерения стоков расстояние между точкой измерения и точкой введения индикатора должно быть достаточно большим, поскольку должно произойти полное смешение. В то же время это расстояние не должно быть слишком большим, чтобы избежать значительной продольной дисперсии или потери индикатора в результате поглощения его дном реки. Бисперсия зависит от гидравлических характеристик отока, и она характерна для этого метода. Поглощение зависит от выбора индикатора и от геологической природы дна реки. Именно с этой точки зрения \(^{3}H\) является идеальным индикатором, поскольку он не поглощается. Однако его трудно измерить, и его прохождение в реке не может быть прослежено непосредственно. Другие индикаторы более удобны, но они могут поглощаться.

Степень поглощения \(^{24}\)Na, \(^{82}\)Br и \(^{32}\)P в виде \(\text{NaHCO}_3\), \(\text{NH}_4\text{Br}\) и \(\text{NaH}_{2}\text{PO}_4\) изучалась путем сравнения данных измерений в нескольких точках вниз по течению о общей точки введения и путем введения меченной тритием воды (HTO) одновременно с введением каждого из трех индикаторов.

Установлено, что \(^{32}\)P, который, как ожидается, будет сильно поглощаться химически, является на самом деле непригодным в качестве индикатора без носителя для измерения речных стоков, хотя с носителем ошибка измерений значительно меньше. \(^{24}\)Na и \(^{82}\)Br не проявляют признаков поглощения их на значительных расстояниях, больше чем необходимо для горизонтального смешения; на расстоянии свыше 660 м в реке с самым медленным течением обнаруживаются признаки исчезновения \(^{24}\)Na.

Представлены некоторые наблюдения относительно степени горизонтальной и продольной дисперсии, которая отражается на общем применении индикаторных методов для измерений речных стоков.

Делается вывод, что все три метода могут успешно применяться. Однако метод непрерывного отбора проб в конечном счете более предпочитителен, поскольку он дает более высокую точность при минимальном количестве индикатора.

COMPARACIÓN DE LOS MÉTODOS RADIOISOTÓPICOS PARA MEDIR EL CAUDAL DE LOS RÍOS. Los autores han empleado los procedimientos de dilución, muestreo continuo y recuento total en un estudio comparativo de los métodos radioisotópicos de medición de caudales fluviales en el arroyo Aylburton, en Gloucestershire, y en los ríos Usway Burn y Alvin, en Northumberland. Se trata de tres ríos de características geológicas diferentes, cuyos caudales oscilan entre 2,5/4 y 3 m³/seg.

En todos los métodos de medición de caudales que emplean indicadores, la distancia entre el punto en que se efectúa la medición y el punto en que se añade el indicador tiene que ser suficientemente grande para que se produzca una mezcla lateral completa. Por otra parte, no debe ser excesiva para evitar que la disipación longitudinal supere cierto valor o que se pierda parte del indicador por adsorción en el lecho fluvial. La disipación depende de las características hidráulicas y de la corriente y es inherente al método. Por su parte, la adsorción depende del indicador elegido y de las características geológicas del lecho del río. Desde este punto de vista, el tritio posee excelentes propiedades, puesto que no es absorbido, pero es difícil de medir y no es posible seguir directamente su desplazamiento en el río. Otros indicadores resultan más convenientes, pero pueden sufrir adsorción.
COMPARISON OF ISOTOPE METHODS

Los autores han estudiado los grados de adsorción del $^{24}$Na, $^{82}$Br y $^{32}$P en forma de NaHCO$_3$, NH$_4$Br y NaH$_2$PO$_4$ respectivamente, que introdujeron en un mismo punto del río junto con agua tritiated (HTO), para comparar después las mediciones realizadas en varios lugares situados aguas abajo.

Se ha visto que el $^{32}$P, para el que se esperaban grandes pérdidas por quimisorción, no sirve para medir el caudal de ríos cuando se emplea sin portador, aunque el error disminuye notablemente cuando se trabaja con portador.

En el caso del $^{24}$Na y del $^{82}$Br, no se observa adsorción a distancias mucho mayores que la requerida para que la mezcla lateral sea satisfactoria si bien a una distancia de 660 m en el río de corriente más lenta se observó que parte del $^{24}$Na había desaparecido.

Los autores formulan algunas observaciones sobre el grado de dispersión lateral y longitudinal que tienen importancia para la aplicación de técnicas radioisotópicas en general a la medición del caudal de ríos.

Llegan a la conclusión de que los tres métodos pueden dar resultados satisfactorios. En definitiva, los autores dan preferencia al método de muestreo continuo, ya que permite alcanzar una mayor precisión para un consumo mínimo de indicador.

1. INTRODUCTION

The use of radioactive isotopes for the measurement of flow has many advantages over conventional velocity × area methods; and, although isotopes cannot be used for continuous measurement, they provide an important means for the calibration of permanent hydraulic structures.

The Dilution, Continuous Sample and Total Count Methods are three systems of flow measurement which are already accepted as providing an accurate method of flow measurement in closed conduits [1]. Their application to river flow is not yet widely established although some work has been reported [2, 3], and it is the purpose of this paper to examine the validity of the methods and to illustrate some of the problems which occur when the techniques are applied to natural streams.

All three methods of flow measurement are based on the injection of a tracer at some point in the river and the subsequent measurement of concentration at a point sufficiently far downstream for complete lateral mixing to have occurred. The correct choice of this point is important to the successful operation of the methods. If the measuring point is too close to the injection point, errors result from incomplete mixing. If the measuring point is too distant, excessive dispersion occurs and the tracer is over-diluted. The tracer must also be selected to minimize losses by adsorption on the river bed. Tritium, which is available as tritiated water, overcomes this problem though its measurement presents some difficulty.

The present investigations were undertaken for comparison of the accuracy of results with three techniques of flow measurement and several different radioactive tracers and for examination of their practical application in the field. In particular, the magnitude of lateral and longitudinal dispersion and the possibility of tracer adsorption was examined.

A small stream (Aylburton Brook, flow 2.6 l/sec) was used for investigation of the behaviour of the tracer under conditions of large dispersion where long transit times allowed measurements to be made at many times the optimum measuring distance.

A small river (River Alwin, flow 260 l/sec) was selected as typical of fast-flowing streams in hilly country with fairly uniform width and depth. Tests for examination of dispersion and adsorption and for comparison of the different techniques were carried out over two stretches at sampling points at various distances from the point of injection.
Tests were also carried out in a second, more mountainous river containing a rectangular weir, and comparisons were made against the weir for flows between 1.7 m$^3$/sec and 3.0 m$^3$/sec.

2. METHODS OF FLOW MEASUREMENT WITH RADIOACTIVE ISOTOPES

2.1. The Dilution Method

Measurement of flow by the Dilution Method is based on a comparison between the concentration $C_1$ of a tracer, introduced into the river at a constant and known rate $q$, with the concentration $C_2$ of samples removed from some position sufficiently far downstream from the injection point for complete lateral mixing to have occurred.

The mass flow of the injected tracer is equal to the mass flow of the tracer as it arrives at the sampling station. Thus,

$$qC_1 + QC_0 = (Q + q)C_2,$$

where $C_0$ is the initial concentration in the stream flowing at a rate $Q$. Hence,

$$Q = q(C_1 - C_2)/(C_2 - C_0). \tag{1}$$

Generally $C_1 \gg C_2$ and $C_2 \gg C_0$ so that equation (1) reduces to

$$Q = q(C_1/C_2). \tag{2}$$

The flow rate can thus be determined absolutely simply by comparison of the concentration of the injected solution with the concentration of samples removed from the stream. The method is completely independent of the velocity of flow and the dimensions of the river bed or variations in these dimensions, should they occur. From equation (2) it is easy to show that $dQ/Q = dC_2/C_2$, i.e. the relative error in the measurement of the flow is equal to the relative error in the measurement of the concentration of the tracer.

An important limitation to the operation of the Dilution Method is the need to remove samples over a finite period of time when the tracer concentration is constant; but, unless some indicator such as a radiation monitor is used, the presence of a region of uniform concentration at the sampling point is not evident. In these circumstances a very large number of samples is required to guarantee coverage of the transit period. When the total dispersion is large (as occurs if the measuring reach is long, or the river is wide and contains stagnant pools, or if the river bed is rocky), equilibrium conditions are never established except with an excessively long injection period and a consequently large injected quantity of radio-tracer.

In practice, the need for inclusion of a continuous and controlled injection system in the procedure can also be a serious limitation to field application. The transport and operation of any type of injector is to be avoided if possible since, apart from such practical difficulties, their use increases the amount of radioactive handling which is necessary.
2.2. The Continuous Sample Method

In this method a volume $V_1$ of solution with radioisotope concentration $C_1$ is injected rapidly into the stream, and the concentration $C$ in the stream is examined at a point sufficiently far downstream to ensure adequate mixing (see section 3.1).

The mass flow equation is now given by

$$C_1V_1 = \int C \, dv,$$  \hspace{1cm} (3)

where $v$ is an elemental volume in the streams.

The total volume $v$ which passes a given point is related to the constant flow rate $Q$ by $v = Qt$. Hence $dv = Qdt$, and

$$C_1V_1 = Q \int C \, dt.$$  \hspace{1cm} (4)

Since $V_1C_1 = A$, the total activity of radioisotope introduced into the stream, the flow rate is given by

$$Q = A / \int C \, dt.$$  \hspace{1cm} (5)

For this expression to be valid for a single sampling point, $\int C \, dt$ must be constant over the entire cross-sectional area.

For determination of the flow rate it is simply necessary to know the total quantity of radioisotope introduced and the value of the integral. There are two ways by which the latter may be determined:

(i) The concentration may be measured continuously by sampling at a fixed point and a concentration time curve constructed and the integral evaluated numerically. Since the concentration is changing smoothly, a strictly continuous measurement is not necessary; and the curve may be derived by measurement of the concentration of discrete samples removed periodically, or

(ii) Using $\int C \, dt = \bar{C} (t_2 - t_1)$, where $\bar{C}$ is the mean concentration over the sampling interval $(t_2 - t_1)$. Provided that $Q$ is constant, $C$ can be determined from measurements of the concentration of samples removed at equal time intervals and mixed together in a suitable container.

The principal advantages of the Continuous Sample Method as compared with the Dilution Method are the following:

(a) It is economical in equipment since apparatus for a continuous injection is not required.

(b) It is less sensitive to the position of the sampling station, provided that the basic requirement of adequate mixing is satisfied, as the problem of operating in a region of constant concentration no longer applies.

(c) By mixing samples removed during the transit period, a single measurement only is sufficient for the determination of flow.

(d) If the concentration $\bar{C}$ of the single sample is made equal to that of a Dilution sample, the amount of activity required to operate
the Continuous Sample Method is approximately half that required for the Dilution technique.

(e) Under conditions of high dispersion a region of uniform concentration may not be established. The Dilution Method then fails completely whereas the Continuous Sample Method is still valid at any sampling point where lateral mixing is complete.

(f) Operation of the Dilution Method depends critically on samples being removed during the period of constant concentration. Such a restriction does not apply in the Continuous Sample Method, and only an approximate knowledge of the tracer distribution is required. Samples must be removed before the arrival of the tracer and may be continued until well after the estimated transit time.

Although in general neither the Dilution nor the Continuous Sample Method is capable of continuous operation since samples have to be removed from the river for assay, measurement of the concentration of samples on site is possible in many locations.

2.3. The Total Count Method

If a counter were to be inserted into the stream at the sampling point in the Continuous Sample Method, the number of counts \( N \) recorded during the passage of the tracer would be given by

\[
N = F \int C \, dt,
\]

where \( F \) is a constant which depends upon the efficiency of the detector.

From equation (6) \( \int C \, dt = N/F \); and, inserting this result in equation (5), we get

\[
Q = AF/N,
\]

which is the equation derived by HULL [2] for the Total Count Method.

In practice \( F \) is obtained by calibration of the detector; and, since this must be carried out under the same geometrical conditions as is the flow measurement, it is more convenient not to immerse the detector in the river but to define a reproducible geometry by the use of a re-entrant chamber through which river water is pumped at a constant rate. This chamber can then be calibrated with a radioactive solution of known specific activity.

The principal disadvantages of the Total Count Method for flow measurement are the low statistical accuracy resulting from a total counting period restricted to the pulse transit time and the need for detecting equipment to operate reliably whilst the tracer is passing.

If counting equipment is available on site, it is doubtful if the Total Count Method can ever compete with the Continuous Sample Method since the concentration of each of a large number of samples or of a single total sample can be measured to as high a statistical accuracy as desired as soon as they are removed from the flow.
3. SOME GENERAL COMMENTS ON THE USE OF TRACERS FOR RIVER FLOW MEASUREMENT

3.1. The mixing length

It is a necessary condition for accurate measurement by any tracer technique that the tracer, which may be injected rapidly or continuously for a finite period of time, should be mixed uniformly with the stream before samples are removed for measurement. The distance needed to achieve such mixing is called the mixing length and is defined as the minimum distance at which the mass transfer and the concentration transfer through a small volume at any point of the cross-section are equal, i.e. \( \frac{dc}{c} = \frac{dm}{m} \).

In the case of a continuous injection, the tracer concentration is uniform throughout any cross-section downstream of the mixing length. Consequently, a rough visual indication of the mixing length may be made by the introduction of a concentrated solution of dye at the injection point and the observation of its general spread up to the selected measuring point. Alternatively, a gamma-emitting tracer and two radiation monitors situated at the centre and close to one bank may be used. The mixing length is exceeded when the detector readings are equal. This technique is particularly useful if stagnant zones are present in the measuring reach and gives a useful qualitative estimate of the mixing length.

With a rapid "pulse" injection, however, equal concentration across the width of the stream is never reached, because velocities vary across the width and lateral and longitudinal dispersions are different. However, we believe that visual estimation of the mixing length, as suggested above, is a useful basis on which to establish the sampling point when a pulse-injection system is used.

Several attempts have been made to calculate the mixing length. Assuming that vertical mixing is much more rapid than is transverse mixing, RIMMAR [4] has considered the problem as a two-dimensional eddy diffusion problem and finds the mixing length \( L_{mix} \) to be given by

\[
L_{mix} = 0.13 \left( \frac{B^2}{H} \right) \left( \frac{C}{g} \right) (0.7C + 6), \tag{8}
\]

where \( H \) (m) is the depth of the stream, \( B \) (m) the breadth, \( g \) (MKS) gravitational acceleration and \( C \) the Chezy coefficient. Usually \( 15 < C < 50 \), depending on the nature of the river bed.

Assuming that transverse and longitudinal mixing coefficients are equal and that the dispersion is a true Gaussian function, Hull finds the minimum distance for complete mixing to be given by

\[
L_{mix} = 50 Q^{1/3},
\]

where \( Q \) is the total mass flow (m\(^3\)/sec). Centre point injection is assumed, and no account is taken of frictional drag.

Once the tracer is completely mixed, samples may, in principle, be removed from any point further downstream. In practice, however, in a natural stream decrease in flow may occur because of seepage, and increase may arise from surface water and groundwater. Seepage which occurs
between the mixing distance and the sampling point will not affect the concentration of tracer and will therefore go undetected. Surface water and groundwater, if completely mixed at the sampling point, will be measured directly. However, for accurate measurement a test section should be chosen in which increase from groundwater and seepage are both negligible.

If it were possible to locate the position at which complete lateral mixing first occurred, this would be the optimum sampling point for all methods. Any extension beyond this distance results in increased longitudinal dispersion and therefore necessitates the use of larger amounts of radioactive tracer.

3.2. Choice of tracer

A practical limitation to the permissible length of test section results from the possibility of tracer adsorption, both by matter in suspension and by material of the river bed. The degree of adsorption depends on the tracer material, which must be chosen to minimize the effect.

The adsorption of radioactive isotopes in very dilute solutions has been studied in connection with the use of tracers to follow underground water, and it has been found that in general anions are adsorbed less than cations [5]. Suitable tracers include Br⁻, I⁻ and complex compounds such as chromium EDTA.

Cationic tracers can be used if the river already contains inactive cations of the same type so that loss of radioactive tracer is reduced. Since Na⁺ normally exists in trace amounts in most rivers, Na²⁴ is a satisfactory tracer.

The problem of adsorption is overcome completely by the use of H³. Only an isotope effect could account for its partial removal from the water; and, even if this occurred, the error involved would be negligible.

Apart from the problem of adsorption, choice of a particular isotope for flow measurement also depends on the following properties:

(i) Stability and solubility in water,
(ii) Radioactive half-life,
(iii) Type and energy of emitted radiation,
(iv) Maximum permissible level in drinking water, and
(v) Maximum specific activity available.

A compromise choice is usually necessary. A long half-life is more convenient for handling and measurement but should be avoided from the point of view of drinking water pollution. For small scale investigations it may be possible to use a short-lived daughter isotope such as iodine-132 (T½ = 2.26 h), which can be prepared on site from the 77-h parent, tellurium-132.

The most sensitive measurements can be made with large volumes containing a high-energy gamma emitter, but these isotopes are difficult to transport in large amounts. A pure beta emitter is more easily handled, but the limited counting volume results in a low sensitivity of measurement. An increase in sensitivity can be obtained by the use of quantitative chemical extraction.

Tritium is unique in a consideration of radioactive water tracers. It is not subject to adsorption; it is available in high specific activity and is not expensive. In spite of its long radioactive half-life, it is one of the least
COMPARISON OF ISOTOPE METHODS

4. EXPERIMENTAL INVESTIGATIONS

4.1. Rivers

The experimental investigation was carried out principally on a small stream (Aylburton Brook, Gloucestershire) and a small mountain river (River Alwin, Northumberland). Some information was also obtained from measurements made on a second mountain river (Usway Burn, Northumberland) during the calibration of a weir. Characteristics of the water from all three water courses are shown in Table I. The Na⁺ content assists as carrier when Na²⁴ is used as tracer.

<table>
<thead>
<tr>
<th>River</th>
<th>pH</th>
<th>Hardness CaCO₃ (ppm)</th>
<th>Na⁺ (ppm)</th>
<th>Cl⁻ (ppm)</th>
<th>Conductivity (μmho/cm)</th>
<th>Total solids (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aylburton Brook</td>
<td>8.3</td>
<td>151</td>
<td>8.3</td>
<td>31</td>
<td></td>
<td>111</td>
</tr>
<tr>
<td>River Alwin</td>
<td>8.0</td>
<td>53</td>
<td>7.8</td>
<td>Trace</td>
<td>126</td>
<td>107</td>
</tr>
<tr>
<td>Usway Burn</td>
<td>7.1</td>
<td>26.5</td>
<td>4.0</td>
<td>Trace</td>
<td>39.2</td>
<td>120</td>
</tr>
</tbody>
</table>

The Aylburton Brook is a small stream with a summer flow rate of about 2.5 l/s, an average gradient of 1.9% and a mean flow velocity of 8 cm/s. The width varies between 50 cm and 75 cm, and the average depth is 5 cm with occasional areas up to 15 cm deep. One tributary in the test sections added about 20% to the flow, and one farm drain may have discharged small quantities intermittently. The bed rock is Old Red Sandstone, and the stream contains small boulders and some silt and vegetation.

The River Alwin is a small mountain river with a flow rate of about 260 l/s, an average gradient of 1.1% and a mean flow velocity of 25 cm/s. The river width is about 6 m and the depth between 15 cm and 30 cm without any large pools or stagnant areas. The stream bed is made up of an alluvial deposit of sand and small boulders of andesite, which produces a flat valley bottom in the hills of igneous rock and lavas. Two test sections were used. One test section in the lower valley was without tributaries, and one in the upper valley included a tributary which entered at about 250 m below the
injection point. Immediately below this tributary the river flowed over a shallow area and bifurcated for about 30 m.

The Usway Burn is also a mountain river, adjacent to the River Alwin, but traverses more rugged ground and, unlike the Alwin, includes a number of pools and deep areas. The average gradient is 1.7%. The river width varies between about 3 m and 6 m, and the depth varies from 20 cm up to 1.5 m. At a flow rate of 1.7 m$^3$/s the average velocity was 85 cm/s. Some of the measurements were made during a period of flood when the river was flowing between 3.0 m$^3$/s and 1.7 m$^3$/s, and the pools then had little effect on dispersion.

4.2. Detection equipment and sample measurement

Na$^{24}$, Br$^{82}$, P$^{32}$ and H$^3$ were used as tracers during these investigations. The samples for $\gamma$-counting (Na$^{24}$ and Br$^{82}$) were taken in 2.2-l polythene bottles. These were then measured with a conventional scintillation counter by transfer of 2.2 l to a polythene bottle 15 cm high $\times$ 12 cm in diameter with a re-entrant base to accommodate a 2-in by 1.5-in diameter NaI(Tl) crystal. The overall efficiency of the detector was 5% for Na$^{24}$, and the background when shielded with 8 cm of lead was about 500 counts/min. About 70% of the samples were counted to provide a theoretical rms deviation of $\pm$ 1%. The deviation associated with the remainder never exceeded $\pm$ 1%.

The Na$^{24}$ samples from Aylburton Brook also contained variable concentrations of P$^{32}$ (relative to Na$^{24}$) which was detected (principally through its
COMPARISON OF ISOTOPE METHODS

bremsstrahlung) with an efficiency of about 0.07%. A thin brass shield over the crystal reduced this to 0.03%, but the correction involved was between 10% and 30% and consequently reduced the accuracy of these Na\textsuperscript{24} results.

The P\textsuperscript{32} samples were measured with a thin-walled Geiger counter surrounded by a sampling jacket containing 10 ml of liquid (20th Century M6H). The counter was 10% efficient and had a background of 20 counts/min within lead shielding. The P\textsuperscript{32} was counted after six days for decay of the Na\textsuperscript{24} to a negligible level were allowed.

Tritium measurements were made with a liquid scintillation system with two photomultipliers in coincidence. Water samples of 1 ml were mixed with 8 ml of scintillator consisting of 100 g/l naphthalene and 6 g/l PPO in redistilled dioxane and counted with 10% efficiency in the presence of a coincidence background of 100 counts/min. An automatic unit counted 24 samples for two separate periods of 10 min each.

All the flow measuring techniques require a comparison between the specific activity of the injected solution and that of the river samples. The concentrated solutions were diluted to approximately the same specific activity as that of the river samples and were measured in the same system. These dilutions, termed "standards", were prepared by the weighing of small aliquots of the original solution (with density corrections made where necessary) and dilution in standard volumetric flasks. At least two standards were prepared independently to avoid accidental errors, and in a series of 12 pairs of standards the average deviation of one standard from the mean of the two was 0.45%, with a maximum spread of 0.6%.

4.3 Isotope handling

The gamma-emitting isotopes were transported to the site as tablets in lead containers.

The Na\textsuperscript{24}, in the form of sodium bicarbonate, was dissolved in warm, dilute acetic acid before use and made up with river water to a volume sufficient for several experiments. The solution was stored in a shielded container and mixed for several minutes with a hand-operated air pump before use.

The Br\textsuperscript{82}, in the form of ammonium bromide, was dissolved in a solution of sodium thiosulphate to convert the free bromine produced by the Szilard-Chalmers reaction during irradiation to sodium bromide.

The P\textsuperscript{32} was in solution in dilute hydrochloric acid with 0.02 g/l of potassium hydrogen phosphate as carrier.

The tritium was in the form of tritiated water and was contained in sealed ampoules. The required amount was added to the sodium and bromine solutions before injection.

4.4 Flow measurement techniques

(i) The Dilution Method

This technique requires the use of constant-flow-rate injection equipment of high stability. Two instruments, both of which were of a positive displacement type and used a piston driven at constant speed in an accurately
machined cylinder, were employed. The displaced tracer solution was delivered in the centre of the river through a narrow bore tube.

In one instrument the piston was driven through gears by a synchronous motor running from a 50-c/s, 230-V supply produced by a constant-speed petrol generator, which was part of the equipment of a mobile laboratory. The generator frequency was measured accurately during each run of 30 min, during which the pump delivered 590 ml of solution. The second instrument was driven by a 24-V battery-operated solenoid controlled by a mechanical clock producing 1-s impulses and giving a pulse discharge of 1950 ± 1 ml in 26 min.

The arrival of the tracer at the sampling point was observed with a portable scintillation counter and ratemeter, type 1413, used in conjunction with a spark recorder. When constant conditions were reached, samples were removed in 2½-l polythene bottles at intervals of one or two minutes. Usually ten samples were taken for each injection.

Solutions for the preparation of "standards" were taken from the injector at the end of the injection period.

(ii) The Continuous Sample Method

The tracer was introduced by the pouring of either a weighted or volumetrically measured amount into the centre of the river. The only apparatus used was a measuring cylinder and handling tongs 1 m long. Most of the injected solutions were mixtures of Na$_{24}$ and tritium or Br$_{82}$ and tritium, but in some experiments tritium was the only tracer. On these occasions the tritium was injected from a screw-top bottle prepared in the laboratory to minimize handling and thus reduce the possibility of contamination on site.

Two sets of samples were taken at a series of fixed sampling points. The samples for gamma counting consisted of 625-ml samples, four of which were mixed in a single 2½-l polythene bottle. Simultaneously small samples were removed for tritium assay in 7-ml polythene "pill packs" or in 25-ml screw-top glass bottles. The sampling interval at a given sample point was constant and determined by the estimated transit time of the tracer. For sample stations close to the injection, samples were removed at intervals of 15 or 20 s and for the more distant sampling stations at intervals of 2 min. A portable detector 50 m upstream of the sampling point indicated the arrival of Na$_{24}$ or Br$_{82}$, and sampling was started at the first indication of a reading on the instrument so as to ensure that the beginning of the pulse was fully recorded. Sampling was continued well beyond the time at which the monitor returned to natural background.

(iii) The Total Count Method

The same injection was used for the Total Count system as for the Continuous Sample Method. The measuring point was at 300 yd, which was considered to be the optimum distance for operation of the method.

The total count was obtained from a 2-in by 1-in diameter NaI(Tl) crystal mounted in a re-entrant brass container. Water from the river was pumped at a fairly constant rate through the container by means of a small, electrically powered centrifugal pump producing a flow of about 125 ml/s. In the investigations on the Aylburton Brook the re-entrant container had a volume of 500 ml, but this was increased to 1850 ml for all other work to improve the sensitivity. The containers were placed in holes in the ground to minimize contributions from the tracer passing in the river. Since the counters
were shielded with soil, the background depended on the nature of the ground and was about 4000 counts/min at Aylburton Brook and between 9000 and 10 000 counts/min at the River Alwin.

The detection system, which included a fast scaler with a resolving time of 5 μs to accommodate the high counting rate during the passage of maximum tracer concentration, was powered by a petrol generator. The detection apparatus used at Aylburton Brook was calibrated in the laboratory, and that used at the River Alwin was calibrated on site.

4.5. Health considerations

In these investigations the concentrations of radioisotopes in the measuring reaches were well below the maximum permissible levels for drinking water as recommended by the I. C. R. P. [6]. These levels are below those at which hazard could arise to fish or fish eggs in the river.

In a practical measurement of river flow, it is desirable that the least toxic isotope should be used at a level sufficient to give the required statistical accuracy. By means of the equipment described in this paper, a flow measurement can be carried out with a statistical accuracy of within 1% in a time of 30 min counting with a sample concentration of Na\textsuperscript{24} or Br\textsuperscript{82} of 1.5 μc/m\textsuperscript{3}, or 0.2% of the I. C. R. P. recommended maximum permissible level for non-occupationally exposed workers. The corresponding value for H\textsuperscript{3} would be 1.5 mc/m\textsuperscript{3}, or 15% of the I. C. R. P. value.

5. RESULTS

Results of flow measurements on the three rivers are presented in Tables II to VI.

Comparisons between the Continuous Sample and Dilution Methods were made on all rivers over the full range of flow rates available, but the Total Count Method was only operated at a single distance on the Aylburton Brook and the River Alwin. These results are shown in Tables II and IV.

Phosphorous was used on the Aylburton Brook, and the loss of this tracer resulting from adsorption on the river bed is shown in Table VI. Evidence of Na\textsuperscript{24} adsorption at long distances is given in Table II.

Comparative measurements with a rectangular weir on the Usway Burn are shown in Table V.

Longitudinal dispersion is illustrated in Figs. 2, 3 and 4. Variation in the lateral distribution of tracer with distance following a continuous, single point injection on the River Alwin is shown in Fig. 5.

6. DISCUSSION

6.1. Comparison of techniques

Results obtained with the Continuous Sample and Dilution Methods using Br\textsuperscript{82} and Na\textsuperscript{24} show a systematic error of 3.1% with a random standard deviation of ±2.4%. One additional comparison carried out at a point where
TABLE II

FLOW RESULTS, AYLBURTON BROOK
(Tritium results shown in brackets below Na$^{24}$ results)

<table>
<thead>
<tr>
<th>Injection data</th>
<th>Sample distance (m)</th>
<th>Flow rate (1/sec)</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Continuous sample</td>
<td>Dilution</td>
</tr>
<tr>
<td>Continuous Injection WITHOUT CARRIER 30 min Na$^{24}$, 2.1 mc H$^{3}$, 470 mc For P$^{32}$ (5.9 mc) see Table VI</td>
<td>98</td>
<td>2.40 (2.41)</td>
<td>2.33</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>253</td>
<td>2.56 (2.51)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>660</td>
<td>3.89 (3.48)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1460</td>
<td>5.29 (4.51)</td>
<td></td>
</tr>
<tr>
<td>Continuous Injection WITH CARRIER 30 min Na$^{24}$, 2.1 mc H$^{3}$, 470 mc For P$^{32}$ (5.9 mc) see Table VI Carrier (1600 g NaCl and 250 g KH$<em>{2}$PO$</em>{4}$ added in 30 min)</td>
<td>56</td>
<td>2.28 (2.54)</td>
<td>2.41 (2.47)</td>
</tr>
<tr>
<td></td>
<td>98</td>
<td>2.34 (2.37)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>253</td>
<td>2.78 (2.77)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>660</td>
<td>3.50 (3.51)</td>
<td></td>
</tr>
<tr>
<td>Pulse injection Na$^{24}$ 0.75 mc</td>
<td>253</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
### TABLE III

**FLOW RESULTS, RIVER ALWIN, LOWER VALLEY**  
(Tritium results shown in brackets below Na\textsuperscript{24} or Br\textsuperscript{82} results)

<table>
<thead>
<tr>
<th>Injection data</th>
<th>Sample distance (m)</th>
<th>Flow rate (l/s)</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Continuous sample</td>
<td>Dilution</td>
</tr>
<tr>
<td>Continuous injection for 30 min</td>
<td>91</td>
<td>268 (279)</td>
<td>273 (269)</td>
</tr>
<tr>
<td>Na\textsuperscript{24}, 90 mc</td>
<td>274</td>
<td>289 (304)</td>
<td>295 (319)</td>
</tr>
<tr>
<td>H\textsuperscript{3}, 4.2 c</td>
<td>823</td>
<td>316 (313)</td>
<td>316 (313)</td>
</tr>
<tr>
<td>Continuous injection for 30 min</td>
<td>91</td>
<td>248 (275)</td>
<td>292 (323)</td>
</tr>
<tr>
<td>Br\textsuperscript{82}, 30 mc</td>
<td>274</td>
<td>272 (296)</td>
<td>289 (338)</td>
</tr>
<tr>
<td>H\textsuperscript{3}, 4.2 c</td>
<td>823</td>
<td>287 (307)</td>
<td>287 (307)</td>
</tr>
<tr>
<td>Pulse injection</td>
<td>91</td>
<td>273 (286)</td>
<td>313 (305)</td>
</tr>
<tr>
<td>Br\textsuperscript{82}, 9 mc</td>
<td>274</td>
<td>313 (305)</td>
<td>308 (304)</td>
</tr>
<tr>
<td>H\textsuperscript{3}, 1 c</td>
<td>823</td>
<td>308 (294)</td>
<td>308 (294)</td>
</tr>
<tr>
<td></td>
<td>1000</td>
<td>313 (266)</td>
<td>313 (266)</td>
</tr>
</tbody>
</table>
### TABLE IV

FLOW RESULTS, RIVER ALWIN, UPPER VALLEY

<table>
<thead>
<tr>
<th>Injection data</th>
<th>Sample distance (m)</th>
<th>Flow rate (1/4)</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Continuous sample</td>
<td>Dilution</td>
</tr>
<tr>
<td>Continuous injection</td>
<td>100</td>
<td>225</td>
<td>232</td>
</tr>
<tr>
<td>for 30 min Na\textsuperscript{24}, 28 mc</td>
<td>273</td>
<td>229</td>
<td>240</td>
</tr>
<tr>
<td></td>
<td>373</td>
<td>234</td>
<td>265</td>
</tr>
<tr>
<td></td>
<td>525</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pulse injection Na\textsuperscript{24}, 4 mc</td>
<td>100</td>
<td>168</td>
<td></td>
</tr>
<tr>
<td></td>
<td>273</td>
<td>242</td>
<td></td>
</tr>
<tr>
<td></td>
<td>373</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td></td>
<td>525</td>
<td>258</td>
<td></td>
</tr>
<tr>
<td>Pulse injection Na\textsuperscript{24}, 3 mc</td>
<td>273</td>
<td>238</td>
<td></td>
</tr>
<tr>
<td>Pulse injection Na\textsuperscript{24}, 4 mc</td>
<td>273</td>
<td>-</td>
<td></td>
</tr>
</tbody>
</table>
### TABLE V

**FLOW RESULTS, USWAY BRUN**

<table>
<thead>
<tr>
<th>Injection data</th>
<th>Sample distance (m)</th>
<th>Flow rate (m³/s)</th>
<th>Weir</th>
<th>Time</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Continuous sample</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Continuous injection for 30 min</td>
<td>230</td>
<td>2.77</td>
<td>2.79</td>
<td>2.16</td>
<td></td>
</tr>
<tr>
<td>Na²⁺, 52 mc</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Continuous injection 30 min</td>
<td>230</td>
<td>2.32</td>
<td>1.82</td>
<td>15.40</td>
<td>Falling flow rate indicated by individual dilution samples</td>
</tr>
<tr>
<td>Br⁻, 62 mc</td>
<td></td>
<td></td>
<td>2.28</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Continuous sample</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Continuous injection 26 min</td>
<td>230</td>
<td>1.71</td>
<td>1.68</td>
<td>1.35</td>
<td></td>
</tr>
<tr>
<td>Br⁻, 26 mc</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pulse injection</td>
<td>230</td>
<td>1.65</td>
<td>1.65</td>
<td>1.33</td>
<td></td>
</tr>
<tr>
<td>Br⁻, 12 mc</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pulse injection</td>
<td>230</td>
<td>1.69</td>
<td>1.69</td>
<td>1.32</td>
<td></td>
</tr>
<tr>
<td>Na⁺, 20 mc</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pulse injection</td>
<td>230</td>
<td>1.67</td>
<td>1.67</td>
<td>1.33</td>
<td></td>
</tr>
<tr>
<td>Na⁺, 20 mc</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pulse injection</td>
<td>230</td>
<td>1.71</td>
<td>1.71</td>
<td>1.32</td>
<td></td>
</tr>
<tr>
<td>Br⁻, 60 mc</td>
<td></td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>Pulse injection</td>
<td>230</td>
<td>1.70</td>
<td>1.70</td>
<td>1.32</td>
<td></td>
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<tr>
<td></td>
<td>550</td>
<td>1.71</td>
<td>1.70</td>
<td></td>
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<tr>
<td></td>
<td>1100</td>
<td>1.70</td>
<td>1.70</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>1650 (weir)</td>
<td>1.72</td>
<td>1.72</td>
<td></td>
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<td></td>
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</tr>
<tr>
<td>Distance (m)</td>
<td>Without &quot;carrier&quot;</td>
<td>With &quot;carrier&quot;</td>
<td>Comments</td>
<td></td>
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<td></td>
<td>From comparison of max. conc.</td>
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<td>From comparison of max. conc.</td>
<td>From comparison of mean conc.</td>
<td></td>
</tr>
<tr>
<td>56</td>
<td>-</td>
<td>-</td>
<td>94.9</td>
<td>-</td>
<td>Evidence of some delay of P(^{32}) without carrier</td>
</tr>
<tr>
<td>98</td>
<td>48.2</td>
<td>44.9</td>
<td>96.9</td>
<td>91.9</td>
<td>Without carrier P(^{32}) delayed relative to Na(^{24})</td>
</tr>
<tr>
<td>253</td>
<td>27.7</td>
<td>43</td>
<td>81.7</td>
<td>76.0</td>
<td>Without carrier P(^{32}) delayed relative to Na(^{24})</td>
</tr>
<tr>
<td>660</td>
<td>(\approx 9)</td>
<td>15.8</td>
<td>(\approx 63)</td>
<td>63.5</td>
<td></td>
</tr>
<tr>
<td>1460</td>
<td>0</td>
<td>0</td>
<td>-</td>
<td>-</td>
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</tr>
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</table>
complete lateral mixing was doubtful was in error by 18%. In all but one of the results the Dilution Method gave a higher reading than did the Continuous Sample Method; and this difference, which was just outside the estimated experimental error, was caused by sampling before equilibrium concentration was fully established.

This problem occurred consistently and resulted from the high degree of dispersion which was present in the three rivers. Typical concentration-
time curves, obtained on the upper Alwin from a rapid injection of Na\textsuperscript{24} are illustrated in Fig. 2. The corresponding rise to equilibrium concentration at 273 m from a continuous injection is shown in Fig. 3. The important advantage of the Continuous Sample over the Dilution Method is thus apparent. The "long tail" can easily be accommodated by the increase of the sampling period without introduction of a systematic error.

In the Dilution Method, particularly when the maximum injection period is limited, a large longitudinal dispersion can prevent the unequivocal establishment of equilibrium conditions. This is demonstrated in Fig. 4, which shows the limited distance over which a plateau occurred from a 30-min injection on the Aylburton Brook. In practice, equilibrium conditions can be proved by comparison of the concentration of several consecutive samples.

The present investigations used mechanical injectors to produce a constant injection rate for the Dilution technique over a period of 30 min. From the results obtained it is apparent that this time was too short and restricted the stretch of river over which a plateau could be established. An alternative method of injecting for longer periods is offered by a constant head device such as the Marriotte vessel [7], which is convenient to use and can be constructed to inject over several hours. It is accurate to within better than \(\frac{1}{2}\%\) and does not involve a prohibitive amount of radioactive handling during operation. Such an instrument was used to obtain the lateral dispersion results.

A comparison of 21 results with H\textsuperscript{3} in the Dilution and Continuous Sample Methods, with either Na\textsuperscript{24} or Br\textsuperscript{82} simultaneously injected, showed 9 out of the 21 H\textsuperscript{3} results deviated by less than 2.5\% from the corresponding results with gamma-emitting isotopes; and all but two of the remainder were in agreement to within 11\%.

Since the comparison between the techniques with gamma-emitting isotopes showed very much closer agreement than this and in view of diffic-
culties encountered with some of the H\textsuperscript{3} samples, which were known to have been slightly contaminated by tritiated water vapour diffusing into the polythene containers, the H\textsuperscript{3} results are now believed to be in error.

Five comparative results were obtained between the Total Count and Continuous Sample techniques with Na\textsuperscript{24} (see Tables II and IV). The Total Count Method indicated a lower flow by about 13\% in all the results. The self-consistency of the results suggests the discrepancy resulted from errors in calibration.

Flow measurements by the Dilution and Continuous Sample techniques with Na\textsuperscript{24} and Br\textsuperscript{82} were compared with the flow measured by a compound rectangular weir and showed the weir to be in error by 20±2\% between the
flow rates of 1.7 and 2.8 m$^3$/s (Table V). This result indicates the importance of periodic, on-site calibrations of permanent hydraulic structures.

6.2. Comparison of isotopes

$^{32}$P was included amongst the tracers selected to demonstrate errors likely to arise in the use of a tracer which is subject to adsorption. Significant losses occurred during transit of the tracer in the Aylburton Brook, as shown in Table VI. Without carrier, adsorption was immediately apparent and the $^{32}$P had disappeared completely by the 1460-m position. Na$^{24}$, injected simultaneously, travelled more rapidly, which is consistent with adsorption of the phosphorus. At 253 m the $^{32}$P peak concentration was 10 min behind the Na$^{24}$ after a transit time of 53 min. When carrier was added along with the radioactive tracer, significant adsorption did not occur until after the 100-m position.

Although adsorption is very marked with $^{32}$P, there is some evidence, as indicated in Table II, that it also occurs with Na$^{24}$ at the 660-m and the 1460-m positions on the Aylburton Brook, where the average transit times were 144 min and 320 min, respectively. There is no similar evidence that adsorption of Br$^{79}$ occurred at the 1650-m position on the Usway Burn (Table V), where the tracer transit time was only 30 min; but this may also result from the very different nature of the river bed.

A number of other isotopes including I$^{131}$, Cr$^{51}$ EDTA [8] have been used for flow investigations, and it is apparent that choice of a suitable tracer must be governed by the amount of adsorption which takes place within the sampling distance. Au$^{198}$ has been used in some investigations [3] but is not preferred by the present authors since it is unstable in dilute solutions and is adsorbed onto mineral surfaces [9].

6.3. Dispersion

The extent of lateral dispersion and the approach to equilibrium concentration from continuous injections at the centre and edge of the upper Alwin are shown in Fig. 5. The central injection is characterized by an immediate deviation of the tracer towards the right bank followed by a return towards the centre farther downstream. The pattern of tracer concentration following edge injection indicates a gradual approach to equilibrium concentration, but in neither of the two is equilibrium concentration reached at a distance of 140 m.

At the 96-m sampling point, lateral mixing was incomplete during the dispersion test. Close to one bank the concentration was approximately 3% of the ultimate equilibrium level and almost 170% at the opposite bank. It would be expected that this variation would have been observed in the flow measurements, but in fact this was not so although there is some indication of incomplete mixing during one test at this distance on the lower Alwin (Table III).

It is interesting to compare the present observations of lateral dispersion with the mixing distance estimated by HULL [3] and RIMMAR [4]. Hull’s formula predicts complete mixing at 46 m for a centre point injection,
whilst Rimmar's equation gives 750 m as the mixing length using a Chezy coefficient of 25 derived from data given by CHOW [10].

The self-consistency of the present results appear to indicate that complete mixing had been established before 250 m and that consequently Rimmar's equation overestimates the distance required for mixing in the conditions of the present test. However, since it is essential that counts should be made only after complete mixing has been established, the use of Hull's equation is inadvisable.

The variation of longitudinal dispersion with distance on the upper Alwin is illustrated in Fig. 2 by the three curves obtained at distances of 100, 273 and 525 m. The magnitude of the dispersion greatly exceeds the simple square-root relationship predicted by TAYLOR [11] and known to be very nearly true for flow in closed conduits. The excessive dispersion which was observed in the present tests is a further indication of the effect of the rocky bed and the dead-water zones which were present.

7. CONCLUSIONS

River flow measurements with the Dilution and Continuous Sample techniques using γ-emitting tracers were reproducible and differed systematically by 3.1%. The operational convenience and flexibility of the Continuous Sample technique makes it the method of choice.

Limited experience with the Total Count Method does not indicate that it has any advantages over the Continuous Sample Method, and it is much less convenient to operate.

Na\textsuperscript{24} and Br\textsuperscript{82} can be used reliably as tracers, but experience with P\textsuperscript{32} demonstrates the errors which may arise from adsorption.

H\textsuperscript{3} is very convenient to use in the field and is not susceptible to errors from adsorption. The difficulties of measuring H\textsuperscript{3} accurately are still formidable, and the isotope should only be used when γ-emitting isotopes are not convenient.

ACKNOWLEDGEMENTS

We wish to record our gratitude to the Northumberland River Board, the Coquet Water Board, the Severn River Board and the Lydney Park Estate Co. for their co-operation and for permission to carry out the present experiments.

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A large number of our own colleagues participated in these investigations, both in the field and in the laboratory, and it is a pleasure to mention the assistance of M.A. Aston, M. Bidgway, I.S. Boyce, G. Bradburn,
J.F. Cameron, W.E. Clark, T.V. Parsons, D.S. Rawson, R. Spackman, Miss A. Nunley and M.J. Joneidi of Tehran University, who was on sabbatical attachment and was one of the field team at the Aylburton Brook.

REFERENCES


DISCUSSION

See the discussion following the paper "Use of radioisotopes for open-channel flow measurements", these Proceedings.
APPAREIL POUR LA MESURE DES VITESSES RELATIVES DES EAUX SOUTERRAINES PAR LA MÉTHODE DE DILUTION PONCTUELLE

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Abstract — Résumé — Аннотация — Resumen

APPARATUS FOR MEASURING THE RELATIVE VELOCITIES OF GROUND WATERS BY THE POINT DILUTION METHOD. Determination of the relative velocities of ground water provides data on the relative porosity of terrains and is therefore of particular interest in problems concerned with the impermeability of soils.

It was with this in mind that the authors undertook the development of an apparatus, described in the paper, to determine the relative velocities of ground water.

The apparatus, which is intended for insertion in a piezometer, enables the water of one section of the piezometer to be isolated. By means of a device controlled from the surface a radioactive (gamma-emitting) tracer can be injected into this section, and an agitator ensures that the solution can be homogenized at any time. The radioactive tracer is scanned by a gamma-radiation detector located at one end of the measurement section. A suitable collimation device prevents this detector from being affected by the tracer located outside the piezometer.

The paper discusses the first results obtained with this apparatus.
устройство для коллимирования предохраняет этот детектор от излучения индикатора, находящегося вне пьезометра.

Дается критический обзор первых результатов, полученных с помощью этого прибора.

APARATO PARA MEDIR LAS VELOCIDADES RELATIVAS DE AGUAS SUBTERRÁNEAS POR EL MÉTODO DE DILUCIÓN PUNTIFORME. La determinación de las velocidades relativas de las aguas subterráneas permite obtener datos sobre la porosidad de los terrenos y reviste, por tanto, un interés primordial para los problemas de estanqueidad de los suelos.

Los autores describen un aparato que han construido con miras a determinar las velocidades relativas de las aguas subterráneas.

El aparato, que se introduce en un piezómetro, permite aislar el agua de una de sus secciones, que contiene un dispositivo, accionado desde la superficie del suelo, con el que se puede inyectar un radionúclido emisor y. Existe también un sistema de agitación para homogeneizar en todo momento la solución.

El paso del indicador radiactivo es observado mediante un detector de rayos y situado en uno de los extremos de la sección de mediciones. Un colimador adecuado impide que el detector registre la actividad procedente del exterior del piezómetro.

La memoria analiza los primeros resultados obtenidos con este aparato.

1. GÉNÉRALITÉS

Nous avons depuis quelques mois entrepris une étude dont l'objet est d'évaluer les possibilités de la très intéressante méthode de mesure de vitesses apparentes des eaux souterraines proposée par MOSER et ses collaborateurs [1]. Cette méthode qui consiste à suivre la disparition d'un traceur dans un tube piézométrique crevé avait déjà fait l'objet d'un certain nombre de travaux avec des traceurs classiques. Moser proposa l'utilisation de traceurs radioactifs. Ces derniers présentent l'avantage de pouvoir être mesurés in situ, et à de très faibles concentrations. La seule méthode en compétition semble être celle qui fait appel à une mesure de conductivité d'une solution saline, en général de NaCl.

Par vitesse apparente il faut entendre la vitesse que l'on peut déduire de la loi de Darcy, ou d'une manière plus générale, la vitesse $v$ qui détermine le débit d'eau à travers une section $S$ de terrain:

$$Q = Sv,$$

$v$ est lié à la vitesse réelle effective $v_e$ par:

$$v = v_e p,$$

$p$ étant la porosité efficace.

La connaissance des vitesses apparentes intéresse les problèmes d'alimentation de nappes aquifères et un bon nombre de problèmes d'étanchement de sols. Elles permettent de calculer par exemple les perméabilités, connaissant la pente hydraulique.

2. RAPPEL DU PRINCIPE DE LA MÉTHODE. ÉQUATION FONDAMENTALE

Nous supposons que l'on ait isolé une section d'un tube piézométrique crevé et que l'injection d'un traceur radioactif ait été réalisée dans cette section.
2.1. Hypothèses et conditions d’application de la méthode

Nous supposons: a) qu’un régime permanent d’écoulement est établi, 
b) qu’il n’existe pas de gradient de potentiel vertical (hypothèse de Dupuit), 
c) qu’à tout instant la concentration du traceur est homogène dans la section de mesure, d) que le volume sensible de la sonde de détection n’intéresse que la section de mesure.

2.2. Equation fondamentale

Une analyse simple du phénomène montre que l’on peut décrire la variation de concentration du traceur dans la section de mesure par:

\[ \frac{C}{C_0} = \exp \left( -\frac{v \sigma t}{V_0} \right) \]

où:

- \( C \): concentration du traceur à la date \( t \),  
- \( C_0 \): concentration du traceur à la date \( t = 0 \),  
- \( v \): vitesse apparente,  
- \( V_0 \): volume de la section de mesure,  
- \( \sigma \): section efficace de la section de mesure. Cette section permet de déterminer le volume d’eau passant à travers la zone de mesure dans un temps \( dt \): \( v \sigma dt \).

Si l’ensemble de détection a une réponse linéaire, l’enregistrement de la fréquence de comptage permet de calculer \( v \) connaissant \( V_0 \) et \( \sigma \). Si \( d \) est le diamètre du tube et \( h \) la hauteur de la section de mesure et si \( \epsilon \) est défini par \( \sigma = \epsilon dh \), on obtient:

\[ \frac{C}{C_0} = \exp \left( -\frac{4 \epsilon vt}{\pi d} \right) \]

3. SECTION EFFICACE DU VOLUME DE MESURE

La détermination de \( \epsilon \) représente le point essentiel dans l’étude de cette méthode. Notons à cet égard que KAUFMAN et MOSER donnent respectivement les valeurs 2 et 1 pour ce paramètre [2].

L’analyse permet de démontrer que dans le cas d’un piézomètre parfait (sans tubage ou dont la perméabilité du tubage est égale à celle du terrain), la largeur de la section efficace est égale à deux fois le diamètre du trou, sans que la vitesse, la perméabilité ou le gradient (\( \epsilon = 2 \)) de l’écoulement n’interviennent (fig. 1).

En réalité ces facteurs, perméabilité et gradient tout au moins, interviennent lorsque l’on introduit un crépinage. Nous avons, dans le but de dégager les lois qui régissent ces phénomènes, entrepris une étude expérimentale en laboratoire. Nous comptons la poursuivre à l’aide d’une méthode d’analogie électrique qui permettra de réduire notablement la durée de cette étude. On obtient directement par cette méthode un tracé, sur un papier conducteur, des courants marginaux qui définissent la valeur de \( \epsilon \).
Nos premiers résultats ainsi obtenus (fig. 2) montrent que le crépinage a une très grande importance, et qu'il doit pouvoir réduire la largeur de la nappe intéressée nettement en dessous du diamètre \( d \) du piézomètre \((\varepsilon < 1)\).

4. DESCRIPTION DE L'APPAREIL RÉALISÉ AU CEA

4.1. Détecteur

Le choix du mode de détection est un point important dans la conception de l'appareil de mesure; il importe que cet appareil ait un régime linéaire et surtout qu'il soit insensible à l'activité hors de la section.

Les émetteurs de rayonnement \( \beta \), dont la portée dans l'eau est de quelques millimètres pourraient apporter une solution intéressante. Cependant, dans un de nos essais réalisés avec du \( ^{32} \text{P} \) et un compteur G-M à parois minces, l'adsorption du traceur sur le verre rendait impossible l'exploitation du signal.
Etant donné par ailleurs que le volume dont on dispose est faible, nous avons donc préféré utiliser un détecteur à scintillation collimaté et placé hors de la section (fig. 3). Cette disposition qui a l'inconvénient de diminuer assez notablement le rendement de détection nous paraît cependant préférable à la solution qui consiste à placer le détecteur au centre de la section de mesure.

Dans ce dernier cas, en effet, le détecteur «voit» l'activité à l'extérieur du tube. Dans l'hypothèse où le traceur n'est pas adsorbé sur le tube ou dans le terrain ceci n'a pour conséquence que d'augmenter le temps de mesure. Celle-ci ne peut commencer que lorsque le front d'activité a atteint les limites du volume sensible. Dès lors l'activité en tous les points du volume sensible du détecteur, hors du tube piézométrique, décroît selon la loi:

\[
\frac{C}{C_0} = \exp \left[ -\frac{\varepsilon v}{\pi d} \left( t - \frac{1}{v_e} \right) \right]
\]

où:
\[
\varepsilon_e = \text{vitesse réelle effective},
\]
\[
t = \text{distance curviligne selon les lignes de courant du point considéré}
\]
aux tube piézométrique.
On pourra peut-être montrer que l'on peut tenir compte de l'adsorption en assimilant son effet à un bruit de fond. Il nous paraît cependant plus sage pour une étude dans laquelle interviennent de nombreux autres paramètres d'adopter un détecteur collimaté hors de la section de mesure.

4.2. Système d'agitation

L'homogénéisation du traceur est réalisée par un dispositif comprenant deux hélices entraînées par un moteur. Les hélices sont placées dans un tube afin d'éviter au maximum les composantes horizontales des vitesses, et de ne pas perturber l'écoulement naturel.

Les lignes de courant dues à l'agitation sont ainsi en majeure partie parallèles à l'axe du tube. Des essais avec un traceur coloré ont montré que les zones les plus efficaces de mélange se situent au voisinage des hélices.
4.3. Détermination de la section de mesure

Il importe que la section de mesure soit délimitée dans le sens vertical afin d'éviter la diffusion du traceur dans cette direction. Nous utilisons à cet effet deux chambres toriques en caoutchouc que l'on gonfle depuis la surface.

4.4. Injection du traceur

Dans la partie inférieure de l'appareil se trouve le réservoir de la solution active. Ce réservoir est sous pression d'air, ce qui permet de réaliser les injections en commandant une électrovanne depuis la surface.

On peut ainsi ajuster, à l'aide d'un bouton poussoir, l'activité injectée au niveau désiré par simple lecture de l'enregistrement. Ce dispositif évite par ailleurs de sortir l'appareil à chaque mesure. Ces mesures sont faites de bas en haut dans le tube piézométrique; il n'y a ainsi jamais d'eau active au voisinage de la sonde de détection.

Notons enfin que la distance et la protection est telle entre le détecteur et le réservoir d'activité que ce dernier n'influence pas pour le bruit de fond.

5. ÉTUDE EXPÉRIMENTALE

5.1. Cuve expérimentale et tube crépiné

Nous utilisons une cuve de section 55 X 86 cm et 1,6 m de long séparée en 5 compartiments délimités par des grilles perforées. Les deux compartiments aux extrémités sont remplis d'eau, et comportent les rampes d'aménée et de sortie d'eau. Les 2 compartiments adjacents remplis de sable servent à homogénéiser les vitesses; le compartiment central de 50 cm de longueur contient le terrain en expérience. L'alimentation est faite par une cuve à niveau constant, et l'eau est siphonnée à travers un orifice calibré en aval.

Le tube crépiné a un diamètre intérieur de 123 mm et une épaisseur de 4 mm. Il comporte 8 fentes verticales de 3 mm de largeur et 100 mm de hauteur. Ces fentes sont imbriquées sans recouvrement.

Dans nos expériences au cours desquelles plusieurs dizaines d'injections ont été réalisées sur le même sol (graviers de rivières) cette cuve s'est fortement contaminée avec de l'iode-131.

Cette contamination est un phénomène gênant dans ce type d'étude; nous voyons ici encore l'intérêt d'utiliser un détecteur collimaté.

5.2. Caractéristiques propres à l'appareil

a) Système d'agitation

Nous avons effectué un bon nombre d'expériences pour déterminer le système d'agitation de l'appareil représenté à la figure 2.

Les courbes de la figure 4 nous montrent par exemple l'hétérogénéité de la solution dans le tube sans système d'agitation. Elles ont été obtenues
à l'aide de 3 compteurs G-M, disposés sur un diamètre du tube, dans la direction du courant.

Avec le détecteur à scintillation collimaté de notre appareil, on obtient sans agitation (et sans le tube central contenant les hélices) des courbes analogues à celles que nous représentons en (a) à la figure 5. Ces courbes qui représentent en diagramme semi-logarithmique les variations du signal de la sonde en fonction du temps ne sont pas exponentielles. La courbe de la figure 5 (b) obtenue avec le système d'agitation est parfaitement exponentielle après une courte période de temps correspondant à la phase d'homogénéisation.

Remarque 1

Une légère contamination de l'appareil entraîne une augmentation du bruit de fond. Cette contamination est mise en évidence lorsque la courbe nette n'est plus exactement exponentielle pour les faibles fréquences de comptage.

Remarque 2

Nous verrons que la diffusion du traceur hors du tube est équivalente à une vitesse d'écoulement. Nous avons mis en évidence que cette vitesse était identique avec et sans agitation.
b) Détermination du collimateur de détection

Le cristal scintillant de NaI(Tl) a pour dimensions 1" X 1" ; le collimateur en plomb a une hauteur de 40 mm et un diamètre de 20 mm. Ces dernières dimensions ont été déterminées en étudiant les variations d'un rapport (signal + bruit)/bruit à l'aide d'une cuve spéciale annulaire. La sonde occupait la partie centrale. Dans la partie annulaire, emplie de sable, était ménagé un secteur de 90° comprenant la solution active et qui simulait l'activité hors du tube piézométrique. A cette activité correspondait une fréquence de comptage appelée bruit. A une activité de même concentration spécifique et placée à l'intérieur du tube piézométrique correspondait une fréquence appelée signal.

5.3. Etude préliminaire de la méthode

Une soixantaine d'essais ont été réalisés au laboratoire avec trois sortes de sables que nous désignons par $g_1$, $g_2$, $g_3$.  

Figure 5

Courbes montrant l'effet de la diffusion.
g_1: sable concassé, grains plats de 4 à 8 mm ×1 mm,
g_2: sable de rivière lavé, de 1 à 5 mm,
g_3: sable de rivière lavé, de 0,2 à 0,8 mm.

Pour ces différents types de sable nous avons obtenu des lois linéaires entre les vitesses vraies et les vitesses mesurées avec l'appareil.

Avec le sable g_1 nous utilisons un tube en matière plastique dont les fentes avaient une largeur de 2 mm. Nous avons obtenu dans la gamme de 0,8 - 6,3 m/j la valeur ε = 0,8.

Avec les sables g_2 et g_3 nous avons utilisé le tube métallique défini précédemment. La gamme de vitesse était de 0,4 à 8 m/j.

La figure 6 représente les résultats de deux séries de mesures. ε a pour valeur 1, et toutes les mesures sont à moins de 10% de la moyenne.

Dans tous ces essais la linéarité entre la vitesse vraie et mesurée est remarquable.
CONCLUSION

Nous avons souligné un certain nombre de points importants dont l'étude devra être parachevée pour donner à la méthode de dilution ponctuelle toute sa valeur.

Nous avons décrit un appareil qui nous paraît très adapté pour cette étude. Nous nous proposons de la poursuivre par des mesures électriques analogiques qui permettront sans doute de dégager un certain nombre de lois dont la vérification se fera en premier lieu par des expériences de laboratoires puis sur des terrains dont les perméabilités auront été mesurées par une méthode hydraulique classique.

RÉFÉRENCES


DISCUSSION

See the discussion following the paper "Use of radioisotopes for open-channel flow measurements", these Proceedings.
USE OF RADIOISOTOPES FOR OPEN-CHANNEL FLOW MEASUREMENTS

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Abstract — Résumé — Аннотация — Resumen

USE OF RADIOISOTOPES FOR OPEN-CHANNEL FLOW MEASUREMENTS. With techniques based upon the salt-dilution method, radioisotopes can be used for discharge measurements in open channels such as canals, streams and rivers. Field measurements with radioisotopes in canals discharging up to 8000 ft³/s have been made by the Bureau of Reclamation. The conditions of the field measurements are described in this paper, and important observations are made relating the results to the use of these methods for discharge measurements in rivers.

The field tests were performed to study the general feasibility of the use of isotopes for discharge measurements in open channels and to investigate some of the important field problems which are still under study. These include the field requirements for necessary transverse mixing of the isotopes with the flowing water; sorption of the isotopes by suspended sediments, channel flow surfaces and aquatic plants; and improvement of field procedures and equipment. Results of present laboratory and field tests lead to a proposed method for employing radioisotopes in river-discharge measurement to provide continuous or periodic discharge determinations.

EMPLOI DES RADIOISOTOPES POUR LA MESURE DU DÉBIT D’EAUX S’ÉCOULANT À CIEL OUVERT.

Avec les méthodes fondées sur la dilution des sels, on peut utiliser les radioisotopes pour la mesure du débit d’eaux s’écoulant à ciel ouvert: canaux, rivières et autres cours d’eau. Le «Bureau de Réclamation» a fait des mesures à l’aide de radioisotopes dans des canaux débitant jusqu’à 230 m³/s. Les auteurs exposent les conditions dans lesquelles se sont déroulées ces mesures et font des observations importantes concernant l’interprétation des résultats dans le cas où l’on voudrait utiliser ces méthodes pour des mesures du débit dans les rivières.

Les expériences avaient pour but de déterminer s’il était possible d’employer des radioisotopes pour la mesure du débit d’eaux s’écoulant à ciel ouvert, et d’élucider certains des importants problèmes pratiques qui sont encore à l’étude, notamment: les conditions qui assurent le brassage transversal indispensable pour que les isotopes se mélangent à l’eau courante; la sorption des isotopes par les sédiments en suspension, les parois du lit et les plantes aquatiques; l’amélioration des méthodes pratiques et le perfectionnement du matériel. Les résultats des expériences qu’ils ont faites en laboratoires et sur le terrain ont conduit les auteurs à mettre au point une méthode d’emploi des radioisotopes pour les mesures, continues ou périodiques, du débit des cours d’eau.

ИСПОЛЬЗОВАНИЕ РАДИОИЗОТОПОВ ДЛЯ ИЗМЕРЕНИЯ РАСХОДА ВОДЫ В ОТКРЫТЫХ РУСЛАХ.

С помощью технических приемов, основанных на методе растворения соли, можно использовать радиоизотопы для измерения расхода воды в открытых руслах - каналах, ручьях и реках. Управление мелиорации были проведены полевые измерения с использованием радиоизотопов в каналах, пропускающих до 230 куб.м.тр. вод в 1 сек. Дается описание условий полевых измерений и сообщаются важные наблюдения, касающиеся результатов использования этих методов при измерении расхода воды в реках.

Полевые испытания были проведены с целью изучения общей возможности использования изотопов для измерения расхода воды в открытых руслах и для исследования некоторых наиболее важных полевых проблем, которые изучаются в настоящее время. К ним относятся: поперечное смешивание изотопов с потоком воды; поглощение изотопов суспензиями дном и берегами каналов, а также водными растениями; улучшение техники полевых опытов и оборудования. Результаты настоящих лабораторных и полевых испытаний приведены к предложенному методу использования радиоизотопов при измерении расхода воды в реках. Метод обеспечивает постоянные или периодические определения расхода воды.
EMPLEO DE RADIOISÓTOPOS PARA MEDIR LA CIRCULACIÓN EN CANALES ABIERTOS. Cuando se trabaja con técnicas basadas en el método de dilución salina, los radioisótopos permiten medir el caudal en canales abiertos, corrientes y ríos. El Bureau of Reclamation los ha utilizado para determinar en canales un caudal de hasta 8000 pies³/s. La memoria describe las condiciones en que se realizaron las mediciones sobre el terreno y, basándose en los resultados obtenidos, formula observaciones relativas al empleo de estos métodos en la medición de caudales fluviales.

Los ensayos sobre el terreno se realizaron con miras a estudiar la posibilidad de emplear isótopos para medir el caudal en canales abiertos y para investigar algunos de los problemas más importantes que se plantean en la práctica y que se están estudiando todavía. Se trata, entre otros, de las condiciones que en la práctica deben cumplirse para que se produzca la necesaria mezcla transversal de los isótopos con el agua en circulación, de la sorción de los isótopos por sedimentos en suspensión, por las superficies laterales y del fondo del canal y por las plantas acuáticas; también es importante mejorar los procedimientos y el equipo que se emplean sobre el terreno. Los resultados obtenidos inducen a los autores a proponer un método que utiliza radioisótopos para medir el caudal de ríos en forma continua o periódica.

BASIC CONCEPTS

Chemicals were first used in large-scale flow measurements approximately 100 years ago, and their importance has increased considerably since that time. The use of radioisotopes as a special form of chemical measurement was first introduced by Joly in 1922, but only in recent years have large-scale measurements, involving both open and closed conduits, been performed [4]. Improved instrumentation and the need for more accurate discharge measurements have been responsible for this renewed and increased interest.

The Bureau of Reclamation is engaged in a research and development programme to investigate the use of radioisotopes for discharge of flow measurements. The programme is broad in scope and covers the study of the basic concepts of several methods of using isotopes; refinement of isotope injection and counting equipment; development of techniques suitable for measurements of flow in natural waterways, canals and through pumps and turbines; and identification and solution of problems arising from field use of the method. The subject of this paper is the use of radioisotopes in discharge measurements in large canals.

Salt dilution using radioisotopes

In one method of using radioisotopes the techniques are directly related to the salt dilution method. In the dilution method a salt solution of known concentration \( C_1 \) is introduced into the flow at a constant rate \( q \). At a point sufficiently distant downstream to assure adequate transverse mixing of the salt solution with the flow the measured salt concentration is \( C_2 \). From considerations of conservation of material, \( C_1 q = C_2 Q \), where \( Q \) is the flow being measured; hence \( Q = (C_1/C_2)q \n
Isotope dilution

Radioactive salt in solution can safely be substituted for ordinary chemical salts to achieve equal or better accuracy. With radioisotopes smaller volumes of the chemical tracer need to be handled and injected, and measurement of the downstream concentration is a simpler field pro-
procedure. These and related advantages come from the high degree of detectability of radioactive materials by their radioactive properties.

The relationship between the use of non-radioactive and radioactive chemicals in the salt dilution method will be discussed without too detailed attention to the instrumentation or laboratory and field procedures, which can vary depending upon the requirements of the particular test. It will be shown that other related methods can be derived from the basic concepts of salt dilution.

Consider the concentration $C_1$ and $C_2$ in the equation for the salt dilution method. When radioactive material is used, these concentrations can be determined by Geiger or scintillation counters which measure the gamma-ray emissions from the radioactive salts introduced into the flow. The emissions sensed by these instruments are counted by a portable battery-operated scaler.

The concentration $C_1$ and $C_2$ can be determined in terms of $\mu c/ml$ by proper calibration of the counting equipment. This can be done in the laboratory. For example, the counting rate is determined for a Geiger counter probe submerged in a container filled with a radioactive solution of known concentration. The container is large enough so that gamma rays originating from the outer regions of the container are not counted by the probe in the centre of the volume. This simulates the action of the probe in the field since gamma rays also occur outside the range of the probe. If $F$ is the calibration factor for a given set of equipment or a given probe, then the counting rate $R$ for a solution of concentration $C$ is $R = FC$, or $C = (R/F)$.

In the field, calibrated equipment is used for the determination of the values of $C_1$ and $C_2$ in the salt dilution method equation. This gives

$$Q = \frac{C_1}{C_2} \times q = \frac{R_1/F}{R_2/F} \times q$$

or $Q = (R_1/R_2) \times q$.

Thus, it is seen that radioisotopes may be substituted for chemical salt in the salt dilution method of measuring discharge. When radioisotopes are used, the technique is known as the isotope dilution method. It is also to be noted from the above equation that the discharge can be determined without a knowledge of the exact value of the calibration factor. The calibration factor need be only sufficiently accurate for the determination of the quantity of radioisotopes necessary to give counting rates large enough for accurate determination of the discharge.

The field procedure in the isotope dilution method is the determination of the concentration $C_1$ of the isotope being introduced into the flow. It is introduced at a constant rate $q$ by a Mariotti flask or constant-flow pump. At a sufficient distance downstream from the point of introduction of the isotope to assure adequate mixing, the concentration of the stream $C_2$ is determined by immersion of a counter, such as a Geiger counter, into the stream. The counting rates obtained for the original solution $C_1$ and the diluted solution $C_2$ provide the data necessary for determination of the discharge for the flow $Q$. 
Pulse or total count method

From the basic considerations of the salt dilution method another technique of using radioisotopes which eliminates the need for continuous application of the tracer at a fixed rate \( q \) can be derived. In the total count or pulse method a known amount of tracer \( A \) is introduced to the flow in a comparatively short time, producing a pulse of radioactivity in the flowing water.

At the measurement point downstream the concentration of the tracer is determined as previously discussed for the isotope dilution method. However, the constant \( C_2 \) in the salt dilution equation is now a variable and a function of time. Again, from considerations of conservation of matter,

\[
A = C_1q = Q/C_2dt
\]

or

\[
Q = C_2q/\int C_2dt.
\]

The latter equation is similar to the equation for the isotope dilution method.

Since \( A \) is known, the rate of introduction \( q \) and the initial concentration \( C_1 \) need not be determined. Since \( C_2 = R/F \),

\[
Q = C_1q/\int (R/F)dt = C_1q/(1/F)\int Rdt.
\]

The total number of counts \( N \) obtained during the passage of the tracer is \( N = \int Rdt \). By substitution, the quantity of flow (discharge) is then found to be \( Q = FA/N \).

FIELD STUDIES

Field measurements in canals were made with the use of the pulse or total count method and the general procedures described above. In addition, specific procedures were as described below.

The quantity \( A \), or the amount of radioisotope to be introduced for each measurement, was determined by measurement of the radioactivity of the solution in a portable standardized counting system which was calibrated in the laboratory. The radioisotope gold-198 was then introduced into the canal stream in a solution of gold chloride poured from a plastic bottle onto the surface of the canal water.

The total number of counts \( N \) was determined by means of a probe consisting of four Geiger counters encased in a plastic tube and submerged in the canal water. The "counting rate" was determined with a companion portable, battery-operated scaler.

Several series of discharge measurements were performed at different times in various canals carrying flows from a few to over 200 m³/s (few hundred to over several thousand cubic feet per second).

Demonstration tests

The first series of measurements was made in April 1961 as a demonstration for exploration of the use of radioisotopes for canal flow
measurements. These tests were carried out co-operatively with United States Geological Survey personnel, who made precise, independent measurements of discharge using the current-meter traverse method.

The April 1961 demonstrations were performed on the Gila Gravity Main Canal and its branches, the Yuma-Mesa Branch and the Wellton-Mohawk Canal. At the point of bifurcation the Gila Gravity Main Canal discharge was 34 m$^3$/s (1200 ft$^3$/s); velocity, 0.70 m/s (2.3 ft/s). The flow division was Wellton-Mohawk discharge 22.7 m$^3$/s (800 ft$^3$/s); velocity, 0.09 m/s (0.3 ft/s); and Yuma-Mesa Branch discharge, 11.4 m$^3$/s (400 ft$^3$/s); velocity, 0.76 m/s (2.5 ft/s).

The locations of the measurements are briefly described in Table I, and Table II describes demonstration tests. It is to be noted that some of the measurements were made at, or across, engineering structures for a study of the influence of the structure or for use of the turbulence introduced by the structure for mixing.

The current-meter flow measurements were made with special care for the best possible evaluation of the radioisotopes measurements. A comparison of the results and an error analysis are given in Table III. The probable error in the current-meter measurements, estimated by the experienced technicians using the current meters, has been used to indicate the outside limit of accuracy of the radioisotope flow measurements achieved. This does not represent a statistical error analysis either on the basis of the results or the total number of counts $N$. A statistical evaluation on the basis of the total number of counts, $100/(N)^{1/2}$, is also included and found to be almost always less than the limits of error from the current-meter measurements.

Additional field measurements

Because of the encouraging results from the demonstrations at Yuma in April 1961, additional measurements were made, with essentially the same techniques, at other locations. These subsequent measurements are summarized in Table IV.

In general, these subsequent tests confirm the encouraging nature of the earlier demonstrations performed on the Gila Gravity Main Canal. The tests were usually performed without great difficulty, effort and preparation; and most tests gave reasonably consistent results one with another and with operational determinations of the discharge.

However, many procedures and details remain to be explored before the full utility of the technique is achieved and minimum requirements for successful discharge determinations are established. Some of the additional measurements were designed to bring out some of the field problems. In other cases unforeseen problems arose.

Transverse mixing

For example, one of the most critical factors is the determination of the necessary length of canal for achievement of thorough transverse mixing. Unless adequate transverse mixing is achieved, inaccurate discharge values
TABLE 1
DESCRIPTION OF TEST LOCATIONS FOR APRIL 1961
DEMONSTRATIONS, YUMA, ARIZONA

<table>
<thead>
<tr>
<th>Test No.</th>
<th>Canal</th>
<th>Canal description</th>
<th>Engineering structures</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Gila Gravity</td>
<td>Trapezoidal, earth-lined, approximate base = 6.7 m (22 ft), depth = 4.1 m (13.54 ft), side slopes = 2:1</td>
<td>Injection at sluice gate discharge to canal, flow very turbulent; counting at Station 30</td>
</tr>
<tr>
<td>2</td>
<td>Gila Gravity</td>
<td>Trapezoidal, earth-lined, approximate base = 6.7 m (22 ft), depth = 4.1 m (13.54 ft), side slopes = 2:1</td>
<td>Injection near entrance to Tunnel 2, 6.10 m (20 ft) diameter and 880 m (2900 ft) long. Counting at tunnel exit</td>
</tr>
<tr>
<td>3A</td>
<td>Gila Gravity</td>
<td>Trapezoidal, earth-lined, approximate base = 6.7 m (22 ft), depth = 4.1 m (13.54 ft), side slopes = 2:1</td>
<td>Injection at upstream end of Gila River Siphon, 5.94 m (19.5 ft) diameter, 610 m (2000 ft) long. Counting at siphon exit</td>
</tr>
<tr>
<td>3D</td>
<td>Wellton-Mohawk</td>
<td>Trapezoidal, earth-lined, base = 13.41 m (44 ft), depth = 2.68 m (8.8 ft), side slopes = 2:1</td>
<td>Injection above wash siphon approximate 4.0 m (13 ft) diameter 60 m (200 ft) long. Counting downstream 1372 m (4500 ft)</td>
</tr>
<tr>
<td>4</td>
<td>Yuma-Mesa Branch (&quot;A&quot; Canal)</td>
<td>Trapezoidal, concrete-lined, base = 2.44 m (8 ft), depth = 3.07 m (10.08 ft), side slopes = 1.5:1. Straight section</td>
<td>Injection 304 m (1000 ft) upstream from pumping plant. Counting 320 m (1050 ft) downstream from pumps</td>
</tr>
<tr>
<td>5</td>
<td>Wellton-Mohawk</td>
<td>Trapezoidal, concrete-lined at injection base 4.88 m (16 ft), depth = 3.9 m (12.8 ft), side slopes = 1.25:1</td>
<td></td>
</tr>
</tbody>
</table>
### Table II

**Demonstration Tests of Flow Measurements with Radioisotopes**

Discharge measurements at various locations of the Gila Gravity Main Canal and Yuma Mesa Canal were made with radioisotopes. The results are given in metric units with the English equivalents in parentheses.

<table>
<thead>
<tr>
<th>Test No.</th>
<th>Location</th>
<th>Location Details</th>
<th>$^{198}$Au (mc)</th>
<th>Injection to count distance (km (mi))</th>
<th>Duration of count (h : min)</th>
<th>Total time (h : min)</th>
<th>$N_{C/S}$ net</th>
<th>$Q$ ($m^3/s$ (ft$^3$/s))</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Station 30</td>
<td></td>
<td>166</td>
<td>0.56 (0.35)</td>
<td>0:12</td>
<td>0:22</td>
<td>2710</td>
<td>39.45 (1393)</td>
</tr>
<tr>
<td>2</td>
<td>Tunnel 2</td>
<td></td>
<td>406</td>
<td>1.37 (0.85)</td>
<td>1:45</td>
<td>2:08</td>
<td>7041</td>
<td>37.21 (1314)</td>
</tr>
<tr>
<td>3A-1</td>
<td>Gila River Siphon</td>
<td></td>
<td>576</td>
<td>0.69 (0.43)</td>
<td>0:32</td>
<td>0:42</td>
<td>10874</td>
<td>34.86 (1231)</td>
</tr>
<tr>
<td>3A-2</td>
<td>Gila River Siphon</td>
<td></td>
<td>498</td>
<td>0.69 (0.43)</td>
<td>0:50</td>
<td>0:48</td>
<td>9436</td>
<td>34.13 (1205)</td>
</tr>
<tr>
<td>3D</td>
<td>Wellton-Mohawk</td>
<td></td>
<td>553</td>
<td>1.40 (0.87)</td>
<td>2:10</td>
<td>1:25</td>
<td>15961</td>
<td>22.37 (790)</td>
</tr>
<tr>
<td>4</td>
<td>Yuma-Mesa Pumping Plant (&quot;A&quot; Canal)</td>
<td></td>
<td>301</td>
<td>1.29 (0.80)</td>
<td>1:20</td>
<td>1:25</td>
<td>17374</td>
<td>11.19 (395)</td>
</tr>
<tr>
<td>5</td>
<td>Wellton-Mohawk Pumping Plant No. 1</td>
<td></td>
<td>395</td>
<td>0.77 (0.48)</td>
<td>1:49</td>
<td>1:43</td>
<td>11387</td>
<td>22.46 (793)</td>
</tr>
</tbody>
</table>
are indicated. The series of tests on the A canal, performed 7 and 8 February 1962, were designed to investigate this problem, both visually and with counting equipment. By injection of fluorescene dye into the canal, it was found that a gentle bend in the canal caused the dye to remain along one side of the canal for greater distances than the casual observation of the flow would indicate. To help evaluate the transverse distribution, measurements were made with separate counting probes located in the left, centre and right portions of the canal and at two locations downstream from the bend. The degree of mixing was evaluated by the relationship

\[
\% \text{ mixing} = \frac{1 - \left( |N_L - N_m| + |N_c - N_m| + |N_R - N_m| \right)}{3N_m} \times 100\%
\]

where \(N_L\) = total counts at the left counter, \(N_c\) = total counts at the centre counter, \(N_R\) = total counts at the right counter and \(N_m\) = means total counts for all three counters.

The results of the tests are shown in Table VI.

From the per cent mixing and the discharge measurements, it is seen that inadequate distribution of the radioisotopes occurred even for a distance

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### Table III

<table>
<thead>
<tr>
<th>Test No.</th>
<th>Current meter (Q) ((m^3/s (ft^3/s)))</th>
<th>Error from current meter ((%))</th>
<th>Radioisotopes (Q) ((m^3/s (ft^3/s)))</th>
<th>Error from radioactive isotope ((%))</th>
<th>Statistical error from (100/(N)^{1/2}) ((%))</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>39.53 (1396)</td>
<td>1</td>
<td>39.45 (1393)</td>
<td>-1</td>
<td>1.9</td>
</tr>
<tr>
<td>2</td>
<td>37.89 (1338)</td>
<td>3</td>
<td>37.21 (1314)</td>
<td>-5</td>
<td>1.2</td>
</tr>
<tr>
<td>3A-1</td>
<td>34.89 (1232)</td>
<td>3</td>
<td>34.86 (1231)</td>
<td>-3</td>
<td>0.97</td>
</tr>
<tr>
<td>3A-2</td>
<td>34.27 (1219)</td>
<td>3</td>
<td>34.13 (1205)</td>
<td>-3</td>
<td>1.0</td>
</tr>
<tr>
<td>3D</td>
<td>22.23 (785)</td>
<td>3</td>
<td>22.37 (790)</td>
<td>-2</td>
<td>0.79</td>
</tr>
<tr>
<td>4</td>
<td>11.70 (413)</td>
<td>3</td>
<td>11.19 (395)</td>
<td>-7</td>
<td>0.76</td>
</tr>
<tr>
<td>5</td>
<td>22.34 (789)</td>
<td>3</td>
<td>22.46 (493)</td>
<td>-2</td>
<td>0.94</td>
</tr>
</tbody>
</table>
## TABLE IV

### ADDITIONAL FIELD TESTS

**Description of location**

<table>
<thead>
<tr>
<th>Date</th>
<th>Location</th>
<th>Structure description</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>19/10/61</td>
<td>Gila River Siphon</td>
<td>Siphon: diameter, 5.94 m (19.5 ft); length, ~610 m (2000 ft)</td>
<td>Concrete-lined inverted siphon</td>
</tr>
<tr>
<td>19/10/61</td>
<td>Gila Gravity Main Canal, Station 5+64</td>
<td>Earth-lined canal: base, 6.7 m (22 ft); depth, 4.1 m (13.54 ft), side slope = 2:1</td>
<td>Maximum capacity 62.3 m³/s (2200 ft³/s)</td>
</tr>
<tr>
<td>19/10/61</td>
<td>Gila Gravity Main Canal, Tunnel 2</td>
<td>Tunnel: diameter, 6.10 m (20 ft); length, 880 m (2900 ft)</td>
<td>Concrete-lined</td>
</tr>
<tr>
<td>20/10/61</td>
<td>Gila Gravity Main Canal, Tunnel 2</td>
<td>Tunnel: diameter, 6.10 m (20 ft); length, 880 m (2900 ft)</td>
<td></td>
</tr>
<tr>
<td>20/10/61</td>
<td>Gila River Siphon</td>
<td>Tunnel: diameter, 6.10 m (20 ft); length, 880 m (2900 ft)</td>
<td></td>
</tr>
<tr>
<td>Date</td>
<td>Location</td>
<td>Structure description</td>
<td>Remarks</td>
</tr>
<tr>
<td>---------</td>
<td>-----------------------------------------------</td>
<td>-----------------------</td>
<td>---------------------------------------------------</td>
</tr>
<tr>
<td>6/2/62</td>
<td>Yuma Mesa &quot;A&quot; Canal, Station 5+00</td>
<td>Concrete-lined canal:</td>
<td>Gentile S-curve at upstream end of test area; maximum capacity, 17.56 m$^3$/s</td>
</tr>
<tr>
<td></td>
<td></td>
<td>trapezoidal base = 2.44 m (8 ft); depth = 3.07 m (10.08 ft); side slopes = 1.5:1</td>
<td></td>
</tr>
<tr>
<td>7/2/62</td>
<td>Yuma Mesa &quot;A&quot; Canal, Station 10+00</td>
<td>Concrete-lined canal:</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>trapezoidal base = 2.44 m (8 ft); depth = 3.07 m (10.08 ft); side slopes = 1.5:1</td>
<td></td>
</tr>
<tr>
<td>8/2/62</td>
<td>Yuma Mesa &quot;A&quot; Canal, Station 60+00</td>
<td>Concrete-lined canal:</td>
<td>Test of split flow:</td>
</tr>
<tr>
<td></td>
<td></td>
<td>trapezoidal base = 2.44 m (8 ft); depth = 3.07 m (10.08 ft); side slopes = 1.5:1</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1. Counter above junction with &quot;B&quot; Canal</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2. Counter below junction on &quot;A&quot; Canal</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>3. Counter below junction on &quot;B&quot; Canal</td>
</tr>
<tr>
<td>25/4/62</td>
<td>Gila River Siphon</td>
<td>Concrete-lined canal:</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>trapezoidal base = 2.44 m (8 ft); depth = 3.07 m (10.08 ft); side slopes = 1.5:1</td>
<td></td>
</tr>
<tr>
<td>25/4/62</td>
<td>Gila Gravity Main Canal, Station 5+64</td>
<td>Concrete-lined canal:</td>
<td></td>
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<td>trapezoidal base = 2.44 m (8 ft); depth = 3.07 m (10.08 ft); side slopes = 1.5:1</td>
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<td>Coachella Canal,</td>
<td>Earth-lined canal: depth = 3.15 m (60 ft); base = 18.28 m (10.33 ft); side slopes = 2:1</td>
<td>Maximum capacity = 70.80 m³/s (2500 ft³/s)</td>
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<td>All-American Canal,</td>
<td>Earth-lined canal: trapezoidal base = 48.77 m (160 ft); depth = 6.28 m (20.6 ft); side slopes = 1.74:1</td>
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<td>Consolidated Canal</td>
<td>Concrete-lined canal: trapezoidal base = 12.19 m (40 ft); depth = 1.52 m (5 ft); side slopes = 1.25:1</td>
<td>1920 m (6300 ft) above diversion gates; maximum capacity = 37.52 m³/s (1325 ft³/s)</td>
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<td>South Canal, Granite</td>
<td>Concrete-lined canal: trapezoidal base = 19.20 m (63 ft); depth = 2.44 m (8 ft); side slopes = 3/8:1</td>
<td>Incomplete mixing occurred; maximum capacity = 46.73 m³/s (1650 ft³/s)</td>
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<td>Consolidated Canal,</td>
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## TABLE V
### ADDITIONAL FIELD TESTS RESULTS

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<th>Net counts (N)</th>
<th>( Q ) radioisotope ((m^3/s (ft^3/s)))</th>
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<td>8.95</td>
<td></td>
</tr>
<tr>
<td>25/5/62</td>
<td>Consolidated Canal - Diversion Gates</td>
<td>305</td>
<td>448</td>
<td>23 878</td>
<td>8.86</td>
<td></td>
</tr>
<tr>
<td>Test No</td>
<td>Counter location</td>
<td>Mixing distance (m (ft))</td>
<td>Q operational (m³/s) (ft³/s)</td>
<td>Q radioisotopes (m³/s)</td>
<td>Total count</td>
<td>Mixing (%)</td>
</tr>
<tr>
<td>---------</td>
<td>------------------</td>
<td>--------------------------</td>
<td>-----------------------------</td>
<td>-----------------------</td>
<td>-------------</td>
<td>------------</td>
</tr>
<tr>
<td>2A-1</td>
<td>Left</td>
<td>696</td>
<td>9.35</td>
<td>281</td>
<td>7.96</td>
<td>15780</td>
</tr>
<tr>
<td></td>
<td>Centre</td>
<td>(2283)</td>
<td>(330)</td>
<td>259</td>
<td>7.33</td>
<td>12040</td>
</tr>
<tr>
<td></td>
<td>Right</td>
<td></td>
<td></td>
<td>378</td>
<td>10.70</td>
<td>11630</td>
</tr>
<tr>
<td></td>
<td>Centre</td>
<td>696</td>
<td>9.43</td>
<td>338</td>
<td>9.57</td>
<td>15760</td>
</tr>
<tr>
<td></td>
<td>Right</td>
<td>(2283)</td>
<td>(333)</td>
<td>358</td>
<td>10.14</td>
<td>15120</td>
</tr>
<tr>
<td>2A-2</td>
<td>Left</td>
<td>776</td>
<td>8.56</td>
<td>302</td>
<td>8.55</td>
<td>12460</td>
</tr>
<tr>
<td></td>
<td>Centre</td>
<td>(2545)</td>
<td>(305)</td>
<td>301</td>
<td>8.62</td>
<td>12280</td>
</tr>
<tr>
<td></td>
<td>Right</td>
<td></td>
<td></td>
<td>297</td>
<td>8.41</td>
<td>12440</td>
</tr>
<tr>
<td>2B-2</td>
<td>Left</td>
<td>776</td>
<td>8.64</td>
<td>294</td>
<td>8.33</td>
<td>12280</td>
</tr>
<tr>
<td></td>
<td>Centre</td>
<td>(2545)</td>
<td>(305)</td>
<td>292</td>
<td>8.27</td>
<td>12180</td>
</tr>
<tr>
<td></td>
<td>Right</td>
<td></td>
<td></td>
<td>304</td>
<td>8.61</td>
<td>11660</td>
</tr>
</tbody>
</table>
of 700 m (2300 ft) between the points of injection and counting. Inadequate mixing occurred in spite of apparently uniform flow, which demonstrates that visual evaluation of flow conditions can be very misleading. When injection location was moved to a point 1676 m (5500 ft) downstream from the bend, satisfactory mixing was achieved and accurate discharge measurements were obtained, as shown by Table VI. In this second test the injection-to-counting distance was 776 m (2545 ft).

The experiment presents an opportunity for evaluation of the formula suggested for determining the minimum required distance for adequate mixing [13]. This formula is:

\[ L = K(Q)^{1/3} \]

where the constant \( K \) is equal to 200 for side injection and 50 for centre injection when \( Q \) is in ft\(^3\)/s. Substituting 330 ft\(^3\)/s for \( Q \) and 50 for \( K \) gives a minimum mixing length of about 350 ft or 107 m. The distance of 107 m (350 ft) was obviously too small in the above tests, and therefore the formula is not applicable to flow measurements in canals.

The simple mixing experiment shows the need for development of accurate criteria for judging the minimum distance required for adequate mixing. This may be especially true for discharge measurements in streams and rivers where the flow patterns are very complex compared with those in canals.

Improving the criteria for determining mixing distances is one approach to the solution of the mixing problem; another is to provide for simultaneous multiple injections across the canal. The latter method tends to produce uniform distribution initially and reduces the required mixing length materially. With the All-American Canal carrying about 225 m\(^3\)/s (8000 ft\(^3\)/s), five equal injections were made at equally spaced intervals across the canal in an apparently successful attempt to achieve complete mixing more quickly. The average measured discharge was within 1% of the discharge measured at a calibrated gauging station. In these tests made on 28 April 1962 the distance between the counting and injection locations was 2100 m (6900 ft). The advantage of simultaneous multiple injections is realized when these figures are compared with the test results previously discussed.

**Split-stream flow**

An interesting result from measurements performed in February 1962 on the A and B canals illustrates the principle of split-stream measurement. Injections were made upstream from the bifurcation, and counts were taken above the bifurcation and in each downstream leg. From the principle of split-stream flow measurement, if complete mixing takes place before the split, measurements downstream in either branch will give the flow in the main stream before the split. The following results illustrate this principle:

<table>
<thead>
<tr>
<th></th>
<th>A main stream</th>
<th>A branch</th>
<th>B branch</th>
</tr>
</thead>
<tbody>
<tr>
<td>First run</td>
<td>8.18 m(^3)/s</td>
<td>8.55 m(^3)/s</td>
<td>8.18 m(^3)/s</td>
</tr>
<tr>
<td></td>
<td>(289 ft(^3)/s)</td>
<td>(302 ft(^3)/s)</td>
<td>(289 ft(^3)/s)</td>
</tr>
</tbody>
</table>
Isotope sorption

Field observations during measurements in May 1962 in the South and Consolidated Canals indicated that aquatic weeds apparently absorbed and/or adsorbed radioisotopes. Large accumulations of weeds which collected on the probes showed this to be so. In canals containing large quantities of weeds serious errors in discharge determination could occur. One possible solution of this problem would be the use of a chemical form of isotope which is extremely stable and less likely to sustain absorption by plants.

Future applications and general observations

With the development of more precise equipment and field procedures the radioisotope method of flow measurement will be used in many field applications, including the rating of hydraulic structures, the calibration of gauging stations and the measurement of seepage losses from canals. For achievement of these objectives, however, minimum mixing distances must be established for use, and all losses of the isotopes must be minimized and/or accounted for.

The results of the field experiments made in canals provide general information which may be of value in the use of radioisotopes for flow measurement in natural waterways.

1. The pulse or total count method, when properly used, can give accurate and reliable results.
2. The techniques and field procedures are relatively simple and uncomplicated.
3. The principle of split-stream flow measurement offers the possibility of injection at a point of good mixing and the choice of a location downstream which may be more convenient for counting.
4. Great caution should be exercised in the choice of the points of injection and counting to assure that adequate transverse mixing has been accomplished.
5. The question of sorption, not only by aquatic plants but by sediments and channel walls as well, should be taken into account.

PROPOSED AUTOMATIC STATION

With the many encouraging results from the field tests and continuing advancement of nuclear and chemical engineering equipment, it is not too soon to propose an automatic stream gauging station employing the pulse or total count principle. The station would consist of two substations, the injection substation and the counting substation.

The injection substation would periodically inject a known quantity of tracer into the stream. A manifold-type injector possibly would be required to assure adequate mixing in a reasonable distance. The counting substation...
would be located downstream where, by trial, adequate transverse mixing was found to occur. A sample of the stream would be continually pumped through a counting chamber containing the Geiger or scintillation counters. The time between a given number of counts would be continuously recorded. For periods between flow measurements the time lapse would be greater, perhaps 100 times or more, than the time between count recordings during a flow measurement. An examination of the record chart would show the exact periods of flow measurements, and by the choice of times before and after the passage of the tracer a gross total count could be obtained. Total counts between flow measurements could be averaged to give an excellent measure of background radioactivity. From this information the net count, and hence the discharge, could be determined.

A station using radioactive tracers would be particularly useful in a stream where submergence or a shifting bottom profile has made it impossible to establish a stage-discharge relationship, i.e., where a staff gauge will not always indicate discharge.

ACKNOWLEDGEMENTS

This paper describes the results of the efforts of many persons, and it is impossible to recognize by name those who contributed to the study described. However, the contributions of all are acknowledged, including the major participants, C. L. Sweet and M. Hastings of the Bureau of Reclamation's regional office at Boulder City, Nevada, who spearheaded much of the field work, and R. L. Hansen and J. C. Schuster of the Bureau's Research Division, Denver, who not only assisted in many of the field measurements but also greatly assisted in the preparation of the technical material in this paper.

REFERENCES

Y. MIYAKE: Dr. Smith mentions in his paper that the loss of phosphorus is due mainly to chemisorption, but it seems to me that there is another way in which it can get lost in river waters, namely, as a result of uptake by various organisms. It is possible that the phosphorus in phosphate form is taken up as nutrient matter by the phytoplankton or weeds present in the water.

Although I do not have here any accurate data on the enrichment factor for gold in the organisms, I might mention in this connection that this factor is known to be fairly high in the case of certain of the metallic elements belonging to the transition group. Some of them may therefore not be suitable for tracer studies in river waters.

D.B. SMITH: I agree with your observations on P32. However, I would like to point out that we are not saying that phosphorus is a good river tracer: we used it fully aware of its drawbacks.

I wonder whether you could give us your views on the possibility of using lanthanum-140 as a tracer. What would be its chemical and biological behaviour in streams and the like?

Y. MIYAKE: I think that lanthanum might be unsatisfactory for this purpose because it has a greater tendency to form a hydroxide precipitate.

L.O. TIMBLIN, Jr.: With reference to his remark on the absorption of gold, does Mr. Miyake believe that chemical complexes such as EDTA could be used to decrease biological absorption?

Y. MIYAKE: Yes, if metallic elements are in the form of chelate compounds, they may in some cases be better tracers in water studies.

D.B. SMITH: Gold has been used by Krone in the United States and by the Australian Atomic Energy Commission for the direct labelling of mud and silt. The gold was adsorbed on silt by mixing gold chloride with a silt suspension. It has been our experience that gold is not very stable in dilute neutral solutions. When mixed with mud, almost all the gold was rapidly taken up by the surface of the silt and I think that to a limited extent, this is bound to occur on river beds. Of all the tracers which we have used and with which we have had experience in other work, we think that bromine-82 is the most satisfactory, particularly if it is used with care and in conjunction with sodium thiosulphate, for the conversion of free bromine to sodium bromide.

L.O. TIMBLIN, Jr.: Further to this work by Krone and others, in studies made a few years ago with Na22Cl the Na22 was completely absorbed by a column of clay. On the other hand, a chelated form of Na22 was not absorbed. It was with the idea that chelated forms of gold might perhaps not be absorbed by clays that I put my question to Mr. Miyake.

F. NEUMAIER: I should like to present some data on our experience in using radioactive tracers for measurement of flow velocity (m/s) and flow rate (m³/s) in open channels. The tracers used were I131, Br82 and Na24. In our method, a known amount of tracer is added to the stream and its concentration-time distribution registered at a measuring point sufficiently far downstream. Depending on the duration of the process, a distinction is made between instantaneous (pulse) and constant-rate (continuous) injection. The tracer is added by means of an injection-rod assembly,
the solution being forced by compressed air from a storage container into the water.

Detection is carried out either by direct measurement in the main stream or in a smaller stream fed from the main flow, or by collection of samples for later analysis. The samples, in turn, can be collected continuously or intermittently and the tracer contained in them measured directly or after enrichment. In one testing arrangement a scintillation counter is immersed directly in the main stream while another is located in a flow tank through which water from the river is pumped.

We sought to determine to what extent the accuracy of the flow rate as ascertained depends on amount of tracer and type of flow. In particular, we established the sensitivities and limits of our detection methods and studied the question of the minimum tracer amounts required to determine flow velocity and flow rate.

For example, to determine the flow velocity of a river having a flow rate of 43 m$^3$/s, we divided a 60-km section into five subsections. The tracer ($^{131}I$) was detected by sampling and chemically treating some 400 5-l samples. The average flow velocity over the whole section was 0.6 m/s. The half-widths, as a function of time of the water passage curves measured at the subsections, varied between 30 and 90 min, depending on the type of flow. Only 10 mc of $^{131}I$ were used for the entire 60-km section. The measurements were carried out with a directly immersed scintillation counter and four flow meters placed across the flow cross-section.

Allowing for the limits of error, the measurement results were found to be independent of the location of the sampling points and the flow cross-section, as was to be expected. It was further proved that tracer methods, provided that thorough mixing takes place, can achieve the accuracy obtainable with flow meters.

L.O. TIMBLIN, Jr.: Could Mr. Guizerix give us further details on his work with ion-exchange resins?

J. GUIZERIX: We have done only one experiment with an ion-exchange resin — for the purpose of assessing the factor which might be expected to increase the detection yield. The resin was mixed with the solution in an 18-l tank and then extracted by filtration. Not all the activity was extracted, but from the difference in counting values obtained from the scintillation probe placed in the centre of the tank, we got a value, $A_1$, which was proportional to the activity localized on the resin. The resin was then placed close to the scintillation crystal and yielded a counting rate $A_2$. We obtained an $A_2-A_1$ ratio of 11.5. Further work would be necessary in order to get a quantitative extraction. We have not done this because, as you will recall, our purpose is to measure large flows, for which we shall certainly have to use tritium.

D. B. SMITH: I see that the methods which Mr. Guizerix and I myself propose for river tracing are basically very similar. I would like to ask him whether he considers that the transport of a pump, battery and large container to the site of measurement is more convenient than the method of taking samples at regular intervals and sending these samples to a base laboratory. Is there any great advantage in the continuous pumping method?
J. GUIZERIX: If I remember correctly, studies made by Électricité de France show that a minimum of 25 samples have to be taken in order to construct the activity curve (your first procedure in the continuous sampling method) and to define its area to within 1%. Now, I believe that the volume of the sample used in your method is about 2 l. I therefore wonder whether the taking of 25 samples of 2 l each would not be as cumbersome as sampling by means of a pump. Our equipment is very light, and while not strictly portable, is nevertheless quite easy to transport. It should also be remembered that we make four samplings per section in our measurements. In the last analysis, however, one has also to face the fact that the gauging of rivers and large channels will always be a cumbersome operation, as far as equipment is concerned.

D. B. SMITH: In our work, we took more than the 25 samples which you mention, but we formed our individual 2-l samples by mixing six samples of about 350 ml each. Hence, all we had to transport from the site were a few 2-l bottles.

R. HOURS: I might mention that a German designer is producing a simple and light plastic device which floats on the river and takes a theoretically perfect average sample. The use of such an apparatus, if necessary with improvements, would doubtless be more convenient than continuous electric pumping or taking numerous samples at regular intervals.

L. O. TIMBLIN, Jr.: In connection with Mr. Guizerix's comments on safety and on the maximum permissible limits (MPL) on the radioisotope, our experiments have shown — and this has probably been your experience as well — that after complete mixing the concentrations were below the MPL's established by the United States Atomic Energy Commission. However, there is some public reluctance to accept the use of isotopes with longer half-lives, even though the MPL's are not exceeded. In the United States, there seems to be a greater willingness to permit the use of an isotope with a short half-life, even though the safety factors may not be any better. This may, in some cases, determine the selection of the isotope with respect to the health question which you discuss.

J. GUIZERIX: In my paper, I emphasized the wisdom of the French regulations with regard to the use of radioactive tracers in hydrology, especially tracers with a half-life of more than one week. Generally speaking, we in France do not like to use long-lived tracers in an extended medium. Since our purpose is to develop tracer applications, it may be assumed that this policy will bear fruit. There is reason to fear the day when large numbers of governmental or private organizations will be using long-lived tracers. I have in mind, in this connection, such things as the use of cobalt to determine blast furnace wear. This is not an application that I personally would favour.

R. HOURS: It is desirable, however, that consideration should be given to the introduction of less conservative regulations, in which greater account would be taken of the half-life of the radioisotope and in which the permissible dose would be calculated in the light of the very short duration of polluted water consumption. But the present regulations have the advantage of already being in existence and of permitting research to be carried on more easily. They are satisfactory for the time being.
J. CAMERON: Mr. Timblin includes in his paper a proposal for an automatic stream-gauging station. Does he consider that we have enough experience at present to predict all the parameters (e.g. vertical and lateral diffusion length, choice and activity of radioisotopes) that would have to be known to design such a station, or would further preliminary experiments have to be conducted?

L.O. TIMBLIN, Jr.: As far as any particular site is concerned, experiments could be performed for the purpose of determining the proper location of the injection and counting station. At present, we do not have sufficient knowledge to predict with certainty, and without experimentation, at what distance complete mixing will have taken place.
THE RADIOISOTOPIC DETERMINATION OF DIFFUSION COEFFICIENTS AND CURRENTS IN NATURAL WATERS. SURFACE COLLECTION OF RADIOACTIVE FALL-OUT ON A LARGE ALPINE LAKE. In an initial stage, the authors brought about the in situ diffusion of a radioactive emitter (sodium-22, in the form of Na$^{+}$) with very low activity, i.e. less than 1 mc. The diffusion laws applied in this case made it easy to predict the rate of diffusing of the "radioactive cloud". In this way, plotting the activities, as measured by two suitably placed gamma detectors, against time permits the estimation of a diffusion coefficient for the emitter used and, if appropriate, the measurement of current velocity at the point of measurement (in the case described, the Lake of Geneva).

Subsequently, and in conjunction with a geophysical study on the general problem of radioactive fall-out, the authors worked on the increases in radioactive fall-out caused by nuclear tests in the atmosphere (October 1961 - January 1962). Measurements of radioactivity were made every day on samples of Lake Geneva water taken at depths of 0 m, 10 m and 20 m and also on a collector of dry and moist atmospheric fall-out. Despite the complex pattern, over a period, of the increases in fall-out, the interpretation of the diagrams obtained can help to explain what happens to radioactive products in the diffusion and current conditions encountered.

The use of the diffusion of radioactive products in very small quantities and of the various highly sensitive techniques of nuclear detection now available can lead to a better understanding of in situ diffusion and of the movement of natural water masses.
L'utilisation de la diffusion de produits radioactifs en très faibles quantités et de différentes techniques de détection nucléaires à haute sensibilité actuellement disponibles peut conduire à une meilleure connaissance de la diffusion in situ et du mouvement des masses d'eaux naturelles.

OPÉRÉLLE COEFFICIENT DE DIFITUSSION ET VOROSTÉ TETSTEKNKES WOD PRI POMOSI YADERNOGO DETEKTIROVANIIA. ÉVOLUTION DE LA SURFACE DES CAPTATES RADIACTIVES SUR UN GRAND LAC ALPIN.

На первом этапе авторы вызвали диффузию "in situ" на радиоактивном излучателе, натрий-22, в виде Na+, с активностью ниже 1 милликюри. Законы диффузии, примененные в этом случае, легко позволяют предвидеть скорость "радиоактивного облака" во время диффузии. Таким образом, регистрация активности в зависимости от времени, измеренная с помощью двух надеждённых гамма-датчиков, привела к оценке коэффициента диффузии для используемого излучателя и к измерению скорости течения в той точке, где производилось измерение (в данном случае озеро Леман).

На втором этапе в соответствии с геофизическим исследованием общей проблемы радиоактивного выпадения, авторы использовали накопление радиоактивных осадков, возникших в результате ядерных испытаний, проведенных в атмосфере в октябре 1961 - январе 1962 г. Каждый день брались пробы воды Леманского озера на глубинах 0, 10 и 20 м, а также сухих и влажных атмосферных выпадений. Несмотря на сложность режима накопления в зависимости от погоды, толкование полученной диаграммы может способствовать разрешению проблемы выпадения радиоактивных продуктов в условиях диффузии и встречных потоков.

Использование диффузии радиоактивных продуктов в очень небольших количествах и различные методы обнаружения при помощи ядерных измерений высокой чувствительности могут привести в настоящее время к наилучшему пониманию диффузии "in situ" и движения масс природных вод.

DETERMINACIÓN POR DETECCIÓN NUCLEAR DE LOS COEYICIENTES DE DIFUSIÓN Y DE LAS CORRIENTES EN LAS AGUAS NATURALES.

Determinación de las precipitaciones radiactivas en un depósito de agua de un gran lago alpino. La primera etapa, los autores harían para la determinación in situ de un emisor radiactivo, el sodio-22 en forma de Na+, de actividad muy baja (inferior al milicurie). La aplicación de las leyes de la difusión a este caso facilita el cálculo de la velocidad de difusión de la "nube radiactiva". Por consiguiente, el registro en función del tiempo de las actividades medidas con dos detectores gamma dispuestos de modo conveniente permite asignar un coeficiente de difusión al emisor empleado y, eventualmente, evaluar la velocidad de la corriente en el punto en que se efectúa la medición (en el caso descrito, el lago Leman).

En una segunda etapa, en relación con un estudio geofísico del problema general de las precipitaciones radiactivas, los autores utilizaron la aportación de las precipitaciones radiactivas debidas a las explosiones nucleares en la atmósfera (octubre de 1961-enero de 1962). Midieron diariamente la radiactividad en muestras de agua del lago Leman tomadas a 0, 10 y 20 m de profundidad y en un colector de precipitaciones atmosféricas secas y húmedas. A pesar de la complejidad del régimen de aportaciones en función del tiempo, la interpretación de los diagramas obtenidos puede contribuir a resolver el problema del destino de los productos radiactivos en las condiciones de difusión y de corriente que reinan en la práctica.

La difusión de productos radiactivos en cantidad muy pequeña, unida a la utilización de diversas técnicas de detección nuclear de alta sensibilidad, permitirán obtener más datos sobre la difusión in situ y el movimiento de las masas de aguas naturales.

INTRODUCTION

Les techniques de détection nucléaire à haute sensibilité ont réalisé ces dernières années des progrès tels qu'il est possible de les appliquer avec succès à différents problèmes de géophysique ou relevant de l'hydro-géologie. En particulier, la détermination de concentrations en émetteurs radioactifs contenus dans les eaux naturelles en quantités extrêmement faibles conduit à la mesure in situ des coefficients de diffusion ou encore contribue à l'étude du mouvement des masses d'eaux et des sédiments.
Pour l'étude in situ des coefficients de diffusion et des courants, la méthode que nous avons utilisée consiste à observer la diffusion et le transport à la suite de l'introduction, dans des conditions définies, d'une quantité faible de produit radioactif [1]. Cette observation se fait au moyen de détecteurs de rayonnement γ, convenablement disposés autour du point d'introduction du produit radioactif. L'étude des diagrammes de variation d'activité γ détectée en fonction du temps permet, à chaque instant, de reconstituer l'allure du nuage radioactif en expansion et en déplacement, et par là, d'évaluer les coefficients de diffusion et les composantes du courant.

Nous avons visé, dans la seconde partie de notre étude, à améliorer la connaissance de l'évolution des produits radioactifs des retombées atmosphériques dans les eaux du Léman (lac de Genève). Pour cela, un vaste bilan systématique a été analysé [2]; des mesures de radioactivité β ont été effectuées périodiquement (pendant seize mois) sur des prélèvements d'eau du lac à 0, 10 et 20 m de profondeur. Des mesures de radioactivité β ont également été effectuées sur les retombées atmosphériques, sèches et humides.

Ces mesures nous ont permis l'observation comparée des évolutions de la radioactivité des eaux du lac et du taux de retombées radioactives en fonction du temps. La notion de surface efficace de collection du lac pour les produits radioactifs des retombées est introduite dans cette étude. La mesure de cette surface effectuée pendant un an pour le Léman permet de représenter le mécanisme de la contamination du lac.

1. DÉTERMINATION DES COEFFICIENTS DE DIFFUSION ET DES COURANTS PAR L'ÉTUDE DE LA DIFFUSION PROVOQUÉE D'IONS 24Na⁺

Les principaux facteurs de la diffusion sont: le phénomène de la diffusion moléculaire et le phénomène de diffusion tourbillonnaire turbulente (ce dernier facteur devenant prépondérant dans la plupart des cas réels) [3,4,5], le transport par voie physique (courants) ou physico-chimique (sédimentation, précipitation, absorption, élution), le transport par voie biologique (plancton, poissons, etc.).

L'application de la loi élémentaire de la diffusion [6] permet de prévoir la distribution des concentrations c(r, t) en cours de diffusion, d'un produit émis en une seule fois, en un point.

\[
c(r, t) = Q \left( \frac{1}{2\sqrt{\pi Dt}} \right)^3 \exp \left(-\frac{r^2}{4Dt}\right).
\]

Cette formule est valable en l'absence de courant.

r = distance du point d'observation à la position initiale de la source,
t = intervalle du temps séparant l'observation de la libération du produit en \( r = 0 \),
Q = quantité initiale de produit rejeté en une seule fois,
D = coefficient de diffusion (supposé constant, indépendant de la direction selon laquelle s'effectue la diffusion).
Traitement du problème avec détection nucléaire

Il est possible au moyen de détecteurs de rayonnement, de mesurer l'activité dans une zone définie entourant le détecteur, après libération d'un produit radioactif [7, 8, 9, 10].

L'étude des variations d'activité conduit à une évaluation des concentrations et permet par application de la formule (1) d'atteindre les valeurs des coefficients de diffusion.

Dans la méthode que nous suggérons, il est possible de n'utiliser qu'un seul détecteur si celui-ci permet d'obtenir la courbe complète et précise de l'activité en fonction du temps en un point situé à une distance donnée de l'endroit où est libéré l'émetteur radioactif.

L'observation du maximum d'activité en fonction du temps et l'application de la formule (1) conduisent alors de façon simple à la valeur du coefficient de diffusion.

Description de la méthode utilisée

La méthode utilisée consiste à libérer rapidement une certaine quantité d'émetteur radioactif dans des conditions ne perturbant pas le milieu étudié. Deux sondes de détection d'activité γ environnante sont utilisées; elles peuvent être disposées soit au contact de la source radioactive initiale, soit à une certaine distance.

Le sodium-24 a été choisi pour différentes raisons: il est un des ions les plus abondants de l'eau de mer et il existe en quantité notable dans les eaux continentales. L'énergie élevée du rayonnement γ du sodium-24 (2,7 MeV) permet une plus grande sensibilité de détection, au moyen d'une méthode de discrimination en fonction de l'énergie du rayonnement γ.

Lors des expériences effectuées dans le lac Léman et décrites ci-après, des précautions spéciales ont été prises afin de veiller à ce que la densité du produit libéré contenant les ions 24Na soit pratiquement égale à celle rencontrée in situ au point de mesure, afin de ne pas perturber le milieu au moment de la libération du produit radioactif.

Le dispositif d'injection du produit comprend un ballon de caoutchouc très fin rempli du liquide-source à émettre, dont la membrane est tendue, et un percuteur mécanique actionné depuis le bateau où se trouvent les ensembles électroniques.

Les deux sondes sont fixées sur un cadre métallique et peuvent être mises en place à différents endroits du cadre, celui-ci ayant en son centre le dispositif d'injection du produit radioactif. La sonde n° 1 contient un cristal d'iode de sodium activé au thallium NaI(Tl) 4,5 cm X 5 cm et un photomultiplicateur 53 AVP. La sonde n° 2 contient un cristal NaI(Tl) de 10 cm X 8 cm, un photomultiplicateur 54 AVP et un préamplificateur à transistors.

Les deux sondes sont reliées chacune par un câble multiconducteur approprié au bateau porteur des ensembles électroniques. Chaque sonde est suivie, sur le bateau, d'un ensemble électronique alimenté à partir d'un convertisseur alimenté à bord et comprenant une échelle de comptage à discrimination d'amplitude et un enregistreur dont la vitesse de déroulement du papier est 10 mm/min.
Deux expériences ont été effectuées en avril 1961 en utilisant ces dispositifs: l'observation de l'allure du nuage de produit en diffusion et de son déplacement par rapport au dispositif fixe a permis, par l'utilisation des formules citées [1], de mesurer les valeurs du coefficient de diffusion $D$ pour l'ion sodium et de la vitesse $V$ du courant à l'endroit de l'expérience:

$$D = 82 \text{ cm}^2/\text{s},$$
$$V = 1,8 \text{ cm/s}.$$  

(Une mesure de courant par une méthode classique de l'hydrologie a pleinement confirmé cette dernière valeur.)

On voit que les effets de la diffusion et du courant ont pu être distingués en une seule expérience. On peut en déduire que la méthode, applicable à divers émetteurs $\gamma$, permettrait de mesurer avec un matériel relativement peu important les coefficients de diffusion in situ à des profondeurs quelconques et, moyennant un appareillage plus important, les composantes du courant.

La connaissance simultanée des valeurs in situ, composantes du courant et coefficients de diffusion pour des émetteurs $\gamma$ (dans des états physico-chimiques quelconques), étant particulièrement précieuse dans de nombreux problèmes de dilution, la méthode proposée peut donner lieu à la conception d'un appareillage particulièrement adapté au problème posé [1].

2. ÉTUDE SYSTEMATIQUE DE L'ÉVOLUTION DES RETOMBÉES RADIOACTIVES DANS LES EAUX DU LAC LÉMAN, (SEPTEMBRE 1961 - DÉCEMBRE 1962)

Détermination de la surface efficace de collection du lac pour les retombées radioactives

Dans les eaux naturelles, les retombées radioactives ayant pour origine les essais nucléaires atmosphériques sont introduites selon un processus involontaire et irrégulier qui conduit à des répartitions beaucoup plus complexes que dans le cas d'expériences de diffusion provoquée.

Néanmoins, l'observation systématique de l'évolution dans l'eau des concentrations en produits provenant des retombées peut être utilisée avec profit pour la solution de nombreux problèmes traitant de leur accumulation et de leur élimination dans les masses d'eaux naturelles [2 et 11].

Nous avons mesuré dans le lac Léman les activités en émetteurs radioactifs artificiels prépondérants dans les retombées atmosphériques depuis la reprise des essais nucléaires dans l'atmosphère en septembre 1961. Des mesures bêta globales ont été pratiquées sur les fractions solubles et insolubles de prélèvements de 2 l d'eau effectués régulièrement à des profondeurs de 0 m, 10 m, 20 m, en un point du lac, situé à 150 m au large de Thonon-les-Bains, Haute-Savoie (fig.1).

La radioactivité bêta globale des retombées sèches ou humides a été mesurée séparément, à partir d'un collecteur de retombées atmosphériques de surface de collection 0,1 m$^2$, du type CRAPAL [12], situé à Thonon-les-Bains, sur un espace dégagé, en bordure du lac (fig.1).
La dynamique de l'enrichissement du Léman en produits radioactifs peut être étudiée à partir de nos mesures. Néanmoins, étant donné la complexité du régime des injections des retombées, il s'avère que l'échelle de temps la plus adaptée à l'observation des phénomènes de contamination est de l'ordre du mois (fig. 1). En dessous de cet intervalle, les fluctuations par trop importantes masquent l'allure générale du phénomène[2].

Dans la période étudiée, les principaux produits radioactifs des retombées étaient: le zirconium-95, niobium-95, le cérium-144, praséodyme-144, le ruthénium-106, rhodium-106. Ces produits ont été identifiés et mesurés par spectrométrie gamma à haute sensibilité [13, 14, 15, 16].

Les mesures effectuées jusqu'en octobre 1962 sur les retombées ont montré une injection, intégrée sur un an dans la région Lémanique, de 0.54 c/km². Ces émetteurs radioactifs se présentaient sous forme de retombées sèches (58%) et sous forme de retombées humides (42%). L'activité totale du lac était, en octobre 1962, de 460 c.

L'atténuation dans le lac de la radioactivité réside principalement dans la décroissance radioactive des émetteurs et non dans le renouvellement des eaux du lac, le débit du Rhône en aval (242 m³/s) [17, 18, 19] ne contribuant que faiblement à l'élimination des produits en accumulation dans le lac.
Les apports dans le lac en nuclides radioactifs ont pour origine l'arrivée sur l'ensemble de la surface du bassin de drainage des produits des retombées; une partie seulement parvient jusqu'au lac lui-même, à cause des effets de décantation ou de rétention dans les sols pour les produits véhiculés par les eaux de ruissellement.

La «surface efficace de collection» représentée pour nous la surface hypothétique de collection totale des produits atmosphériques vers le lac, alors que la surface du bassin versant, déterminée selon la topographie [18], est la surface maximale de collection des eaux d'origine atmosphériques et phréatiques.

Le rapport de l'aire efficace de collection à l'aire du bassin versant permet d'estimer la fraction de l'ensemble des produits atmosphériques tombés sur la surface du bassin versant, et qui, transportés par les eaux de ruissellement, atteignent effectivement le lac.

La connaissance simultanée de la radioactivité totale en fonction du temps, du Léman et des injections rapportées à une surface connue permet d'accéder à la mesure de l'«aire efficace de collection» pour les produits considérés.

L'évaluation de l'aire efficace est effectuée par comparaison entre l'activité réelle observée dans le lac et l'activité théorique des eaux de ce lac que l'on peut reconstituer d'après les mesures effectuées sur les retombées collectées. Cette méthode est décrite en annexe.

L'injection totale en retombées radioactives peut être déduite des mesures effectuées sur ces collections périodiques sur 0,1 m². En effet, l'homogénéité climatique de la région lémanique est connue [18], les pertur-
bations microclimatiques sont localisées, et l'effet des chutes de neige sur les versants jurassiques du bassin de la Venoge et préalpins et alpins des bassins de la Dranse et du Rhône ne fausse pas les conclusions que l'on peut tirer de la représentativité globale du collecteur de retombées à l'échelle du bassin de réception du Léman.

La connaissance des coefficients de diffusion, des processus limnologiques et les mesures complémentaires d'ordre hydrologique [17,18] effectuées simultanément pendant l'étude autorisent à considérer les prélèvements comme représentatifs de la totalité des eaux du lac, lorsqu'il y a homogénéité de ce lac holomictique et monomictique chaud.

L'évolution de l'aire efficace du bassin de drainage alimentant le lac Léman a pu être observée sur une année (fig. 2). Cette aire est considérablement inférieure à l'aire totale du bassin (8900 km²) et de l'ordre de grandeur de la surface réelle du lac (582 km²); ceci provient des effets de décantation et de rétention par le sol des atomes radioactifs [20,21] des retombées avant que ceux-ci aient pu atteindre le lac. Au mois de juin, l'augmentation considérable de l'aire efficace qui est observée (fig. 2) est due au phénomène de la fonte des neiges, alimentant le lac en produits des retombées accumulées au cours du printemps (période de maximum dans le régime des retombées), associé à l'effet de stratification thermique estivale.

CONCLUSION

Nous pouvons maintenant comparer les renseignements donnés par chacune des deux méthodes exposées plus haut.

Les conditions d'emploi ont été exposées: la première méthode consiste à libérer une certaine quantité d'un émetteur radioactif et à observer sur place immédiatement la diffusion du produit libéré [1 et 22]; la seconde, utilisant l'injection involontaire des émetteurs radioactifs des retombées atmosphériques, consiste à mesurer en laboratoire différentes grandeurs accessibles — radioactivité des eaux, taux d'injection en émetteurs radioactifs, caractéristiques des émetteurs, etc.

La première méthode permet d'accéder à la mesure des coefficients de diffusion et de courants. La seconde ne permet pas d'atteindre de tels résultats dans le cadre des moyens et des échelles de temps et d'espace utilisés. La recherche analytique des paramètres de mouvement des produits dans les eaux du lac nécessiterait l'enregistrement continu des radioactivités observées in situ, au moment d'une pluie fortement radioactive par exemple. Malgré la grande sensibilité de nos détecteurs, les ordres de grandeur des activités rencontrées dans les retombées atmosphériques sont tels qu'il apparaît encore difficile d'envisager une telle expérience, nécessitant de plus une implantation permanente in situ des dispositifs détecteurs.

La deuxième méthode, fondée sur l'utilisation des prélèvements discontinus d'eau du lac et de relevés périodiques de collecteurs de retombées ne peut éclaircir l'aspect microphénoménologique du lac devant de nouvelles injections. Nous avons donc délibérément adopté une échelle de temps — moyennes mensuelles — permettant l'intégration des fluctuations non sig-
nificatives et, à partir de l'homogénéité habituelle du lac, de déduire les caractéristiques de la surface efficace de collection des retombées atmosphériques. L'application de cette méthode conduit à un critère pour l'aptitude à la consommation des eaux naturelles malgré une augmentation du taux de retombées radioactives: à volume disponible égal, les bassins où la contamination est la moins importante sont ceux pour lesquels la surface efficace de collection est la plus faible.

ANNEXE

L'équation suivante, appliquée à la totalité du lac Léman, peut rendre compte des différents processus d'accumulation et d'élimination des produits radioactifs des retombées:

\[
\frac{dN}{dt} = \frac{0.693}{T} N - \frac{0.693}{r} \frac{V}{T_R} n_t - \frac{V}{T_R} n_t + Q.
\] (1)

\(N\): nombre total d'atomes radioactifs, présents dans le lac Léman, fonction du temps \(t\).
\(T\): période de décroissance radioactive, supposée identique pour toutes les espèces de nucléides. La période \(T\) est liée à la constante de désintégration \(\lambda\) par

\[
\lambda = \frac{0.693}{T}.
\]

\(r\): période d'élimination moitié par processus divers: absorption, sédimentation, etc.
\(T_R\): temps de résidence des eaux.
\(n_t\): concentration moyenne en atomes radioactifs, principalement à l'entrée du Rhône dans le lac Léman.
\(n_s\): concentration en atomes radioactifs, à la sortie du lac Léman, en général, \(n_s \neq N\).
\(V\): volume total du lac.
\(Q\): flux total de l'injection atmosphérique pour la surface de collection du lac Léman en atomes radioactifs (retombées directes), fonction du temps \(t\), soit \(Q(t)\).

Ainsi, les trois premiers termes du second membre, de forme comparable, sont respectivement inversement proportionnels à \(T\) période radioactive, à \(r\) période d'élimination et à \(T_R\) temps de résidence des eaux. On voit ainsi que l'effet d'élimination prépondérant correspond à la période ou au temps de résidence le plus court.

Les périodes apparentes \(T\) mesurées d'après la décroissance de l'activité des prélèvements en fonction du temps sont comprises en 120 et 360 j (tableau 1). Le temps de résidence \(T_R\) des eaux dans le lac Léman est de 4230 j[18]. Le volume total du lac Léman a été estimé à 89 km\(^2\). Le débit de l'émission du Rhône à Genève est de 243 m\(^3\)/s en moyenne et a été réparti comme suit:

- Rhône à son entrée dans le lac Léman : 181 m\(^3\)/s,
- Affluents rive française : 32 m\(^3\)/s,
- Affluents rive suisse : 16 m\(^3\)/s,
- Eau atmosphérique (apport direct) : 3 m\(^3\)/s,
- Eau de ruissellement : 11 m\(^3\)/s.

L'analyse sur un an des diagrammes des activités des prélèvements [2] (fig. 1) conduit à évaluer le temps d'élimination pour ces nucléides à environ 6 mois, ce qui est une grandeur comparable aux périodes de décroissance radioactives mesurées par ailleurs. L'effet d'élimination par adsorption et précipitation n'apparaît donc pas comme prépondérant en général.

L'effet d'atténuation opposé à l'accumulation de l'activité due aux injections peut donc être expliqué principalement par la décroissance radioactive, et on peut écrire (1), en regroupant les termes d'élimination et les termes d'injection:

\[
\frac{dN}{dt} = -\frac{0.693}{T} N + Q.
\] (2)
### TABLEAU I
COMPARAISON DES RÉSULTATS

<table>
<thead>
<tr>
<th></th>
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</thead>
<tbody>
<tr>
<td>$n_1$ pc$_1$/l</td>
<td>1.43</td>
<td>3.00</td>
<td>3.64</td>
<td>5.16</td>
<td>-</td>
<td>-</td>
<td>4.0</td>
<td>6.1</td>
<td>5.3</td>
<td>11.1</td>
<td>5.3</td>
<td>4.4</td>
<td>4.3</td>
<td>5.2</td>
<td>4.6</td>
<td>5.4</td>
</tr>
<tr>
<td>$q_1$ pc$_1$/0.1 m$^2$</td>
<td>150</td>
<td>20300</td>
<td>9360</td>
<td>12940</td>
<td>-</td>
<td>-</td>
<td>8720</td>
<td>12250</td>
<td>20980</td>
<td>7520</td>
<td>6280</td>
<td>5360</td>
<td>9620</td>
<td>13100</td>
<td>14260</td>
<td>11970</td>
</tr>
<tr>
<td>$\Sigma q_1$ corrigé pc$_1$/0.1 m$^2$</td>
<td>150</td>
<td>20400</td>
<td>25500</td>
<td>32800</td>
<td>26300</td>
<td>20800</td>
<td>25400</td>
<td>33000</td>
<td>49000</td>
<td>48400</td>
<td>47600</td>
<td>41300</td>
<td>49500</td>
<td>53900</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$T$ (journée)*</td>
<td>380</td>
<td>412</td>
<td>330</td>
<td>178</td>
<td>-</td>
<td>---</td>
<td>364</td>
<td>243</td>
<td>293</td>
<td>192</td>
<td>255</td>
<td>355</td>
<td>480</td>
<td>255</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$S$ (10$^3$ km$^2$)</td>
<td>-</td>
<td>1.3</td>
<td>1.3</td>
<td>1.3</td>
<td>-</td>
<td>-</td>
<td>1.4</td>
<td>1.6</td>
<td>1.0</td>
<td>2.0</td>
<td>1.0</td>
<td>0.9</td>
<td>0.8</td>
<td>0.9</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* C'est-à-dire $\frac{1}{K} \sum_{K=1}^i q_K e^{-\lambda(t-K)\Delta t}$, cf. formule (7)

** Période apparente en jours, calculée d'après deux mesures successives effectuées sur les prélèvements d'eau du lac. La première de ces deux mesures a été effectuée environ 17 jours après le prélèvement; la seconde au cours du mois de janvier 1963.
La radioactivité bêta globale des produits en solution et en suspension dans deux litres de prélèvement a été mesurée séparément. Ces mesures ont été quotidiennes du 12 septembre 1961 au 16 décembre 1961 pour des prélèvements effectués à 0 m, 10 m, et 20 m de profondeur en un point situé à 150 m au large du Centre de recherches géodynamiques de Thonon-les-Bains, par 53 m de fond ; elles ont été bi-hebdomadaires de mars à décembre 1962, dans les mêmes conditions.

Le nombre total N d'atomes radioactifs contenus dans le Léman est déduit de chaque mesure par application de la formule suivante, dans laquelle on néglige la correction due à l'intervalle de temps t séparant la mesure nucléaire du prélèvement, dont la valeur moyenne égale à 17 jours est petite par rapport aux périodes de décroissance radioactive mesurées :

\[ n = \frac{v}{V} N e^{-\lambda t} \]  

(3)

\( v \) : volume du prélèvement.
\( V \) : volume total du lac Léman.
\( t \) : intervalle de temps compris entre la date de la mesure et celle du prélèvement.

La formule suivante, comparable à la formule (3), relie l'activité bêta globale \( q \) des mesures nucléaires effectuées sur les collections de retombées sur la surface \( s \) au flux total \( Q \) de l'injection atmosphérique rapporté à l'aire efficace de collection du lac :

\[ q = \frac{S}{S} Q \Delta t e^{-\lambda t} \]  

(4)

avec les mêmes hypothèses simplificatrices que celles adoptées pour la formule (3).

L'intégration de la formule (2) conduit au calcul théorique des nombres d'atomes radioactifs contenus dans le lac :

\[ N = \sum_{k=1}^{i} q_k \frac{S}{S} e^{-\lambda (i-K)\Delta t} \]  

(5)

\( N \) : nombre calculé total d'atomes radioactifs présents dans le Léman, en moyenne pour le \( i \)-ième intervalle de temps \( \Delta t \).
\( Q \) : moyenne pour le \( k \)-ième intervalle de temps \( \Delta t \) de l'activité bêta globale des retombées (avec \( i > k \)).

De plus, la formule (3) peut être écrite

\[ \frac{N}{N} = \frac{\bar{n}}{v} \]  

(6)

\( N \) : nombre moyen total d'atomes radioactifs présents dans le Léman, déduit des moyennes mensuelles des activités \( n \) du prélèvement.

La confrontation des formules (5) et (6) conduit à l'obtention des aires efficaces de collection du lac Léman pour les produits considérés et les intervalles de temps \( \Delta t \) envisagés :

\[ S = \frac{v}{\bar{V}} \frac{\bar{n}}{\sum_{k=1}^{i} q_k \frac{S}{S} e^{-\lambda (i-K)\Delta t}} \]  

(7)

avec \( \frac{v}{\bar{V}} = 8,9 \cdot 10^6 \text{ km}^2 \)

Les résultats obtenus d'après cette confrontation sont contenus dans le tableau I et la figure 2 ; on voit que les aires efficaces moyennes de collection pour un mois sont comprises entre 800 km² et 2000 km², soit de 1,4 à 3,5 fois la surface réelle du lac ou de 0,10 à 0,25 fois la surface du bassin de drainage [2 et 18] .
Références


Discussion

P.C. LÉVÊQUE (Chairman): I wonder whether the rather abrupt variation in the effective collection-area curve does not correspond to the influx of large quantities of clay minerals that takes place when the snows melt. I am thinking of the fairly considerable capacity for fixation possessed by these minerals, particularly with respect to cations.

R. HOURS (on behalf of R. Chesselet et al.): I think that two effects are possible when there is a sudden prolonged influx of water from melting snow. In the first place, the water is liable to bring with it suspended particles of clay which have already fixed radioactive cations. This rapid discharge of clay into the lake would give rise to an increase in specific activity at the water levels recorded (0, 10 and 20 m) in spite of the fact that there would have been no simultaneous increase in actual fall-out. There would be a concomitant increase in the effective collection area, as is shown in Figs. 1 and 2. The particles would then settle on the bottom and this would bring about a decrease in the specific activity in the area of measurement. The latter effect would tend to be counteracted by the thermal stratification
in the summer months. There is a discussion of these various phenomena at the end of the paper.

Secondly, the clay particles that were not contaminated would fix the ions already present in the lake before the influx. It would be possible for these particles, on settling, to remove some of the radioactive ions or aerosols from the surface waters of the lake.

It is clear that the specific-activity measurements have to make allowance for the activity of these suspended particles if a correct estimate of the effective collection area is to be made.

Y. MIYAKE: I would like to mention that a similar attempt is being made at my own laboratory to estimate diffusion coefficients. In our case, however, we are studying the diffusion coefficients in the open seas by measuring the horizontal and vertical distribution of Sr$^{90}$ and Cs$^{137}$.

R. HOURS: It is in fact intended to use this technique for studying oceanic diffusion with a view to obtaining data on the behaviour of radionuclides in solution or suspension in sea water, as the result of atmospheric fallout or the disposal of effluents on the sea bed. The dispersion of radionuclides will be measured down to the depths of 5000 or 6000 m; it is planned to use a more elaborate device for the purpose, consisting of six scintillation probes spaced out at regular intervals over the surface of a sphere.

In this connection, I might also mention the somewhat larger scintillation device that has been developed by the William Johnston Laboratory in the United States. It consists of a 1-m diam. ring of 16 scintillation probes, and measurements are made of the time taken by the radionuclide to pass each of these detectors. As you see, the main purpose here is to measure current velocity and direction. With the French technique I have just described, however, the aim is to obtain data on diffusion — although obviously certain assumptions have to be made with regard to the diffusion equation. In the paper I presented, only isotropic diffusion is considered, but reference [1] also deals with the problem of anisotropic diffusion in various directions.

Y. MIYAKE: Do you think that the immersion of the detectors in any way disturbed the diffusion-coefficient measurements?

R. HOURS: When detector No. 1 is very close to the initial source, I suppose there might be some slight disturbance. When the detector is far from the point source, it would be negligible. I would emphasize, however, that I am not the author of the paper.
AN INVESTIGATION OF THE HYDRAULICS OF AN UNDERGROUND POOL WITH IODINE-131

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Abstract — Résumé — Аннотация — Resumen

AN INVESTIGATION OF THE HYDRAULICS OF AN UNDERGROUND POOL WITH IODINE-131. Water movement in a natural underground pool, semi-tidal and partly saline, was investigated with a radioactive tracer.

50 mc of iodine-131 was rapidly mixed with the water in the pool at low tide, and the volume of the pool was established by the dilution technique to be 3075 m$^3$. Subsequent dilution resulting from tidal movement showed that the pool was part of a larger water system, and measurements permitted the throughput of tidal water and the dispersion of the tracer into adjoining parts of the system to be estimated. After the initial dispersion the pattern of the decreasing tracer concentration was followed for four days and permitted the fresh water throughput to be estimated at 26 m$^3$/h.

This information will assist engineers to design a method for the extraction of potable water from the pool.

ETUDE DES CARACTÉRISTIQUES HYDROLOGIQUES D'UNE NAPPE SOUTERRAINE, À L'AIDE D'IODE 131.
On a étudié, à l'aide d'un radioindicateur, le mouvement des eaux d'une nappe souterraine naturelle, soumise à l'action des marées et partiellement salée.

On a mélangé rapidement 50 mc d'iode 131 à l'eau de la nappe, à marée basse; à l'aide de la méthode de dilution, on a évalué à 3075 m$^3$ le volume de la nappe. La dilution ultérieure due à la marée a révélé que la nappe faisait partie d'un réseau hydrologique plus vaste; des mesures ont permis d'évaluer la quantité d'eau amenée par la marée ainsi que la dispersion de l'indicateur vers les parties avoisinantes du réseau. Après la dispersion initiale, on a suivi pendant quatre jours la baisse du degré de concentration de l'indicateur, ce qui a permis d'évaluer à 26 m$^3$/h l'heure le débit de l'eau douce.

Cette étude aidera les ingénieurs à établir une méthode pour extraire de l'eau potable de la nappe.

ИЗУЧЕНИЕ ГИДРАВЛИЧЕСКИХ СВОЙСТВ ПОДЗЕМНОГО БАССЕЙНА С ПОМОЩЬЮ ЙОДА-131. С помощью радиоактивного индикатора изучалось перемещение воды в естественном подуширном и частично засоленном подземном бассейне.

50 милликюри йода-131 быстро смешивались с водой в бассейне при небольшом приливе; объем бассейна, измеренный методом разбавления, оказался равным 3075 м$^3$. Последующее разбавление в результате движения прилива показало, что бассейн является частью более крупной водной системы. Измерения дали возможность определить количество приливной воды и дисперсию индикатора в сообщающейся части системы. После определения величины первоначальной дисперсии в течение четырех дней прослеживался процесс уменьшения концентрации индикатора, что дало возможность определить скорость поступления свежей воды, которая оказалась равной 26 м$^3$/час.

Эта информация поможет инженерам при разработке метода извлечения питьевой воды из бассейна.

ESTUDIO DE LA HIDRÁULICA DE UN ESTANQUE SUBTERRÁNEO CON YODO-131. Los autores estudiaron con ayuda de un indicador radiactivo los movimientos del agua en un estanque subterráneo natural, parcialmente salino, y algo influenciado por las mareas.

Durante la marea baja, añadieron rápidamente al agua 50 mcuries de yodo-131 y, por la técnica de dilución, determinaron el volumen del estanque (3075 m$^3$). La dilución ulterior originada por la subida de la marea demostró que el estanque forma parte de un sistema mayor y las mediciones permitieron evaluar las variaciones de volumen debidas a las mareas y a la penetración del indicador en otras partes adyacentes del sistema. Después de la dispersión inicial, observaron la curva de disminución de la concentración del indicador durante cuatro días y calcularon en 26 m$^3$/h el caudal de agua.

Estos datos ayudarán a los ingenieros a establecer un método para extraer agua potable del estanque.
INTRODUCTION

This investigation was carried out on an underground pool situated near sea level in a natural cavern in a coastal karstic area. The pool contains partially saline water and is a potential source of supply of fresh water. For assistance in the extraction of potable water more information is required concerning the hydraulics of the pool.

The pool was formed along a fault plane running N-S in the limestone (Fig. 1). Its surface area is 300 m² and the average depth about 5 m. At the south end the depth increases, and a chamber 4 m square leads diagonally downwards for over 30 m.

![Fig. 1](image-url)
The water level is about 1 m above mean sea level and rises and falls with the tide. The tidal range in the pool is about 20 cm compared with a range of 120 cm in the sea, and the pool cycle lags behind the tidal cycle by about 1.5 h.

The existence of the level difference between the sea and the pool indicates that a considerable volume of fresh water is probably held within fissures in the rock. This arises from the percolation of rain through the limestone over a limited catchment area. The time lag in the tidal cycle shows that the connection between the pool and the sea offers considerable resistance to flow, and it is unlikely that water involved in the pool cycle is exchanged rapidly with the sea.

Pumping data and salinity measurements indicate that there is a flow of fresh water into the pool during times of rain and that there is a relation between the amount of fresh water available and the recent rainfall on the catchment. If the pumping rate is excessive, the pool soon becomes highly saline.

HYDRAULIC PROBLEMS

For exploitation of the water resources of the pool further information is required on the hydraulics of the system. One problem is the determination of whether there are extensive water bodies of large volume adjacent to the known pool. Associated with this, information is required on whether the pool is part of the main water system of the limestone or is an offshoot in the form of a "cul de sac" at the end of part of the system. An estimate of the fresh water throughput during a short period could be used in conjunction with rainfall data to indicate the potential fresh water supply obtainable from the pool.

RADIOACTIVE TECHNIQUES

Two radioisotope tracer techniques were used in the study of the hydraulics of the pool.

(a) Direction of flow of currents
By observation of the direction of movement of a pulse of radioactive solution near openings in the walls or floor of the pool it was hoped to establish the general direction of movement of water during the flooding and ebbing tide. The value of the method is dependent on the general turbulence of the pool producing lower water velocities than do the tidal currents.

(b) Dilution and mixing investigation
By the mixing of a known amount of radioactive tracer with the pool and measurement of the radioactivity per unit volume after the mixing is complete, the volume of the pool can be calculated.

The subsequent reduction of concentration of tracer within the mixed pool can give information about tidal throughput, mixing into adjacent volumes of water and the fresh water throughput of the system. A single set of measurements can give information about these three factors but cannot differentiate unambiguously between the three.
EXPERIMENTAL PROCEDURE

(a) Direction of flow of currents

A buoyant "spider" made up of six radial spokes each 150 cm long was constructed to float on the surface of the pool. Six 24-cm halogen-quenched Geiger counters in waterproof housings were suspended singly from each of the six extremities of the spokes and were adjusted to hang at the required depth. About 10 ml of radioactive solution was released by compressed air from a container suspended at the centre of the spider. The movement of the tracer was followed by measurement of the count rate of each of the counters in turn on a ratemeter to which the counters were connected by 15 m of single-core, armoured cable. A six-way switch enabled any counter to be selected.

The radioactive tracer used in the investigation was iodine-132 (half-life, 2.26 h). This isotope emits gamma rays with an average energy of about 0.7 MeV and can be obtained by elution of an alumina column containing its parent isotope, tellurium-132, (half-life, 78 h) with water. The short half-life of the iodine enables repeat experiments to be carried out without interference from injections on the previous tide. Approximately 1 mc was used in each injection.

The spider was assembled in the south corner of the pool (see Fig. 1) opposite the known chamber leading downwards. An existing walkway and small dinghy afforded access. The detectors were adjusted to 7 m depth, which placed the most northerly near the bed of the pool.

After a trial run, an injection was made on a rising tide, 2 h after low tide in the pool. Indications of tracer were obtained on the three most northerly counters within 3-6 min. The activity rose to a maximum of about 1000 cpm on the north counter after 15 min and on the two adjacent counters in 50 min. All counts were near background after 80 min.

A second injection was made 1½ h after high tide in the pool. High counting rates (up to 1500 cpm) were measured on the south counter, with the maximum after 1 h. Fairly high counts were registered on the southwesterly counter, with the maximum after 1½ h. During the period up to 2 h all counters showed some significant reading, indicating the degree of turbulence in the pool.

The spider was moved 2 m north for a third injection 1 h after high tide in the pool. This indicated water movement between northwest and south-west on a falling tide.

The three injections showed that the south end of the pool formed an entrance area for water during the flood tide and an exit during the ebb. It appeared that the southwesterly wall formed the exit point in the region where the known large chamber led downwards from the base of this wall.

(b) Isotope dilution investigation

For a study of the volume of water in the pool and water movements through the pool, the radioactive isotope was mixed as rapidly as possible with the visible water in the pool. Mixing was continued throughout the in-
INVESTIGATION OF POOL HYDRAULICS

vestigation, and the subsequent decrease of specific activity of the tracer was measured.

For obtaining information during the first tidal cycle, the tracer had to be mixed with the pool as rapidly as possible. An electric pump capable of recycling 50 m³/h of water took water from about 3 m depth at the north end of the pool and discharged at two points near the centre and on the east wall. This discharge caused little disturbance and did not appear to cause effective mixing.

Three 5-cm airlines were weighted and sunk in the pool to depths of 4-6 m at positions shown on Fig. 1. Two compressors supplied approximately 20 m³/min of air to these lines. Enormous turbulence was caused by the air, columns of water and air rising from each airline outlet. Water dispersed radially on the pool surface from above each point, and the pool became opaque in all parts within a few minutes of injection of the air.

The radioisotope selected for this investigation was iodine-131. The half-life (8.04 d) was suitable, and the I⁻ ion is not normally adsorbed during passage through limestone rock. For reduction of the chance of adsorption, about 250 g of potassium iodide carrier was added to the pool. The natural chloride and bromide content of the saline water would also reduce any tendency to adsorption.

Approximately 50 mc of iodine-131 was used in 16.35 ml. Before this was added to the pool, two weighted aliquots of 0.1352 g and 0.1379 g were taken and diluted with inactive potassium iodide carrier to approximately $8 \times 10^{-5}$ mc/ml to form two standards from which the specific activity of the pool and hence its volume could be calculated.

The iodine solution was added to the centre of the pool at low tide in the pool. The mixing was observed on a submerged scintillation counter operating into a chart recorder. Large fluctuations of counting rate did not occur, and the tracer was progressively diluted to give a steadily declining reading after about $\frac{1}{2}$ h.

Quantitative measurements of the specific activity of the pool were obtained by means of single 2-l samples taken every half hour. The samples were assayed on a scintillation counter with a 2-1 re-entrant polyethylene bottle and a 2-inch sodium iodide crystal with an efficiency of detection of about 6% for iodine-131 in this geometry. The detector was surrounded by an 8 cm lead castle and the background in the counting bottle was checked frequently. Instrument stability was measured with a cobalt-60 check source. A total of about 10 000 counts was taken on each sample.

Mixing and sampling were continued from the injection time for $\frac{43}{4}$ d. The results show that mixing was not complete until $\frac{3}{2}$ h after injection. After this time the specific activity fell rapidly and exponentially for $\frac{3}{2}$ h (see Fig. 2). It then fell irregularly for a further 24 h before showing a slow exponential decline for the following 3½ d.

The counting rate of the samples was initially about 4000 counts/min and finally fell to 1000 counts/min. The background produced an additional count of 600 counts/min, and hence it was important to determine background accurately throughout the experiment. No contamination of the counting bottles arose from the samples.

The counter stability, checked with a cobalt-60 source, was within the statistics of counting and showed no measurable drift throughout the in-
investigation. Since the counting was continued for 5 d, an accurate half-life correction had to be made for the isotope, and regular measurements were made on one of the iodine standards to check the decay. The values fitted a half-life of 8.04 d within the statistics of counting and served as a check of both the radiochemical purity of the tracer and the stability of the detector.

Two standard dilutions of the parent iodine solution were made by weight with dilution factors of $5.90 \times 10^8$ and $9.27 \times 10^8$ to give counting rates similar to those expected from the pool. The two standard solutions actually counted several times as fast as the samples from the pool. Their counting rates were in agreement within 0.8% of each other, and an average value was taken for the calculation of the results.

**DISCUSSION OF THE SPECIFIC ACTIVITY MEASUREMENTS IN THE POOL**

The results of the specific activity measurements are shown in Table I and Fig. 2. They are plotted as a graph of the log counting rate against time. The sample counting rate is corrected for background and the decay of iodine with 8.04-d half-life. The time of high and low water in the pool is also shown in Fig. 2.

The interpretation of the results of a single investigation of this type is necessarily open to ambiguity. When the form of the curve in Fig. 2 is taken together with some of the known hydraulic characteristics of the pool, alternative explanations can be examined and a number of conclusions drawn concerning the characteristics of the movement of the water in the pool.
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(a) Pool volume

The samples measurements show that mixing of the pool was complete after 2\(\frac{1}{2}\) h (point A in Fig. 2). This is a longer period than was indicated by the immersed detector in the centre of the pool, but this detector averaged the specific activity over a much larger volume than the 2\(\frac{1}{2}\)-l sample and also, for reasons of safety, the latter was taken from near the edge of the pool.

At the time of complete mixing, the volume of the pool can be calculated from the specific activity of the pool compared with the diluted standard solutions. The volume of water into which the activity had mixed was 3075 m\(^3\). By extrapolation of the initial linear portion of the curve (section A to C) back to the time of injection, the volume of the pool into which mixing initially occurs (probably the visible water within the pool) is estimated to be 2700 m\(^3\).

If the initial part of the curve is assumed to indicate mixing into water in the vicinity of the pool (including tidal water) then the volume into which this mixing occurs can be estimated by extrapolation of the linear portion F-G of the curve back to zero. This eliminates the effect causing the slope F-G and indicates a value\^ of 4400 m\(^3\) for the volume of water into which the tracer has mixed after 30 h. This is probably the total volume of the pool plus the oscillating tidal water and adjacent pools with which water is exchanged.

(b) Tidal throughput and tracer dispersion

The initial dilution of the tracer (sections A to F) will be governed principally by the tidal characteristics of the pool and by the spreading of the tracer into adjacent water bodies.

The effect of a throughput of water of Q m\(^3\)/h in the constantly mixed pool of volume V m\(^3\) is that the concentration C of a tracer in the pool after time T hours is related to the initial concentration C\(_0\) by the expression

\[ C = C_0 \exp(-Q/V \times T). \]

An exponential relation of this form is found during the first tidal input (assumed to have a constant value Q since the accuracy of the measurement does not warrant a more detailed analysis). From the slope of the curve and with a value of V = 2700 m\(^3\), Q = 138 m\(^3\)/h or 830 m\(^3\) in a tidal half cycle of 6 h. This value is a maximum for the tidal throughput since it assumes that no reduction of specific activity occurs as an effect of dispersion of the tracer. However, this latter effect would not be expected to follow an exponential relationship, and the linearity of section A to B of Fig. 2 supports the theory that the tidal current is the primary control during this time.

Since the level of the pool rose 20 cm because of the incoming tide which required only 60 m\(^3\) of water, most of the 830 m\(^3\) of water passes through the pool to raise the water table "upstream" of the pool.

When the tide turns (point B), the tracer which had passed upstream might be expected to return, thus causing a reduction in the rate of dilution of the tracer. This only occurs for between 1 and 2 h, after which the specific activity of the pool continues to fall almost as rapidly as during the previous flood tide. This could be taken to support the tracer dispersion hypothesis.
and could result from the water's passing through an enclosed passage before being diluted into a fairly extensive water body upstream, from which it returns at very low specific activity. Alternatively, it could be due to the tidal water returning through the pool together with an additional flow of fresh water (see following section). Since the pool required up to 2½ h to mix near the edges, only a limited amount of tracer would pass "upstream" during the initial flood, and the larger volume of water passing through on the ebb because of the fresh water component would result in the observed dilution. The measurements cannot differentiate uniquely between these two possibilities.

When the tide has ceased to ebb (about point C), for the following 3½ h there is very little change of concentration, indicating that the first part of the returning flood tide consists of water which has previously been in the pool. The current direction investigation indicated that this water would be contained in the steep 4-m-square passage leading downwards from the south end of the pool, and this apparently causes only limited tracer dispersion.

As outlined in the previous section, the mechanisms of tracer dilution resulting from tidal throughput and pulse dispersion appear to be complete 30 h after injection (point F) when the tracer has been mixed with about 4400 m³ of water.

(c) Fresh water throughput

After 30 h the curve showed a slow exponential reduction of the specific activity. This relation persisted for at least the following 85 h. Two mechanisms can cause this effect.

The first is the possibility of adsorption of the tracer on the limestone. This is unlikely since the I⁻ ion is not subject to adsorption on limestone, although some effect could occur during extensive percolation of water of low carrier ion content. The presence of Cl⁻ and Br⁻ ions in the partially saline water, together with the added I⁻ carrier are considered sufficient to inhibit this effect.

The more probable mechanism is that of dilution by fresh water flow through the pool. Since the pool level is above that of the sea, it is not likely that a current from the sea could cause removal of the tracer from the system.

During the time interval F to G, measurements relate to the whole water system characterized by point H on Fig. 2. With the value of 4400 m³ as the volume of the system, the fresh water throughput is estimated to be 26 m³/h.

By a mathematical analysis of the linearity of the curve F to G it is found that a cyclical component is present. This is such that the period is that of the tide, and the specific activity tends to a maximum positive displacement from the average just before high water and a maximum negative displacement just before low water. The effect is small but statistically significant when examined over six tidal cycles. This effect indicates that the fresh water input into the system is on the upstream side of the pool. Thus, the flooding tide holds back the incoming fresh water, resulting in a slow fall of the specific activity during the flood, and the ebb carries the fresh water into the system so that the specific activity reduces more rapidly.
This result is of considerable importance if any engineering is carried out in the pool with the object of restricting the access of saline sea water on the downstream side of the system. Such a restriction would not be expected to cut off the fresh water.

CONCLUSIONS

A study of the dispersion and dilution of radioactive iodine-131 mixed with the water of an underground pool enabled quantitative values of several hydraulic parameters of the pool to be obtained.

By accurate measurements of the specific activity in the constantly mixed pool for a period of 5 days, several effects which cause reduction of the specific activity can be seen and examined separately. The volume of the pool can be determined and also the volume of water in the whole pool system including the oscillating tidal water and the adjacent water bodies into which the tracer disperses. The volume of water entering the pool during the flood tide can be determined, but not without some ambiguity since the effect cannot be uniquely separated from that of tracer dispersion. Finally, the throughput of fresh water and its position of entry into the pool system relative to the movement of tidal water can be determined from the rate of reduction of specific activity after the other effects mentioned above have reached equilibrium. The value for fresh water throughput obtained in this way is a maximum, because some additional tracer dispersion will occur during these measurements. However, the exponential nature of the relationship between specific activity and time indicates that the effect is caused by water throughput.

A single investigation of this nature cannot hope to differentiate among all the causes of reduction of specific activity in the pool. Confirmation of the interpretation outlined in this paper could be obtained by further experiments. In particular, an investigation starting at high tide would examine the dispersion on a falling tide, which would make an interesting comparison with that found on the flood; and the two results should assist in the differentiation between tidal throughput and tracer dispersion. The conclusion on the volume of fresh water throughput could be examined by an investigation carried out in late summer when a long period of dry weather would reduce the fresh water flow to a negligible value.

Although this investigation has yielded some information about the hydraulic behaviour of the underground pool, more valuable definite data could be obtained by a series of several investigations under different hydraulic conditions.

DISCUSSION

B. R. PAYNE: The authors are to be congratulated on what is, I think, a novel application of radioisotopes. There is, however, one question I should like to ask: have you given any thought to the possibility of iodine-131 being lost in the form of iodine? The difference you mentioned between your own pool-volume data and the estimates of the engineers could be indicative of such losses. I would have thought that the turbulent air-mixing might have been of some importance in this connection.
D. B. SMITH: Since the maximum possible rate of iodine loss is represented by the final exponential slope of the concentration-time graph, any error arising from iodine loss during the first few hours of the investigation would be negligible and could not account for the difference between the engineers' estimate of the pool iodine and the isotope estimate. Incidentally, the engineers' estimate did not include water outside the immediately visible volume of the pool; such water would of course be included in the isotope measurement.

It is possible that the air mixing affected the iodine during the investigation but the addition of the carrier would normally reduce this effect. Only further investigations, however, could clear up this point satisfactorily.

R. AMBROGGI: It might perhaps have been useful to have checked your results with a microcurrent meter. This would have given you information on the saline and fresh-water influx areas.

D. B. SMITH: Microcurrent measurements would possibly have been valuable as an additional aid, especially as this would have enabled us to examine smaller entrance and exit points than was possible with the large-diameter equipment we used. On the other hand the turbulence of the water in the pool was such as to make it difficult to obtain very good measurements. The direction of the currents was variable and I don't think that the entrance and exit points would have been very easy to locate. Results with the spider were not particularly good. We had expected that the radioactivity would be carried from the centre past one counter only, but in fact it was very often registered on all six. So you see, there was quite a lot of turbulence in the pool.

P. C. LÉVÉQUE (Chairman): You could perhaps have made use of the techniques employed by Moser or Guizerix for determining the velocity and the direction of water currents.

D. B. SMITH: I think that this would indeed have been an improvement on the method we used. In any case, we shall certainly not use the spider again. I think that Moser's technique might have given us the information we needed more easily.

P. C. LÉVÉQUE (Chairman): On the whole I think that the complexity of the lines of current flow would make it very difficult to use a microcurrent meter. There would always be a danger of the vane turning at negative speeds. Karstic-type holes are too irregular to enable even high-precision mechanical devices to be used for measuring the velocity and thus the flow rate of currents. Moreover, it is very likely that there would be a difference between ebb and flow measurement results.

J. HONSTEAD: I gather that the air-injection mixing device was kept in operation continuously during the experiment. I wonder whether it might not have been possible, by switching off the mixer, to have studied the appearance of concentration gradients. This might have provided information on the source and rate of inflow of fresh water.

D. B. SMITH: An investigation of this sort might well yield interesting results. It would not, however, be possible to combine it with the experiment described in our paper since turbulence would continue long after the air mixing was stopped. Since the investigation would be concerned with
changes in small volumes of water, it might be possible to obtain higher resolution by using a pure beta-emitting isotope and scanning possible entrance and exit points with a beta counter.
A STUDY IN RIVER ENGINEERING BASED ON THE RESULTS OF FIELD MEASUREMENTS OF FLOW VELOCITIES WITH RADIOISOTOPES IN THE SORACHI RIVER, JAPAN

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AND
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Abstract — Résumé — Аннотация — Resumen

A STUDY IN RIVER ENGINEERING BASED ON THE RESULTS OF FIELD MEASUREMENTS OF FLOW VELOCITIES WITH RADIOISOTOPES IN THE SORACHI RIVER, JAPAN. In a joint project of the Hokkaido Development Bureau and the research group at the University of Tokyo, radioactive tracer experiments were made in the summer of 1961 on the Sorachi River from the Kanayama Dam site to the Akabira intake, ranging 90 km. The objective of the experiments was to obtain precise information on the flow times to each intake and the flow conditions of the water released from the dam, which is now under construction and is to be used for various purposes.

A radioactivity of about 60 mc of sodium-24 was introduced into the stream in each reach tested. The lower reaches from the dam were divided into five reaches, each of which was about 15 to 20 km in length, and the tests were carried out five times in turn from the lowest reach. The test procedure consisted in the direct measurement by underwater detector and in the periodic collection of one-liter samples. In the case of the former, detecting efficiency for sodium-24 was $29 \times 10^8$ counts/cm$^3$/per min/μc with a 1 3/4-in diameter x 1-in NaI crystal. In the case of the latter, sodium-24 was caught by an ion-exchange column in a recovery of 95%. The results by the two detection methods were figured as concentration versus time flow-through curves, and the peaks of both curves coincided very well. With the aid of such a procedure, the maximum distance traced with 60 mc was found to be about 30 km.

Then, based on the results obtained, a study from the standpoint of river engineering was made. That is, the authors calculated the flow time of released water for irrigation, checking the reliability of the flow time estimated by velocity measured with the current-meter method at a representative discharge observation station in each longitudinal flow section and by the flow time actually measured through fairly long reaches of each section, using radioisotopes. Next, they calculated the longitudinal diffusion coefficients of tracers in the Sorachi River, finding that each longitudinal diffusion of tracers correlates to the sectional equivalent roughness of the river in each longitudinal section of the stream.

ÉTUDE D'HYDRAULIQUE FLUVIALE FONDÉE SUR LES RÉSULTATS DES MESURES, A L'AIDE DE RADIO-ISOTOPOS, DE LA VITESSE DES EAUX DU SORACHI AU JAPON. Dans le cadre d'un projet conjoint, le Bureau d'aménagement de l'Ile d'Hokkaido et le Groupe de recherches de l'Université de Tokyo ont procédé, au cours de l'été 1961, à des expériences à l'aide de radioindicateurs sur la partie du cours du Sorachi qui va du barrage de Kanayama à la prise d'eau d'Akabira et s'étend sur 90 km. Ces expériences avaient pour objet de réunir des renseignements précis sur le temps mis par l'eau pour atteindre chaque prise d'eau et sur le régime d'écoulement de l'eau depuis la sortie du barrage à fins multiples qui est actuellement en construction.

Dans la partie du cours d'eau qui a fait l'objet des expériences, on a délimité cinq sections, d'une longueur d'environ 15 à 20 km chacune; on a introduit environ 60 mc de sodium-24 dans chacune d'elles, en commençant par la plus en aval. On a employé deux méthodes de détection: mesure directe de la radioactivité à l'aide de détecteurs immergés et prélèvements réguliers d'échantillons d'un litre. Avec la première méthode, l'efficacité de détection de sodium-24 a été de $29 \times 10^8$ coups cm$^{-3}$/min/μc avec un cristal de NaI de 4,5 cm x 2,5 cm. Pour la seconde, le sodium-24 a été retenu par une colonne échangeuse d'ions jusqu'à concurrence de 95%. A l'aide des résultats obtenus par ces deux méthodes, on a établi deux courbes du débit
indiquant la concentration en fonction du temps; pour le deux courbes, les pics coïncidaient parfaitement. On a ainsi constaté que la distance maximum qui pouvait être étudiée avec 60 mc était d'environ 30 km.

Se fondant sur les résultats obtenus, les auteurs présentent ensuite une étude d'hydraulique fluviale. En d'autres termes, ils ont calculé le temps d'écoulement de l'eau destinée à l'irrigation, en comparant, aux fins de contrôle, les valeurs du temps estimées d'après la vitesse (méthode du moulinet) en des points d'observation représentatifs des différentes sections longitudinales du cours d'eau et les valeurs du même temps, mesurées à l'aide de radioisotopes, sur une assez longue partie de chacune de ces sections. Ils ont ensuite calculé les coefficients de diffusion des indicateurs dans le Sorachi et constaté que, pour chaque section considérée, il existe une corrélation entre la diffusion et la rugosité du lit.

**INGENIERÍA FLUVIAL: ESTUDIO BASADO EN LA MEDICIÓN DE VELOCIDADES DE FLUJO MEDIANTE RADIOISÓTOPOS EN EL RÍO SORACHI (JAPÓN).**

En el verano de 1961, el Hokkaido Development Bureau y el grupo de investigadores de la Universidad de Tokio realizaron conjuntamente experimentos con indicadores en el río Sorachi, desde la presa de Kanayama hasta la salida de Akabira (90 km). Se trataba de obtener datos precisos sobre las velocidades de circulación en cada salida y las condiciones en que fluye el agua de la presa actualmente en construcción y que se destinará a diversos fines.

En cada uno de los tramos estudiados, se introdujeron en la corriente unos 60 mc de sodio-24. El curso del río se dividió, a partir de la presa, en cinco tramos de unos 15 a 20 km, en cada uno de los cuales se realizó un ensayo comenzando en el tramo inferior. Las mediciones se efectuaron directamente con un detector sumergible, tomando periódicamente muestras de 1 l. En el primer caso, se utilizó un cristal de NaI de 1.75 pulg de diámetro y 1 pulg de altura, cuya eficiencia de recuento para el sodio-24 era de 29·10^6 impulsos/cm²·min·mc. En el segundo, se recuperó el 99% del sodio-24 mediante una columna de intercambio iónico. Con los resultados obtenidos al aplicar estos dos métodos, se trazaron curvas de la concentración en función de la velocidad de flujo y se observó que los máximos coinciden satisfactoriamente en ambos casos. Los autores hallaron que la distancia máxima que se puede estudiar con 60 mc es de unos 30 km.

A continuación, examinaron los resultados desde el punto de vista de la ingeniería fluvial, es decir, calcularon la velocidad de circulación del agua destinada al riego a partir de la velocidad determinada con el aparato medidor de corriente en un punto de observación en cada tramo y la compararon con la velocidad...
1. INTRODUCTION

The Sorachi River is 166.2 km in length and drains an area of 2663 km\(^2\); it is the largest tributary of the Ishikari River, which is the second largest river in Japan. Upon the upper reaches of this river, the Kanayama Dam Reservoir, to be used for many purposes such as flood control, irrigation and hydroelectricity, is now under construction. Irrigation water supplied from the Kanayama Dam Reservoir flows 90 km downstream, meandering in the Sorachi River channel until reaching the farthest intake (Fig. 1). Therefore, an investigation of the flow conditions of the water is quite important for control of the future operation of the dam.

![Map of the Sorachi River](image)

This investigation was carried out by tracer experiments with saltwater and the radioisotopes I\(^{131}\) and Na\(^{24}\) in every August, a season of water shortage, for four years. It was found that with salt water the maximum tracing distance at a time was about 4 km at the most, while it was as long as 30 km with radioactive tracers. Thus the latter method offers much clearer data than does the former. In the radioactive-tracer experiments, direct measurement making use of the underwater detector and, parallel to it, the periodic collection of samples for determination of the absolute concentration in the water were adopted.

In this paper first several matters in connection with radioactive-tracer techniques and then methods of estimation of the flow times to the respective intakes will be described. Further, the calculation of the longitudinal diffusion coefficient in each reach and some factors affecting this coefficient will be discussed.
2. RADIOACTIVE-TRACER TECHNIQUES

In the first radioactive-tracer experiment, carried out as a pilot experiment in 1960, $^{131}$I of about 1.6 c was used, for short-lived radioisotopes had not been produced in this country in large quantities at that time. In the second experiment in 1961, however, $^{24}$Na of about 750 mc was used as a principal tracer and $^{131}$I of about 600 mc as an auxiliary. Below, mainly the techniques and methods used in the second experiment will be described.

$^{24}$Na of about 150 mc was produced by the JRR-1 reactor, transported by air, prepared in the form of a tracer and then injected into the river after a decay time of about 18 h. Such procedures were carried out five times for nine successive days. $^{131}$I was provided for use when $^{24}$Na was not available because there was no airplane flight or no reactor run.

There were some limits to the use of larger amounts of $^{24}$Na than that mentioned above, including the small production capacity, high cost of transportation by air, poor facilities in the field, etc. Consequently, radioactivity strength at the time of injection was about 70 mc. The maximum distance for which water flow can be traced with this amount was estimated to be about 20 km, when account is also taken of other factors such as sensitivity of detection, recovery of the tracer, discharge and dispersion coefficient of the river. Then, it was finally decided to divide the whole reach of about 80 km to be tested into five reaches.

In order to carry out the experiment safely the following precautions and procedures are necessary: When there is an intake in the reach to be tested, the radioactivity of the flowing body of water must not exceed 0.1 of the maximum permissible concentration at that point. Accordingly, the distance between the injection point and the intake in which $^{24}$Na of 70 mc dilutes to 0.1 MPC was calculated with the longitudinal diffusion coefficient, $1.8 \times 10^5$ cm$^2$/s, which was obtained in another experiment [1] on this river. In addition, as the calculation was made on the assumption that the tracer was injected uniformly in the cross-section, the calculated values were multiplied by a safety factor of 2 or 3. Thus, the injection points had to be located farther than the distances upstream from the intake in the respective reaches shown in Table I.

Before the experiment, some samples of the river water were taken and analysed (Table II) and throughout the reach of the river the backgrounds under and over the water surface were surveyed. These results showed no unusual values.

According to the air transportation regulations concerning radioactive materials, solutions and powders must be packed tight in metallic cans. $^{131}$I could be prepared to tracer form before being sent. $^{131}$I mixed with sufficient carrier material was enclosed in four glass ampoules, each of which contained a NaI solution of 50 ml and 150 mc, respectively; and the ampoules were then transported by air in package form, the four ampoules being packed in a steel can which was further packed in a leaden container of 80 kg. The total activity was 600 mc.

With $^{24}$Na the energy of $\gamma$-rays is very high, and thus the weight of the leaden container is excessive. So as not to exceed the weight limit of 100 kg for air transportation, $^{24}$Na had to be transported in powder form, which was
TABLE I

DISTANCE FOR DILUTION TO 0.1 MPC

<table>
<thead>
<tr>
<th>Reach</th>
<th>Discharge (m³/s)</th>
<th>Dilution factor</th>
<th>Distance to be diluted to 0.1 MPC (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kanayama-Tonashibetsu River junction</td>
<td>24</td>
<td>0.0103</td>
<td>1100</td>
</tr>
<tr>
<td>Tonashibetsu River junction-Shimanoshita</td>
<td>47</td>
<td>0.0202</td>
<td>600</td>
</tr>
<tr>
<td>Shimanoshita-Nokanan</td>
<td>65</td>
<td>0.0279</td>
<td>300</td>
</tr>
<tr>
<td>Ashibetsu power station-Akabira intake</td>
<td>58</td>
<td>0.0248</td>
<td>300</td>
</tr>
</tbody>
</table>

not tracer form. Five pieces of a sodium target were irradiated in the through tube of JRR-1 reactor for 2 h a day; each of these consisted of 10 g of powdery Na₂CO₃ enclosed in water soluble PVA (polyvinylalcohol) bag and packed in an aluminium can (25-mm diam. X 28 mm). After irradiation, the radioactivity of each target was about 30 mc. The five containers were transported via air to the site of the experiment in Hokkaido, a distance of about 1000 km.

A temporary radiochemical laboratory attached to our base camp was built for the preparation of the tracer, chemical analysis and radioactivity measurement of sampled water and storage of radioisotopes, etc. When the containers arrived at the base camp, the dose rate of the isotopes enclosed in the aluminium cans was checked with an ion-chamber dosimeter. Then, in this laboratory the tops of the aluminium cans were opened by a cutter made for this purpose, and the Na₂CO₃ enclosed in the PVA bags was removed and dissolved in HNO₃ solution, through a chemical reaction changing to NaNO₃, which is much higher in solubility in water than is Na₂CO₃. When the reaction ended after several minutes, the radioactive tracer as a solution of 200 ml was finally made up and promptly transferred to a glass flask (250 ml) specially made for injection and transported by car to the injection place.

In the tracer injection process (Fig.2) the flask in the chamber is crushed under water, and the radioactive solution flows away through the many holes in the wall. For easy operation in water, a float is attached to the injector. After the radioactive tracer is introduced, the injector is carried to the centre of river, set afloat and operated from a rubber boat.
**TABLE II**
CHEMICAL ANALYSIS OF WATER ALONG THE SORACHI RIVER

<table>
<thead>
<tr>
<th>Items of analysis</th>
<th>Kanayama</th>
<th>Ponmojiri</th>
<th>Ashibetsu</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>7.5</td>
<td>7.4</td>
<td>7.3</td>
</tr>
<tr>
<td>Total acid</td>
<td>0.05 epm</td>
<td>0.07 epm</td>
<td>0.06 epm</td>
</tr>
<tr>
<td></td>
<td>2.4 ppm</td>
<td>3.4 ppm</td>
<td>3.0 ppm</td>
</tr>
<tr>
<td>Alkalinity</td>
<td>0.34</td>
<td>0.40</td>
<td>0.42</td>
</tr>
<tr>
<td></td>
<td>17</td>
<td>20</td>
<td>21</td>
</tr>
<tr>
<td>Evaporation residue</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>51</td>
<td>99</td>
<td>91</td>
</tr>
<tr>
<td>Combustion residue</td>
<td>-</td>
<td>79</td>
<td>62</td>
</tr>
<tr>
<td>Hardness</td>
<td>0.38</td>
<td>0.57</td>
<td>0.59</td>
</tr>
<tr>
<td></td>
<td>19</td>
<td>28.4</td>
<td>29.4</td>
</tr>
<tr>
<td>Cl'</td>
<td>0.044</td>
<td>0.11</td>
<td>0.035</td>
</tr>
<tr>
<td></td>
<td>1.6</td>
<td>3.8</td>
<td>3.0</td>
</tr>
<tr>
<td>SO₄²⁻</td>
<td></td>
<td>17</td>
<td></td>
</tr>
<tr>
<td></td>
<td>18</td>
<td></td>
<td>22</td>
</tr>
<tr>
<td>NO₃⁻</td>
<td></td>
<td></td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>0.02</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>C.O.D.</td>
<td>0.46</td>
<td>0.35</td>
<td>0.38</td>
</tr>
<tr>
<td></td>
<td>3.7</td>
<td>2.8</td>
<td>3.0</td>
</tr>
<tr>
<td>Cu</td>
<td>0.020</td>
<td>0.020</td>
<td>0.022</td>
</tr>
</tbody>
</table>

* epm = mg equivalent/1
For detection of radioactivity dispersed in water, direct measurement by the underwater detector and periodic collection of 1-l samples were used simultaneously. In the pilot experiment a G-M type underwater detector, which contains a bundle of three large G-M tubes to increase detecting efficiency, was fabricated. However, as its counting rate from the background in water was large, two scintillation detectors with 1\text{\textfrac{4}{10}}\text{-in diam.} \times 1\text{-in NaI} crystals were used in the second experiment. The detecting efficiencies of these detectors were measured with a water basin (1.5 \times 1.5 \times 1.5 \text{m}) in which radioisotopes had been dispersed uniformly. These basic experiments were carried out with the radioisotopes \text{I}^{131}, \text{Na}^{24} and \text{Br}^{82}. The efficiency was indicated by the ratio of the counting rate to the concentration of radiotracer in water, as tabulated in Table III. This shows that the scintillation detector is more useful than is the G-M detector. Therefore, in the field experiment, the G-M detector was set at the first observation station and the scintillation
<table>
<thead>
<tr>
<th>Radioisotope</th>
<th>Type of detector</th>
<th>Detecting efficiency (cpm/μc per cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na²⁴</td>
<td>1 3/4-in diam. X 1-in NaI**</td>
<td>29 x 10⁶</td>
</tr>
<tr>
<td></td>
<td>1-in diam. X 1-in NaI</td>
<td>6 x 10⁴</td>
</tr>
<tr>
<td></td>
<td>G-M tube* X 3</td>
<td>2.5 x 10⁴</td>
</tr>
<tr>
<td>I¹³¹</td>
<td>1 3/4-in diam. X 1-in NaI**</td>
<td>9 x 10⁴</td>
</tr>
<tr>
<td></td>
<td>1-in diam. X 1-in NaI</td>
<td>1.4 x 10⁴</td>
</tr>
<tr>
<td></td>
<td>G-M tube* X 3</td>
<td>0.3 x 10⁴</td>
</tr>
<tr>
<td>Br⁴²</td>
<td>1 3/4-in diam. X 1-in NaI</td>
<td>36.6 x 10⁴</td>
</tr>
</tbody>
</table>

* Halogen G-M tube (made in Japan): effective volume, 50X330 mm; effective surface area, 518 cm²; BG in water, 9 cps.

** BG in water, 5-6 cps.

detectors at the second and third stations. Detecting pulses were fed to a rate-meter via a 50-m coaxial cable and recorded. Then, through such procedures as conversion of the counting rate to concentration with detecting efficiency and correction for nuclear decay after injection, flow curves were calculated; an example is shown in Fig. 3.

In the sampling method more than 1 l of water was collected by the water sampler near the underwater detector. After rough filtration, exactly 1 l of sample water was taken. Analysis of I¹³¹ was performed by the following method: Add 7-8 ml of concentrated H₂SO₄ to adjust the pH to 0.5-1.0. Add 1 ml of 0.1 N NaI and 1.2 ml of 0.1 N AgNO₃ to precipitate the AgI. Stir the sample violently, and heat slowly for 10-15 min. Filtrate AgI with a special filter. Lay the precipitate on a counting tray, dry under a heat lamp and count its beta activity.

By this method 85% of the I¹³¹ was recovered. Other methods were also tested. These yields are tabulated in Table IV.

Analysis of Na²⁴ was performed as follows: Add 0.2 ml of 1 N NaCl and stir slowly. Pour the sample in the upper basin, and adjust its passage through the ion-exchange column to a rate of 18-20 ml/min. Extract the collected Na ion with 30 ml of 3 N HCl solution at 2 ml/min.

Then, by means of the conventional process, a sample for beta counting was prepared and measured. The recovery of Na²⁴ by this method was 95%, and over-all efficiency including counting efficiency amounted to 23%. Another method was also tested, but it resulted in less recovery than that of the ion-exchange method (Table IV).

Conversion of the observed beta-counting rates to disintegration rates, based upon the calibrated geometrical efficiency of the counting apparatus,
for Na\textsuperscript{24} and upon comparison with a standard source for I\textsuperscript{131}, was made. Further, radioactivity concentrations of these samples were corrected to that at the time of injection and were plotted, as shown in Fig. 3.

Both flow curves obtained by these different methods coincided very well in peak time, and the discrepancy in concentration resulted from various error factors such as the variation of geometry at the point at which the underwater detector was set in the river, the difference of position between sampling and setting points, experimental errors in chemical treatment, etc. Finally, for calculation of the mean flow velocity the peak-to-peak times were used.
As for precautions in the use of radioisotopes, the radioactive tracer experiment was carried out safely. The exposure dose of each person who worked on the experiment for 10 d was less than 180 mR; two were above 100 mR; one, 30 mR; and a few others, less than 10 mR. Two days after the experiment ended, a survey of the background in the river through the whole reach tested was carried out, and there was no contamination.

3. CALCULATION OF FLOW TIME

Generally speaking, the methods for measuring the flow velocity in river channels may be classified roughly into two kinds. One is estimation from the data actually observed at some representative stations in a reach. The other is calculation of the mean flow velocity within a reach from the result of tracer experiments.

The representative stations were set at intervals of about 5 km. On the assumption that most of the tracer flows down the centre of the river, the relation between mean maximum velocities observed by current meters at two or three representative stations and the flow velocity obtained by tracer experiments were examined. As a result, it was clear that the maximum velocities vary appreciably at each station, but the flow velocity does not change relatively. Accordingly, it seems that the first method is not accurate.

The tracer experiment with salt water was carried out in 1958 and 1959. At the same time, some plywood (15 cm × 15 cm) was floated as an auxiliary measure. The ratio of mean flow velocity \( V_m \) of the salt water to the surface velocity \( V_s \) of the floats tends to increase with flow distance.

Further, comparison was made between the results with radioactive tracers and those with salt water. When the term "peak duration" is defined as shown in Fig. 4, the measurement accuracy may be indicated by the ratio of the peak duration to the flow time. Such measurement accuracies corresponding to the respective flow distances were plotted (Fig. 5). From this figure it was found that the accuracy for a distance of 10 km by radiotracers is nearly equal to that for 2 km by salt water.

The calculations of the flow time from the dam to each intake was based on the results traced for 80 km from the Kanayama Dam site to the Akabira intake in 1958 to 1961. With the mean velocity formula, \( U = (1/n)R^{1/m} \), and the mean sectional area \( A \), \( A = aH^2 \) through a reach, the relation between discharge \( Q \) and flow velocity \( U \) is shown as follows:

\[
Q = AU = a(n/1^m)^{5/8} U^{5/6} = KU^{6/7},
\]

where \( R = \) average hydraulic radius, \( l = \) average gradient of river, \( n = \) average river roughness, \( l, m = \) constant index, \( H = \) depth of river, nearly equal to \( R \), \( S = \) index relating to the shape of the cross-section, and \( a = \) specific constant.

\[
K = a(n/1^m)^{5/8}, \quad \beta = s/l + 1.
\]
Measurement accuracy $t'/t_0$

- $a$ - value:
  - 0.017 mol/m$^3$ for salt water tracer
  - 0.1 µCi/m$^3$ for isotope tracer

Fig. 4

Comparison between the accuracy of measurement of the two tracers, salt water and radioisotope
- Salt water, NaCl
- Radioisotope

Fig. 5

Calculated as the gradient of the Q-U line plotted on log-log paper. Thus, the relation of discharge to flow velocity in each reach was computed as shown in Table V.

Water from the dam generally passes representative discharge observation stations, which are called operating stations. Therefore, it is necessary to study whether the discharge observed at these stations correlates with the flow time. In this work the following method was adopted as it seemed most reasonable: First, we calculated the ratio of the discharge obtained by tracer experiments to that obtained by water gauges at the operating stations.
<table>
<thead>
<tr>
<th>Reaches</th>
<th>Relation between flow velocities and discharge in each reach</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kanayama Dam Site—the confluence of the Tonashibetsu River</td>
<td>$Q = 35.6 V^{1.980}$</td>
</tr>
<tr>
<td>The confluence of the Tonashibetsu River—Yamabe bridge</td>
<td>$Q = 42.0 V^{2.060}$</td>
</tr>
<tr>
<td>Yamabe bridge—Nunobe bridge</td>
<td>$Q = 32.0 V^{2.660}$</td>
</tr>
<tr>
<td>Yamabe bridge—Furano Intake</td>
<td>$Q = 28.3 V^{2.120}$</td>
</tr>
<tr>
<td>Nunobe bridge—Sorachi bridge</td>
<td>$Q = 34.0 V^{2.550}$</td>
</tr>
<tr>
<td>Sorachi bridge—Ochibe</td>
<td>$Q = 46.0 V^{2.450}$</td>
</tr>
<tr>
<td>Ochibe—Takisato bridge</td>
<td>$Q = 36.0 V^{4.060}$</td>
</tr>
<tr>
<td>Takisato bridge—Ponmojiri hydraulic power station</td>
<td>$Q = 48.0 V^{2.700}$</td>
</tr>
<tr>
<td>Takisato bridge—Nokanan bridge</td>
<td>$Q = 82.0 V^{2.500}$</td>
</tr>
<tr>
<td>Ashibetsu hydraulic power station—Heihan bridge</td>
<td>$Q = 178.0 V^{2.300}$</td>
</tr>
<tr>
<td>Heihan bridge—Oketo bridge</td>
<td>$Q = 94.0 V^{2.800}$</td>
</tr>
<tr>
<td>Oketo bridge—Akama bridge</td>
<td>$Q = 150.0 V^{2.900}$</td>
</tr>
<tr>
<td>Akama bridge—Akabira Intake</td>
<td>$Q = 290.0 V^{4.370}$</td>
</tr>
</tbody>
</table>
ating stations, observed at the same time, and then estimated the discharge through each reach in accordance with the various discharges which had been observed at operating stations.

Then, we calculated the respective flow times, using the values in Table V, and summed them up in separate reaches from the dam site to an intake. In such a manner Fig. 6 was obtained.

![Flow Time to Each Intake](Image)

Flow time to each intake plotted against respective discharges at the operating stations

**4. SOME THEORETICAL CONSIDERATIONS**

*(a) Diffusion coefficient*

Generally speaking, the diffusion equation of a tracer is as follows:

\[
\frac{\partial C}{\partial t} + \frac{\partial (uC)}{\partial x} + \frac{\partial (vC)}{\partial y} + \frac{\partial (wC)}{\partial z} = \frac{\partial}{\partial x} \left( D_x \frac{\partial C}{\partial x} \right) + \frac{\partial}{\partial y} \left( D_y \frac{\partial C}{\partial y} \right) + \frac{\partial}{\partial z} \left( D_z \frac{\partial C}{\partial z} \right) + W_0 \frac{\delta C}{\delta z} - \alpha C, \tag{3}
\]

where \( C \) = concentration of a tracer; \( u, v, w \) and \( D_x, D_y, D_z \) = velocity components and diffusion coefficient of \( x, y \) and \( z \) directions, respectively; \( W_0 \) = precipitating velocity of tracers; and \( x, y, z \) axes are taken in the flow, cross and depth directions, respectively.
Now, neglecting the change of density, precipitation and decay of tracer in the y and z directions, and taking the origin of the co-ordinates at the centre of the flowing tracer, we may simplify this as follows:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2},$$

(4)

where $x = \text{longitudinal distance from flowing origin}$, and $D = \text{diffusion coefficient in the x direction}$. However, this symbol $D$ represents exactly the apparent coefficient, because we neglected the self-purification decay.

Assuming that the tracer is injected into the river at a very small width, the following solution can be obtained:

$$C(x, t) = \left[\frac{ug}{2Q_0} (\pi Dt_0)^{1/2}\right] \exp\left(-\frac{u^2 t'^2}{4Dt}\right),$$

(5)

where $g = \text{quantity of tracer injected into the river}$, $Q_0 = \text{discharge at } t_0 = 0$, $t_0 = \text{mean flow time}$, and $t' = t + t_0$.

Then, assuming that $C = C_{\text{max}}$ at $t' = 0$ ($t = t_0$), the longitudinal diffusion coefficient is as follows:

$$D = \frac{Uy}{4Q_0^2 C_{\text{max}}^2 t_0 \pi},$$

(6)

where $C_{\text{max}} = \text{maximum concentration of observed tracer}$.

By substitution of the data from the radiotracer experiment into equation (6), the values shown in Table VI were obtained.

For the cross-sectional diffusion, only one radioactive tracer experiment was carried out in the reach from the Sorachi Bridge to the Shimanoshita operating station, where two detectors were set, one in the centre of the river and the other near the river bank. The results show that the maximum concentrations at these two points are different from each other, but the beginnings and the ends of the flow curves are similar. So, it seems that a flow distance of about 5 km is not enough to diffuse the tracer in the cross-sectional direction.

The diffusion in the cross-sectional direction at other stations was studied as follows: The total quantity of detected tracer is given by

$$g = \sum_{i=1}^{n} Q_i \int_{0}^{\infty} C \, dt,$$

(7)

where $Q_i = \text{discharge at the zone assumed to have an equal velocity in the cross-section}$, $C = \text{concentration of a tracer}$, and $t = \text{measuring time}$.

With an initial quantity $g'$ of the tracer injected, the ratio of $g$ to $g'$ versus flow distance is shown in Fig. 7. In this experiment, when the flow distance is shorter than about 10 km, $g/g' > 1$. Therefore, it seems that most of the tracer tends to flow along the centre of the river, and its diffusion in the cross-sectional direction is insufficient.

(b) On some factors affecting diffusion:

The longitudinal diffusion coefficient is given by equation (6). From the equation, the dilution factor of the tracer, which is also important in
### TABLE VI

**CALCULATION OF LONGITUDINAL DIFFUSION COEFFICIENT D**

| Tracer point       | Observation point | \( u \) (cm/s) | \( g \) (μc) | \( C_{max} \times 10^6 \) (μc/cm\(^2\)X10\(^-6\)) | \( Q_x \) (cm\(^3\)/s X10\(^6\)) | \( \tau_c \) (s) | \( \frac{u^2}{g} \) X10\(^6\) | \( \frac{\Delta C_{max}}{Q_x} \) X10\(^6\) | \( D \) (cm/s X10\(^-6\)) |
|--------------------|------------------|----------------|-------------|----------------------------------|---------------------------------|----------------|----------------|----------------------------|----------------|----------------|
| Meiho bridge       | Akabira intake   | 58.2           | 47,000      | 0.99                             | 53.39                           | 4250           | 0.549          | 1.48                      | 0.37            |
| Akama bridge       | Meiho bridge     | 76.1           | 28,000      | 0.56                             | 53.40                           | 4880           | 0.454          | 0.54                      | 0.86            |
| Ashibetsu hydraulic power station | Helian bridge | 75.8           | 54,000      | 0.66                             | 50.00                           | 6450           | 1.675          | 0.89                      | 1.88            |
|                     | Oketo bridge     | 71.1           | -           | 0.32                             | 51.78                           | 12450          | 1.474          | 0.42                      | 3.51            |
|                     | Akama bridge     | 67.1           | -           | 0.22                             | 51.78                           | 18810          | 1.313          | 0.31                      | 4.24            |
| Shimano-shita      | Ochibe           | 65.4           | 59,000      | 1.35                             | 37.30                           | 3900           | 1.484          | 1.24                      | 1.20            |
|                     | Tatsuto bridge   | 97.4           | -           | 0.63                             | 37.77                           | 10320          | 3.302          | 0.73                      | 4.59            |
|                     | Nokanan bridge   | 73.8           | -           | 0.33                             | 40.94                           | 20580          | 1.895          | 0.50                      | 3.79            |
| Sorachi bridge     | Shimanoshita     | 83.2           | 65,000      | 1.75                             | 24.31                           | 6214           | 2.925          | 1.42                      | 2.06            |
|                     | Sorachi bridge   | 83.7           | 35,000      | 0.58                             | 27.78                           | 8940           | 0.838          | 0.27                      | 3.12            |
| Yamabe bridge      | Heiwa bridge     | 79.2           | 68,000      | 1.79                             | 25.27                           | 6000           | 2.906          | 1.66                      | 1.86            |
|                     | Nunobe bridge    | 82.4           | -           | 0.76                             | 27.45                           | 13320          | 2.723          | 0.76                      | 3.58            |
|                     | Sorachi bridge   | 82.5           | -           | 0.43                             | 27.78                           | 21300          | 2.731          | 0.37                      | 7.38            |
| Uryu hydraulic power station | Shimei bridge | 68.8           | 52,000      | 2.62                             | 17.97                           | 4830           | 1.286          | 1.35                      | 0.95            |
|                     | Shimokanayama    | 72.5           | -           | 1.16                             | 15.46                           | 11490          | 1.421          | 0.48                      | 2.99            |
|                     | Yamabe bridge    | 73.7           | -           | 0.78                             | 24.23                           | 16230          | 1.469          | 0.74                      | 1.99            |
|                     | Sorachi bridge   | 87.8           | -           | 0.24                             | 27.78                           | 38150          | 1.679          | 0.24                      | 6.99            |
sanitary engineering, is obtained as follows:

$$\frac{C_{\text{max}}}{C_0} = \frac{6}{(4\pi t_0 D)^{\frac{1}{4}}}$$

(8)

where $C_{\text{max}}$ = maximum concentration of observed tracer, $C_0$ = initial concentration just after injection, and $\delta$ = initial width of the tracer in the cross-sectional direction.

Assuming that the tracer is injected into river in an unit width, the dilution factor of the tracer may be given as follows:

$$\frac{C_{\text{max}}}{C_0} = \frac{1}{2(\pi t_0 D)^{\frac{1}{4}}}$$

(9)

From this equation, the dilution factor of the tracer is influenced only by the diffusion coefficient if the flow time remains constant. So we considered the factors affecting the diffusion of the tracer.

Generally, we may enumerate the following factors: irregularities in cross-sectional and longitudinal directions of the river channel, channel separation, depth of water, velocities and river roughness through the reach, etc.

Irregularities in cross-sections of the river are considered to increase the longitudinal and cross-sectional diffusions. As mentioned previously, with equations (1) and (2) the index of the shape of the cross-section $S$ can be calculated from the following equation:

$$S = \ell (\beta - 1).$$

(10)

As the value of $\beta$ is given by Table V, and with $2/3$ taken as the value of $\ell$, as in Manning's formula, $S$ can be calculated. In Fig. 8 the relations of $S$ to $A$ and $D$ to $S$ are shown. $D$ may have a tendency to increase in relation to $S$.

On the other hand, to study the effects of irregularities in the longitudinal direction, we carried out an experiment. In the experiment a portion of the tracer flows through a driving channel of the Ponmojiri power station and other portion of the tracer through the reservoir. As a consequence, the latter was found to be greatly diffused as compared with the former, having passed through a short path. Though the flow times differ greatly, this
diffusion phenomena seems to be similar to the variation of a flood wave through the reservoir. Such an irregularity may be one of the main causes of an increase in the diffusion.

In addition to the above considerations, the correlations of D to velocity and roughness were investigated. The relation between D and flow velocity is plotted in Fig. 9. As it was impossible to distinguish the effect of flow velocity alone, a significant correlation could not be obtained. By the use
of Manning's formula of the mean velocity through a reach, the average river roughness $n$ is given as follows:

$$n = rac{1}{v}.$$

(11)

In calculating $n$, we used a value of $H$ at the cross-section downstream as an average depth in the reach but used a mean value of the depths both upstream and downstream when they are appreciably different from the average depth. This average river roughness $n$ covers all indefinite factors such as bends in river channels, irregularities of longitudinal river beds and cross-section. Thus, the relation shown in Fig. 10 between $D$ and $n$ was obtained. The diffusion coefficient tends to increase in proportion to the average river roughness.

In natural river channels, such values of $n$ cannot be estimated even by the current-meter method at many stations located in a reach. However, an easy estimation is possible only when the flow velocity is measured by means of radioactive tracers over a fairly long distance.
5. CONCLUSIONS

As regards radiotracer techniques in field experiments on river flow, it is concluded that:

1. For direct measurement with underwater detectors, the scintillation-type detector is much more sensitive than is the G-M type. The scintillation-type of underwater detector used in this experiment, equipped with 1\frac{3}{4}-in diam x 1-in NaI crystal, has a detecting efficiency of $9 \times 10^4$ cpm/uc per cm$^3$ for $^{131}I$, $29 \times 10^5$ cpm/uc per cm$^3$ for $^{24}Na$ and $36.6 \times 10^5$ cpm/uc per cm$^3$ for $^{82}Br$.

2. As to the choice of radioisotopes for this kind of experiment, $^{82}Br$ may be better than $^{24}Na$ in such respects as detection sensitivity, longer life and more ease in shielding because of the lower energy of the gamma rays; hence it is possible to send large amounts by air.

3. In this experiment water flow could be traced for 30 km at the most with 52 mc of $^{24}Na$, but it may be possible to extend the traceable range with $^{82}Br$.

4. It seems to be difficult to measure the absolute radioactivity concentration in a river by direct means only because of the variation of the geometry at an observation point. Therefore, it is advisable to use a sampling method at the same time if concentration measurements are needed.

From this series of studies in river engineering, our conclusions concerning the method for flow velocity measurements, the flow time for water supplied from the dam to each intake and the diffusion of tracer are as follows:

1. The estimation of flow velocities from the measuring data actually taken at the representative stations is not accurate in a reach of more than about 5 km. The ratios of the average surface flow velocities obtained by the float measurements to flow velocities of water mass through the reaches vary with the length of the reach. The measurement of flow velocities with radioactive tracers is much more accurate than that with salt water.

2. We were able to make a diagram of the flow time to each intake plotted the individual discharges of the operating stations.

3. The values of the longitudinal diffusion coefficient in the Sorachi River were within $(0.4-7) \times 10^5$ cm$^2$/s, and the concentration gradient of the tracer in the cross-sectional direction was recognizable in reaches shorter than 10 km. Irregularities in the cross-sectional and longitudinal directions of the river channels and average river roughness throughout the reach are factors influencing the diffusion of tracers.

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REFERENCES

DISCUSSION

J. F. CAMERON: Could you perhaps indicate what maximum permissible concentration (MPC) value you use in the hydrological work? Do you have any special regulations for short-term experiments in Japan?

M. KATO: The MPC that we use corresponds to the value laid down for drinking water in the Japanese legislation on radiation protection. This is in line with the value recommended by the International Commission for Radiological Protection (ICRP). There are special regulations governing field tracer experiments, for which a government license is needed.

L. O. TIMBLIN: Dr. Kato, your injection device is very interesting in that it provides for subsurface injection. In what respects do you feel that this technique is superior to ordinary surface injection?

M. KATO: Since our aim was to inject the tracer at the point of maximum velocity, we were very much concerned with trying to prevent the waves on the water surface from interfering with tracer diffusion immediately after the injection. A further advantage of the technique is that it lowers the exposure of operating personnel.

D. B. SMITH: Since you used sodium as a tracer, do you have any figures similar to those in Table II for the natural sodium content of the river?

M. KATO: We did not measure this value specifically for our experiment. On the basis of the available data, however, I would say that natural sodium could be present in anything up to ppm quantities.

P. C. LÉVEQUE (Chairman): In Table II of your paper, mention is made of evaporation residue and the values quoted are low. I take it that the water in question moves through crystalline, metamorphic or volcanic formations. There are probably very few sedimentary deposits in the river basin. Do you not think that one of the factors involved might possibly be the natural radioactivity of the ground and perhaps even the water?

M. KATO: I am afraid I can give no very definite answer since we did not investigate this point specifically. However, before we embarked on the experiment, we took water samples and studied the natural background along the entire stretch of the river using portable counters. An appreciable level of activity was found in one spot only and on the whole the natural activity in both the rock and the water was very low and did not obstruct the experiment in any way.

P. C. LÉVEQUE (Chairman): I should like to ask one more question, Mr. Kato, again in connection with the low value quoted in Table II for the evaporation residue. Do you not think that the adsorption and consumption of radioisotopes are liable to be much higher when the water contains fairly small amounts of soluble salts than when it contains say three or four times the quantity of evaporation residue per litre?

M. KATO: According to our experimental data, the self-decay resulting from absorption or adsorption is not particularly high. Only in one reach
Y. MIYAKE: I should like to make one brief comment on this problem of loss of radioisotope through adsorption and other processes. In the case of sodium or other main constituents of river waters, the losses are probably not great. In the case of iodine, however, which is present only in pp 109, there may easily be some loss of iodine through adsorption or biological uptake, particularly when the iodate-iodide ratio is favourable.

P. C. LÈVÈQUE (Chairman): That would confirm what I suspected, at least for iodine. I believe that in some experiments carried out by the French Commissariat à l'énergie atomique, which involved the injection of iodine into silty ground, the loss of tracer was so serious as to impair the value of the whole investigation—although, as far as I remember, the percolation distance was only between 3 and 7 m. Of course, the conditions were not quite the same as in a river or an open channel. However, sometimes even anionic radioisotopes are affected by loss or absorption.

G. H. LEAN: In the case of a straight channel, it is usually assumed that the diffusion coefficient D is proportional to the product of the shear velocity and the depth. I wonder if any better correlation could be obtained with this factor in Figs. 8-10?

Y. MORITA: In the natural river channels we studied, the cross-section and slope of the river bed varied appreciably—sometimes to quite a remarkable extent—even within one reach. This made it difficult to determine the representative depth H and the shear velocity $V_s$ in any particular reach. We did try to investigate the correlation between $H$ and $V_s$ at each representative station, but because of these variations it proved impossible to obtain a good correlation. We decided therefore to abandon any attempt to investigate the relationship between $D$ and $V_s$. In our view, the many factors that are liable to affect $D$ are provided for in the average $n$, as defined in our paper and estimated on the basis of flow-velocity data. The correlation between $D$ and $n$ is fairly good as Fig. 10 shows. Of course, $n$ correlates with $H$, and $V_s$ is an important factor from the point of view of $D$, turbulence and tractive force, particularly in a straight channel where the cross-section does not vary.

In a future study, we shall be examining the possibility of averaging the products of $H \times V_s$ by surveying the cross-sections at many stations in each reach.

P. C. LÈVÈQUE (Chairman): There are a few general comments I should like to make on the first three papers of these proceedings. First of all, I think it is important to bear in mind the possibility of water penetrating through permeable strata and reaching a river in between the point of injection and the point at which the measurements are made. Of course, there are cases where this is not likely to happen to any appreciable extent; e.g. when the river cuts through an area of marl, clay, or even very marly limestone. On the other hand, if the river passes through a real limestone or sandstone region, the flow can quite easily increase by something on the order of 2 to 5%. Before blaming flow increases on faulty electronics or imperfect geometrical conditions one should, I feel, look into this possibility of lateral influxes. This is particularly important when the measurements...
are made on stretches of tens of kilometers as in the case of the Sorachi river.

Another interesting point is that none of the three papers made any reference to major difficulties arising from the dissolved salt content of the water. I wonder whether this might not perhaps indicate that the measurement techniques employed are insufficiently fine. Mr. Kato, and Mr. Guizerix have spoken of the stringent safety standards which are applied to experiments of this sort in their countries. At least this is a spur to our ingenuity and it may even give rise to interesting advances in fine-counting techniques.
METHODS FOR MEASURING SAND TRANSPORT USING RADIOACTIVE TRACERS

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Abstract — Résumé — Аннотация — Resumen

METHODS FOR MEASURING SAND TRANSPORT USING RADIOACTIVE TRACERS. Radioactive glass particles are sometimes used in the field for investigating the behaviour of sand and silt. Experiments have been performed in a flume with a rippled sand bed to compare the rate of movement of sand and glass particles of the same size and density. Several methods of measuring sand transport in the flume using radioactive tracers are described and compared. Two of these methods have been investigated in a series of tests; the first, usually termed the space integration method, involves the measurement of the spatial distribution of particles from a strip of the bed after various times; the second, referred to as the time integration method, requires the measurement of the passage of the tracer in time at a station downstream. These methods have been used to measure the velocity of several size fractions in a sand mixture.

The practical difficulties of applying these methods to measurements in the field are briefly examined.

EMPLOI DE RADIOINDICATEURS POUR MESURER LE MOUVEMENT DU SABLE. On utilise parfois des particules de verre radioactif pour étudier in situ le comportement du sable et de la vase. On a fait des expériences dans un cours d'eau à lit de sable râpé pour comparer les vitesses de déplacement du sable et des particules de verre de même dimension et de même densité. Les auteurs décrivent et comparent plusieurs méthodes de mesure faisant appel aux radioindicateurs. Deux de ces méthodes ont été étudiées au cours d'une série d'essais; la première, généralement appelée méthode d'intégration dans l'espace, consiste à mesurer la distribution spatiale des particules provenant d'une bande du lit à divers moments; la deuxième, appelée méthode d'intégration dans le temps, consiste à déterminer dans le temps le passage de l'indicateur à une station située en aval. Ces méthodes ont servi à mesurer la vitesse de plusieurs fractions de tailles différentes dans un mélangé de sable.

Les auteurs passent rapidement en revue les difficultés que soulève l'application de ces méthodes dans le cas de mesures in situ.

МЕТОДЫ ИЗМЕРЕНИЯ ПЕРЕНОСА ПЕСКА С ПОМОЩЬЮ РАДИОАКТИВНЫХ ИНДИКАТОРОВ. Частичи радиоактивного стекла используются иногда при изучении поведения песка и ила. Для сравнения скорости движения песка и частиц стекла того же размера и плотности эксперименты проводились в горном русле с неровным песчанным дном. Дано описание и сравнение нескольких методов измерения переноса песка в горном потоке с использованием радиоактивных индикаторов. Два из этих методов исследовались в целом ряде испытаний. Первый, обычно называемый методом пространственного интегрирования, заключался в измерении пространственного распределения частиц определенной полосы для через различные промежутки времени. Второй, называемый методом временного интегрирования, требует измерения времени прохождения индикатора до станции, расположенной ниже по течению. Эти методы использовались для измерения скорости частиц нескольких размеров в смеси песка.

Кратко рассмотрены практические трудности применения этих методов в полевых условиях.

MÉTODOS PARA MEDIR EL TRANSPORTE DE ARENAS CON INDICADORES RADIÁCTIVOS. En algunos casos se emplean partículas de vidrio radiactivo para investigar los desplazamientos de arenas y aluviones. Los autores han estudiado una acequia con fondo de arena ondulada con el propósito de comparar los movimientos de la arena y de partículas de vidrio del mismo tamaño y densidad. La memoria describe y compara diversos métodos para medir el transporte de arenas en acequias por medio de indicadores radiactivos. Dos de ellos fueron investigados en una serie de ensayos; el primero de los métodos, generalmente denominado de integración...
1. INTRODUCTION

It is nearly ten years since radioactive tracers were first used for the examination of sediment movement. During that time, with many countries conducting field studies into the behaviour of silt, sand and shingle, experimental techniques have been refined, but no equivalent improvement of interpretation analysis has been forthcoming. As with the first experiments, results are only qualitative, showing the direction of movement and giving some measure of the speed of movement, but without providing the volume of sediment moved over a certain time. Such quantitative results have been outside the scope of the work carried out hitherto, and it is the purpose of this paper to discuss the methods by which these might be attained in the particular case of unidirectional sand transport.

If a quantitative assessment of sand transport is the aim, then, as a first essential, the tracer chosen to represent the movement under study must behave in the same way as the sediment in the hydraulic processes. Sand particles, surface-labelled with a radioisotope by adsorption from a radioactive solution, have been employed by some workers. However, owing to the vulnerability of the surface label to abrasion, glass particles incorporating an element which on irradiation possesses the desired radiation properties are often preferred for long-term experiments. An early experiment carried out in a laboratory channel at this Station by the staff of Wantage Research Laboratory had indicated that there was a difference between the behaviour of glass and sand particles of the same size [1]. In that experiment glass particles, volume-labelled with rubidium-86, and sand particles, surface-labelled with barium-lanthanum-140, were used. After irradiation, samples of glass and sand particles were mixed and laid at a point on the bed, and the movement of the tracer down the flume was measured after various running times. However, the different method of labelling, by volume in the first case and by surface-labelling in the second, together with the problem of distinguishing the activity-contributions from each of the radioisotopes, necessitated lengthy experimental procedures for determination of the relative concentration of the tracers along the flume. This involved sampling and the counting of sieved fractions of each sample on a 100-channel pulse analyser. The results suggested that the glass moved faster than did the sand, but it was not possible to analyse sufficient samples for this result to be conclusive. For this reason the simpler experiment described in section 2, in which only a single size fraction was used and the movement of glass and sand was examined in separate tests, was undertaken.

There are four main ways in which the discharge of sand or silt particles along a channel can be measured by tracers. These may for brevity be termed the space-integration method, the time-integration method, the steady-dilution method and the cloud-velocity method. Apart from the first, these have their counterparts in the measurement of the discharge of liquids,
METHODS FOR MEASURING SAND TRANSPORT

In the first method the sample is injected at a section and after a time long enough for the particles to "forget" their initial situation and for them to wander over the whole field; the velocity of their centre of gravity is determined by successive measurements of their concentration in space. In the second method the concentration is measured at a fixed section which is sufficiently far downstream for the initial circumstances of injection to be lost and for the particles to have taken up the average velocity of the particle flow. The steady-dilution method is a variant of the second method in which the injection is continuous at a constant rate and the same restriction on the distance between the injection section and the measuring section is necessary as in the other methods. The cloud-velocity method is essentially the same as the first method, but in its application to the flow of liquids it has been more usual to measure not the successive distributions of the dilutant in space but the average time for its passage between two sections.

Experimental work has been carried out in a laboratory channel on some of the above methods, employing tracer sand grains of different particle diameters in transport in a rippled sand bed. The laboratory experience has allowed an evaluation of the various methods and at the same time demonstrated the difficulties, peculiar to each method, that will be encountered on extension of the work into the field.

2. THE SUITABILITY OF GLASS PARTICLES AS A SAND TRACER

Boron-free soda glass incorporating a small percentage of scandium-46, iridium-191, zinc-65 or some other element which exhibits suitable radiation properties has often been used for sand movement studies. Although matched in size and density, the glass grains tend to be more angular than are fluvial or marine sand particles, and there has been some doubt as to whether such shape differences are significant. To resolve this question we compared the movement of sand and glass particles in a flume in which the conditions were held constant.

For simplification of the measurements only a limited size fraction of the sand was used as tracer. Treatment of the flume sand was necessary before irradiation [2] to remove minerals of specific gravity higher or lower than that of quartz, the principal component of the sand. The specific activity of the irradiated quartz grains was low, sodium-24 and bromine-82 being responsible for the short-lived gamma radiation; and the flume experiments were carried out with a 150-g sample, closely graded to 152 to 178 μm particle diameter, with an activity of about 1 mc.

A melt made up of SiO₂ 67.2%, CaO 13.4%, Na₂O 14.4% and PbO 5.0% gave a glass of specific gravity 2.645, satisfying the density-matching conditions to within 0.2%. The molten glass was dropped into cold water for rapid cooling and later annealed. This process resulted in irregular glass lumps of about 0.5 cm diameter, having little internal stress. The lumps were shattered in a grinding mill and particles in the diameter range 152 to 178 μm separated by sieving. The particles produced in this way were slightly more angular in shape than were the equivalent sand grains (Fig.1). 20 g of particles of this size was irradiated to give a sodium-24 activity of 0.5 mc at the commencement of the experiment.
The channel used to compare the behaviour of the two tracers was 350 ft long by 5 ft wide; in it a water discharge of 10 ft$^3$/sec was run over a self-formed sand bed at a depth of 1.36 ft. The bed was replenished at the upstream end by the feeding of sand at a constant rate while sand in movement was trapped in a settling basin at the downstream end of the flume. At the time of the tests the longitudinal profile was in approximate equilibrium, with the transport rate reasonably steady at 90 to 100 lb/h. The mean size of the sand on bed was 180 $\mu$m, and movement occurred both by surface creep and in suspension. The bed itself was highly rippled, the ripples being irregular both in plan and in section. The rms elevation of the ripple surface relative to mean bed level was 0.030 ft; and the wavelength of the ripples, defined as the average distance between alternate zero crossings, i.e. the crossing of the ripple profile with the mean bed level, was 0.70 ft. About six months elapsed between the glass and equivalent sand tracer tests. During the intervening period the flume was in operation; and although the water discharge, water depth and sand feed rate were held constant, small changes in the bed profile did occur, and the transport rate was not reproducible to closer than 10%.

The experimental procedure was the same for both tests. A strip of the bed, 1.5 ft long across the complete width of the flume and to a depth of 0.125 ft below the mean bed level, was replaced with a dilute mixture of tracer and bed sand. The subsequent spread of the tracer downstream, after 4 h and 32 h of flume running, was measured by means of a scintillation counter. The glass experiment could not be continued beyond 32 h owing to the rapid decay of sodium-24 (half-life, 15 h). For each measuring survey the flume was drained to within 0.2 ft of the ripple crests. A sodium iodide crystal, 1.5 in in diameter, was held at a fixed distance above the water surface and the activity registered on a ratemeter or, when greater sensitivity was required, on a scaler. Although the detector was shielded by a series
of lead rings, scattered radiation was still responsible for a significant contribution to the total radiation measured; and it was necessary to apply scatter corrections to all readings to obtain the activity for the area of bed directly beneath the detector. This correction was particularly important in the early stages of the experiments when the concentration gradient was steep. Tests showed that, for the limited depth range of sand movement experienced in the flume, the recorded activity level was virtually independent of tracer distribution in depth, so the readings could be interpreted as directly proportional to the weight of tracer per unit area.

The dispersion of the two tracers after 4 h and 32 h is compared in Fig. 2. The readings at 4 h were in close agreement, but at 32 h glass concentrations were low for a distance of 80 ft downstream from the origin. However, it will be noted that, at the origin itself, glass tracer was persisting at 32 h in concentrations four times in excess of the sand tracer.
For the glass experiment the average thickness of bed removed from the origin after 32 h was only 0.08 ft, compared with 0.11 ft for the sand tracer experiment at the equivalent time. The amount of tracer leaving the origin is closely related to the prevailing ripple configuration. It appears that the period 4 h to 32 h of the glass experiment corresponded with abnormally high bed elevations at the injection strip, leading to a starving of the area immediately downstream during that time and the trapping of the tracer at the origin. The average glass particle velocity moving in a layer extending to 0.12 ft below the mean bed level was 2.33 and 1.90 ft/h after 4 h and 32 h respectively, compared with the equivalent sand tracer velocities of 2.00 and 1.98 ft/h.

In addition to a survey of the spatial distributions of tracer after certain periods of flume running, the passage of the tracer through a section 12 ft downstream of the injection strip was measured. A continuous record of the activity at this section was taken for the duration of flume running by means of G60H Geiger counters, each 2.46 ft long and 0.12 ft in diameter, set across the flow and buried in the sand bed to a depth greater than that reached by the deepest troughs. The passage of sand and glass tracer par-

![Graph](image-url)
METHODS FOR MEASURING SAND TRANSPORT

Particles is compared in Fig. 3. The velocity of the tracer particles can be derived from these curves of tracer concentration, being inversely proportional to the area beneath the curves. Integration of the concentrations from 0 h to 32 h shows the glass tracer to be travelling at an average velocity 6% greater than that of the sand tracer.

The small difference of velocity between the two tracers revealed in both the spatial- and time-integration tests is almost certainly due to slight changes in the flume conditions between the tests and random deviations from the average ripple pattern which would occur in a short experimental time.

3. TRACER METHODS FOR DISCHARGE MEASUREMENTS IN LIQUIDS

Before discussing the methods outlined in section 1 for measuring sand transport, let us consider the application of analogous methods for discharge measurement in liquids. We shall see that the problems have several features in common. Suppose liquid flows through a uniform open channel and dilutant is introduced uniformly over a section \((x = 0)\) in a length \(X\) small compared with the depth \(h\). Initially the dilutant is convected with the flow and after a time \(t\), which is short compared with that for dispersal by molecular or turbulent diffusion, each element of dilutant is carried downstream a distance \(x = Ut\), \(U(y)\) being the local velocity in which the element was placed.

For a wide channel and ignoring the lateral velocity variation, if the initial concentration is \(C_0\), the mass of dilutant between \(x\) and \(x + \delta x\) is given by

\[
-C_0 \delta x(x/t) \frac{dy}{dU},
\]

where \(y\) is the distance from the bed.

The average concentration \(C_m\) at \(x\) is given by

\[
C_m = \left(\frac{C_0}{h}\right) \left(\frac{x}{t}\right) \frac{dy}{dU} \bigg|_{0}^{x/t}, \quad 0 < x < U_0 t
\]

where \(U_0\) is the maximum velocity across the section.

As the motion proceeds, the concentration gradients generated by convection tend to be smoothed out by diffusion. The time required for diffusion effects to become appreciable is not easy to calculate since it depends both on the variation of velocity and turbulent diffusion in depth. However, we may obtain a rough indication of its magnitude by assuming that the velocity and diffusion coefficient \(D_1\) are constant across the section and by calculating the time required for an initial variation of concentration proportional to \(\cos \pi y/h\) to decay. The time \((t_1)\) for the variation in depth to fall \(1/e\) of its initial values is given by

\[
t_1 = \frac{h^2}{\pi^2 D_1},
\]

and during this time the dilutant will have travelled through a distance

\[
L_1 = \overline{Ut_1} - \overline{U} h^2 / \pi^2 D_1.
\]
BAILEY and GOGARTY [3] have shown that, for laminar flow in a pipe, a time approximately equal to $5t_1$ must be allowed before radial changes due to diffusion are rapid compared with longitudinal changes due to convection. If it is assumed that a similar factor applies to turbulent flow in a wide channel,

$$L_1 \approx \frac{\bar{U}h^2}{2D_1}.$$  \hspace{1cm} (4)

For turbulent flow through a channel, the average value of $D_1$ over the section is given by

$$D_1 \approx 0.067 U_T h,$$ \hspace{1cm} (5)

in which $U_T$ is the friction velocity ($U_T = (gih)^{1/4}$, $i$ is the water surface slope in uniform flow), and substitution gives

$$L_1 \approx 7.5 \frac{\bar{U}h}{U_T}.$$  \hspace{1cm} (6)

In our experiments $U_T = 0.135$ ft/sec and $\bar{U}h = 2$ ft$^2$/sec, giving $L_1 \approx 110$ ft.

If sufficient length is allowed for variations of concentration over a section to be small compared with those in the longitudinal direction, TAYLOR [4, 5 and 6] has shown that the distribution of dilutant originally uniformly spread across a section is given by

$$C_m = \left(C_0 X/2(\pi D t)^{1/2}\right) \exp\left(-\frac{\xi^2}{4Dt}\right),$$  \hspace{1cm} (7)

in which $\xi = x - \bar{U}t$ and $D$ is the effective diffusion coefficient which is associated with the combined action of local turbulent diffusion and the longitudinal dispersion due to the variation of velocity over the cross-section. This coefficient is much greater than that ($D_1$) resulting from turbulence alone. ELDER [7] gives for a wide channel

$$D = 5.86 U_T h.$$  \hspace{1cm} (8)

It will be clear from (7) that at any time the pulse of dilutant is distributed in a Gaussian manner about a point centred on a section moving with the mean velocity. Also, the distribution of the pulse moving through a fixed section is not Gaussian in time but displays a long tail. However, at a section which is sufficiently distant even the time distribution tends to the normal form. This is easily shown by substitution of $T + \tau$ for $t$, in which $T$ is the time at which a point travelling with mean velocity $\bar{U}$ passes $x$ and $\tau$ is the time taken for a particle to travel from $x$ to $\xi$ so that $\xi = U_T \tau$ and

$$C_m \approx (C_0 X/2(\pi DT)^{1/2}) \exp\left(-\bar{U}^2 \tau^2/4DT\right).$$  \hspace{1cm} (9)

We now proceed to examine the application of tracer methods to discharge measurement.
(i) The spatial-integration method

It will be evident that, if the fluid is originally uniformly labelled with dilutant across the section, the velocity of the centroid of the dilutant is equal to the discharge velocity. In steady flow this result appears to hold whether or not the channel is straight and follows from the fact that the dilutant is supposed to be hydrodynamically indistinguishable from the liquid. It is clear also that the time that has elapsed from the initial injection is unimportant. In (1), for example, immediately after injection the centroid velocity

\[ \frac{1}{C_0} \int_0^t C_0 \delta x \frac{X}{t} (dy/dU)(x/t), \]

with \( x = Ut \) is equal to the mean velocity

\[ \bar{U} = \frac{1}{h} \int_0^h U(y)dy. \]

If the flow is not uniformly labelled over a cross-section, clearly some time must be allowed to elapse for the dilutant to spread over the flow field.

(ii) The time-integration and steady-dilution methods

In the time-integration method the concentration at a fixed section downstream from the point of injection is measured, and the velocity given by

\[ \frac{V}{A} \int_0^\rho C_m dt \]  

is sometimes taken as the discharge velocity. (\( V \) is the mass of dilutant injected, and \( A \) is the cross-sectional area.) However, for (10) to give the discharge velocity some degree of mixing is essential. Let us consider the case of an initial uniform labelling over a section. After a short time, before appreciable mixing has taken place, \( C_m \) is given by (1) and the velocity given by (10) is

\[ C_0 Xh/ \int_{t=\rho/U_0}^\rho (C_0/h)(X/t)(dy/dU)_{U=\rho/t} dt = \frac{1}{h} \int_0^h [1/U(y)] (dy/h), \]

which is only equal to the discharge velocity if the velocity is uniform over the depth. It is clear therefore that the validity of the time-integration method depends on mixing. It might be conjectured that the method will only yield the discharge velocity if the distance, "the accommodation length" between the injection and measuring sections, is sufficient to allow each particle of dilutant to have wandered over the whole section. As we have seen, this
is the condition that (7) should represent the distribution, and it is therefore not surprising that substitution of (7) in (10) immediately yields the discharge velocity. It may be observed that this conclusion holds whether the dilutant is spread uniformly over the section initially; i.e. the accommodation length is independent of the initial distribution over the section.

Similar limitations apply to the steady-dilution method. In this method the dilutant is continuously supplied to the flow at a constant rate (q) and the steady concentration $C_s$ is measured at a section downstream. We may note that $C_s$ may be considered as made up of a succession of contributions from a series of instantaneous injections $V$ at intervals of time $\delta t$, and

$$C_s = \frac{1}{\delta t} \int_0^\infty C_m dt$$  \hspace{1cm} (12)

Evidently the condition of mixing required for (10) to be valid applies equally to the steady-dilution method, and the velocity given by

$$\frac{q}{AC_s}$$

will only be equal to the discharge velocity if the distance between the points of injection and measurement is greater than the accommodation length.

(iii) The cloud-velocity method

The cloud-velocity method relies on the circumstance that, if a cloud of dilutant is released in the flow, its average velocity will eventually be equal to the discharge velocity. In this form the cloud-velocity method is identical with the space-integration method, but in practice it is more usual to measure the average time taken by the cloud to pass two sections at a given distance apart. If the cloud originally consists of dilutant uniformly spread over a section, the average time for the cloud of particles to travel a distance $x$ from the origin is given by

$$T = \frac{1}{C_0 X A} \int_0^\infty t dt \left[ \frac{\partial}{\partial t} \int_0^x A C_m dx \right].$$  \hspace{1cm} (13)

The velocity $\frac{dx}{dT}$ with which the cloud moves through a section is given by

$$\frac{dx}{dT} = \frac{C_0 X A}{(d/dx)} \left\{ A \int_0^\infty t dt \left[ \frac{\partial}{\partial t} \int_0^x C_m dx \right] \right\} = \frac{V}{A} \int_0^\infty t (\partial C_m/\partial t) dt.$$

Integrating by parts and inserting the limits gives

$$\frac{dx}{dT} = \frac{V}{A} \int_0^\infty C_m dt,$$  \hspace{1cm} (14)
which is equal to the velocity given by the time-integration method. In this interpretation of the cloud method, therefore, the time-integration and cloud methods are equivalent, and the same limitations regarding dispersal and accommodation length apply to both.

With the distribution (7) it is easily shown that the velocity with which the maximum concentration of the pulse passes, as determined from the \( C_m-t \) curves at consecutive stations, is closely equal to \( U \).

4. TRACER METHODS FOR SAND TRANSPORT

When we turn to the application of the methods just described to the measurement of sand transport, we immediately encounter two important differences. The mean velocity of the particles is much smaller than the liquid, and so also is their rate of dispersion. The median sand fraction for example moves with an average velocity of 2.0 ft/h, and after 100 h it is dispersing about the centroid position with an effective diffusion coefficient of the order of \( 1 \times 10^{-2} \text{ft}^2/\text{sec} \), compared with 1.0 ft\(^2\)/s for the water flow. The time for the particles to pass a given section is proportional to \( (xD/U)^{3/2} \) so that the sand cloud takes about \( 10^4 \) times as long as a diluant in the water.

The reason for the slow velocity and dispersion of the particles is the trapping effect of the ripples. When particles resident in a strip of the bed are exposed by the ripple movement, they are taken up by turbulence at the bed and convected for a certain distance before being deposited. If they alight in an area of local accretion, on the leeside of a ripple, for example, they may be buried for some time before being released again. This has the effect of reducing both the velocity of the particles and the average time available for diffusion.

We may gain an insight into the factors affecting the dispersion by considering a simple hypothetical model of the movement in which a fraction \( c \) of the ripple layer is assumed to move through an average distance \( L \) in time \( T \). After a time \( t = nT \) the fraction of material \( \mathcal{G} \) at a distance \( x = mL \) is given by

\[
\mathcal{G} = nC_m c^n (1-c)^{n-m},
\]

and the mean particle speed \( u \) is given by

\[
u = cL/T.
\]

When the motion has been continuing long enough to make \( cn \) large, the distribution tends to become Gaussian, i.e.

\[
\mathcal{G} = \exp[-(x-ut)^2/2ut(1-c)L]/[2\pi c(1-c)(t/T)]^{1/2},
\]

giving a longitudinal distribution which is symmetrical about a point moving with the mean particle speed.

This equation has obvious similarity with equation (7) for the distribution of diluant in the flow, the effective diffusion coefficient for the particles
being given by

$$\frac{1}{2}u(1-c)L.$$  \hspace{1cm} (17)

The diffusion coefficient evidently depends both on the mean particle speed and on the length of hop (L). This is illustrated by the curves of Figs. 4, 5 and 6, which show the longitudinal distribution of labelled particles, after various times, having mean sizes of 110, 160 and 270\(\mu\)m. The average velocity of the particles determined from the movement of the distributions were 7-10, 2.0 and 0.43 ft/h respectively. The finest particles dispersed so rapidly that the activity had values close to background after only 32 h. Much greater initial activities would have been required to enable this size fraction to be tracked for a longer time. After 100 h the diffusion coefficient for the coarse particles was of the order of \(1 \times 10^{-3}\) ft\(^2\)/sec, indicating that their hop lengths were less than half that of the median fraction.

(i) The accommodation length

In section 3 it was noted that for liquids, if the distance between the injection and measuring sections was larger than the accommodation length, the time-integration method for determination of the mean velocity was valid and the distribution would conform with equation 7. With particle motion, however, it is not necessary to achieve this distribution for the time-integration method to succeed. For example, with the simple binomial distribution given by (15)

$$u = 1/\sqrt{\frac{1}{2}} (\xi/L)dt$$

after only a single hop length. However in general the hop length is not constant but is distributed over a wide range even for particles of similar size. When this factor is taken into account, in general the time-integration method gives the mean particle speed only after many hops have been performed. It appears to be essential to allow sufficient hops for each particle to have had the opportunity of performing a variety of hops, i.e. for each particle on its successive flights to have wandered over the whole flow field. This condition is analogous to that for the time-integration method to be valid for liquids, and we might surmise that an indication of the accommodation length for particles could be calculated in the same way as for liquids. Dobbyns [8] has given an expression for the damping of initial perturbations of concentration of suspended solids superposed on the equilibrium distribution for flow at uniform velocity with constant turbulent diffusion coefficient (\(D_1\)).

For an initial concentration perturbation proportional to \(\cos \pi y/h\), the time for the concentration to fall to \(1/e\) of its initial value (corresponding to (2)) is given by

$$t_2 = h^2/(\pi^2D_1 + 2wh),$$  \hspace{1cm} (18)
Fig. 4
Tracer distributions after stated times of flume running
Coarse size fraction

Fig. 5
Tracer distributions after stated times of flume running
Median size fraction
when \( \text{wh}/D_1 \) is small, \( w \) being the fall velocity of the particles. The distance for concentrations of fine particles to spread across the field is therefore about

\[
(1 + 2wh/\pi D_1)^{-1}
\]

(19)

times the distance required for the dilutant, i.e. about 60 ft for the fine particles. For the large particles the distances given by (19) will be too large since the particles will travel close to the bed where the velocities are lower.

Measurements of the velocity of the particles by the time-integration method roughly bear out these results [9]. It was found, for example, that an accommodation length of 24 ft was required for the time-integration method to give a velocity within 5% of the average particle velocity for the median particles. For the coarse particles an accommodation length of only 6 ft was required, and at 12 ft the velocity from the time-integration method for the fine particles was only about half the particle velocity.
(ii) **The depth of movement**

So far we have been mainly concerned with the measurement of the mean velocity of the particles. For derivation of the volume of material in transport the velocity must be multiplied by the quantity of material in movement in unit length of flow. With liquids no difficulty arises since the cross-section of flow is easily measured, but with sand and silt moving in ripples the boundary is less well defined. The ripples are irregular in section, both longitudinally and laterally, and the bed elevations are distributed approximately normally about the mean. Thus at any point, as the ripples move, the bed is disturbed to an increasing depth.

If all the particles in a typical longitudinal strip are labelled down to a known depth below the lowest layers which can be disturbed, the average thickness of material which has been moved can be obtained from the loss of activity from the strip. If the particles are spread on the surface originally, the movement of the centroid in a given time corresponds to the velocity of the particles in the average depth of the bed subject to movement during this time. The latter is obtained by an analysis of ripple elevation records; the depth of movement coinciding approximately with the lowest elevation occurring at a point since the introduction of the tracer.

When the volume of tracer per unit length is measured, the time-integration method essentially gives the particle velocity and the auxiliary measurement of the thickness of movement must be made to derive the volume transport. It may be noted that with liquids it is more usual to measure the concentration \(c\) at a point in the section rather than the volume of tracer per unit length. Application of the time-integration method gives the volume transport

\[
Q = \frac{V}{\int c \, dt},
\]

provided that the accommodation length is exceeded, in which case \(\int c \, dt\) is constant at all points of the section [10]. In the case of sand transport, equation (20) can still be applied if the concentration at some fixed level in the ripples is measured throughout. Unfortunately the concentration varies rapidly in depth in the ripples because of the absence of diffusion in the ripples themselves, so that the level has to be specified with some exactitude. It may be noted that, if the time integration in (20) is stopped after a long time, it has been found that a close approximation to the transport can be obtained with the thickness of movement applicable to that time.

5. **FIELD APPLICATIONS**

(i) **Injection**

In the flume there was no difficulty in introducing the tracer uniformly over the width and throughout the thickness of movement. This will seldom
be possible in the case of a river. As far as dispersion throughout the ripple layer is concerned, it has been shown [11] that the actual manner in which the tracer is introduced is of little consequence. However, it will usually be an advantage to introduce the tracer close to the bed to localize its starting position and ensure that the tracer has an early opportunity to mix with the bed. An attempt has been made to solve this problem by the enclosure of the tracer in small soluble capsules which can be attached to a weighted cord run across the river. So far, however, no completely satisfactory material for the capsule has been discovered. Flume experiments indicate that most materials dissolve unevenly, and the tracer tends to become lodged in the fragments of the capsule.

(ii) Detection

In the flume the restricted depth of sand movement in the ripple layer in conjunction with the high-energy gamma emission of the tracer permitted the interpretation of the weight of tracer per unit area directly from the activity readings, irrespective of the tracer distribution in depth. With such measurements the thickness of movement of the bed must be determined independently for derivation of the transport rate.

In some field problems the thickness of bed involved in movement may be very great due to the presence of large dunes or shoals which may lead to burial of tracer particles under several feet of sediment. In these cases the activity measured by a detector resting on the surface of the bed will take little account of the deeply buried grains, owing to the absorption of overlying sand. For the flume methods to be applied it would be necessary for the detector to be calibrated for the particular vertical distribution of tracer existing at the measuring point. The vertical distribution may be obtained from an examination of undisturbed core samples. This information, together with continuous records of the changes of ripple and dune elevations should also permit the thickness of movement to be derived.

The vertical distribution of concentration in cores may be measured by injection of a large quantity of fluorescent particles simultaneously with the radioactive tracer. The visual counting of fluorescent particles under ultraviolet light in successive layers of a core is more straightforward and sensitive than scanning with a collimated radiation detector.

Each of the methods analysed in section 4 have inherent features that restrict their application in the field, and we now describe a number of the more important points that need to be considered in their future use.

(iii) The spatial-integration method

The spatial-integration method has the advantage that no allowance has to be made for accommodation length. Subsequent to lateral and vertical dispersion, an analysis of the longitudinal distribution of the tracer after any time yields the average particle velocity, i.e. the centroid velocity. In practice the determination of the centroid position is not easy; small
quantities of tracer at large distances contribute significantly to the centroid determination yet necessitate measurement very close to the natural background radiation level. At present no theoretical treatment has permitted the prediction of the shape of the head of the tracer movement from the higher and more readily measured tracer amounts nearer the origin. There is little doubt that eventually the spatial distribution will be Gaussian, with coincident positions of the concentration maximum and centroid. The flume experiments suggest that the time required to attain this distribution would be very long except for particles moving close to the bed and the concentrations would then have fallen to values which would make the definition of the maximum difficult. There is some evidence, based on the indications of a three-layer model of the movement, that the shape of the head may become Gaussian in form at an earlier stage. The approach of the head to this form appears to depend on the variety of hop length that particles can perform in their movement. This is a subject which could profitably be investigated in very long artificial channels or canals.

(iv) The steady-dilution and time-integration methods

In both the steady-dilution and time-integration methods, even with initial uniform injection across the section, the measuring section must be sited at least one accommodation length downstream for a correct evaluation of the transport. This length will vary for each particular case. It seems to be dependent on the average hop length and the width of the velocity spectrum that the tracer particles experience when in movement and so is small for heavier particles travelling in surface creep.

Both methods possess the advantage that the transport rate can be obtained directly from measurements of concentration without any definite knowledge of the thickness of movement, provided sufficient length is allowed for particles to have moved over the whole thickness. Normally, intensity measurements, such as those made in the flume, do not yield a concentration unless the variation of concentration in depth is known. These methods are therefore more suited to concentration determinations by recovery of bed samples for subsequent laboratory measurement. In the time-integration method the position of sampling in the bed is critical, being confined to the same stratum within the moving bed layer at all times during the passage of the tracer. The use of a tracer emitting soft gamma rays offers the possibility of measurement of concentrations without sampling. For example, the energy level could be chosen so that tracer particles buried under a few millimeters of sand were not measured due to absorption. The mean activity over the troughs and crests of the ripples would then give a direct measure of the concentration at the average level of the surface of the bed layer.

In some respects the steady-dilution method has advantages compared with the time-integration method. The technique for sample recovery need not be so precise (since eventually the concentration is constant across the thickness of movement), and only one tracer concentration is required. In practice, several such measurements would be made to ensure that the steady value had been reached. For the time-integration method the concentration must be measured many times for a plot of the rise and fall of concentration
throughout the tracer passage. The contributions to $\int c\,dt$ from the slow-moving tail are particularly difficult to measure as concentrations are so low. The main disadvantage of the steady-dilution method is the practical difficulty of carrying out continuous injection over a very long period. In order to arrive at the equilibrium concentration the tracer injection must continue at least as long as the time required for a single injection of tracer to pass.

In natural channels bends and cross-currents give rise to differing velocities across any section and the transport is not constant throughout the width of the channel. If the tracer is introduced at a point or a line source, a length additional to the accommodation length must be allowed to permit all particles to experience every element of the flow field. The criterion for judging the adequacy of post-injection mixing in the steady-dilution method is the establishment of uniform concentration over the whole measuring section; similarly, in the time-integration method the required condition is uniformity of $\int_0 c\,dt$ over the section. Where the transport varies considerably over the width and lateral dispersion is slow, enormous quantities of tracer would be necessary for the concentration to be measurable so far downstream. In such cases it would be more reasonable to divide the river into longitudinal strips and measure the transport rate in each strip independently. For this approach to be valid the transport must be nearly uniform across each individual strip and mixing between strips over the accommodation length should be minimal.

(v) The cloud method

The cloud method has superficially several important drawbacks. It is essentially a method of measurement of particle velocity; and, as in the spatial method, the thickness of movement must be determined independently before the volume transport can be derived. Secondly, even when the tracer is initially laterally dispersed across the flow, a distance equal to the accommodation length must be allowed to the first measuring station to permit the tracer to disperse. Thirdly, at distances which are not much greater than the accommodation length the shape of the pulse will be markedly asymmetric, attenuated and extended in time, and no means are available for prediction of the average time directly from the shape of the pulse. This being so, it is surprising that in the flume the velocity of the maximum through consecutive measuring sections gave values which were quite close to the mean particle velocity for the median size sand. This held even for distances which were smaller than the accommodation length, both for initial tracer distributions which were well mixed in depth and those which were laid on the surface.

With the finer-sized particles the sections were too close together for resolution of the passage of the maximum. If further measurements showed that the maximum and mean particle velocities agreed, this method would be especially suitable for the finer-sized and faster-moving fractions.
6. CONCLUSION

The difficulties of measuring sand and silt transport in rivers with tracers are formidable; and, although all the methods discussed can be applied, each has its peculiar difficulties. In the present state of knowledge no particular method can be advocated, and it seems likely that the choice of method or combination of methods will depend very largely on the particular situation. In stretches of river which are straight with a well marked channel and without extensive berms on either side, the tracer can be injected across the width of the river, and any of the methods can be used. If the practical difficulty of continuous injection for prolonged periods can be faced, the constant-dilution method is practically the most attractive, since bed sampling to determine the concentration of tracer may be fairly crude and no auxiliary measurement of the thickness of movement is required. For rivers with irregular or divided channels and wide berms, measurement of the transport by injection across the width encounters severe difficulties of lateral mixing, and the best hope would appear to be a piecemeal evaluation of the transport from measurements taken over individual longitudinal strips.

The spatial- and time-integration methods appear to be very well suited to the measurement of "bed load", i.e. the transport of the heavier fractions of the bed mix, which travel close to the bed. The lighter fractions which travel mainly in suspension can be measured by conventional methods, so that the methods supplement one another. A preliminary appraisal suggests that the cloud method may also be specially suited to the measurement of the finer particles, but more research is needed to establish the limitations of this method.

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REFERENCES

DISCUSSION

R. HOURS: I have a couple of comments on the part of Dr. Lean's paper dealing with the identical hydraulic behaviour of natural sand and ground glass. The first has to do with a very simple experiment which was carried out at the Chatou National Hydraulics Laboratory several years ago, for the purpose of comparing the movements of sand and glass. In the experiment 200 kg of ground glass and 200 kg of granulometrically identical natural sand were distributed in a glass flume in two parallel and adjacent longitudinal strips, over a distance of several meters. Water was then turned on and permitted to flow through the flume for a considerable period. Observation of the displacement of the two materials and measurement of their weight upon arrival at the end of the channel did not show any significant differences in their transport properties. No account of this experiment has ever been published, but it has contributed to our extensive use of ground radioactive glass in France. Secondly, although the differences in the behaviour of the glass and sand are small, it has been our experience that the manner in which the glass is ground up has an important bearing on the form of the glass particles obtained. For example, a roller mill produces needles, which are very different from natural sand, and are therefore not suitable. A ball mill, or a bar mill, on the other hand, produces grains with smooth edges which—at least when viewed through a magnifying glass—are quite similar to natural sand. Moreover, the grinding should be halted frequently, to permit sieving, and then resumed. A hammer mill yields fine grains immediately. The identical character of the particles of glass and sand is also dependent on the diameter which is sought. For example, gravel cannot be properly imitated. It would be desirable for grinding and sieving procedures to be standardized and also for them to be described in greater detail in published material.

G.H. LEAN: We did not think that the variations in activity between glass and sand shown in Figs. 1 and 2 of the paper were significant. What differences there were could, I think, be attributed to the chance occurrence of deep ripples. I am not quite sure how the glass was prepared in this case, but in any event glass drops of the order of half an inch in diameter were crushed merely by means of a pestle and mortar. The photographs of fragments of glass and sand shown in Fig. 1 do not reveal any particular differences in shape except that the glass might perhaps be considered slightly more angular.

D.B. SMITH (Chairman): I understand from the description of your time integration method that you are calculating the average velocity of transport and that to obtain the mass transfer you also need to know the area of cross-section over which lateral movement takes place. This is not quite analogous to the corresponding time integration technique used for the flow
of liquids and is more like a pulse velocity determination. Does your technique enable you to dispense with the requirement of complete lateral mixing across the flume?

G. H. LEAN: I would not like to give the impression that we do not consider complete mixing to be necessary, because we certainly do. In the time integration method for determining the transport of sand, we have measured not the concentration at a point, but the weight of tracer per unit area which is proportional to the mean concentration over the depth of movement. The reason for this is that the concentration is markedly dependent on the level in the bed. If the concentration at a certain point is measured continuously the mass transfer can be obtained without a knowledge of the thickness of movement, as in the case of liquids. A rather precise positioning of the sampling point would be necessary however. The requirements for lateral mixing are the same, whether you measure the weight per unit area or the point concentration.
RADIOACTIVE TRACER STUDY OF SEDIMENT TRANSPORTED IN ALLUVIAL RIVERS. For some years we have been developing techniques and equipment for studying the movements of sediments in rivers by means of radioactive tracers.

Qualitative studies: The first tests were purely qualitative in nature, being concerned primarily with gathering information on the development of zones of transport in relation to liquid discharge, on the formation of bars and on the rate of advance of dunes, either for checking the satisfactory geometric and kinetic similarity of transport phenomena in a scale model (calibrating phase) or for evaluating the possibilities of correcting a channel for purposes of navigation. With these tests in view, measurements were made on the Niger, in the Republic of Mali (both in the river itself and on a model), and on the Loire, in France.

Quantitative studies: The lack of a truly satisfactory method for determining the amounts of sediment transported in rivers led us to consider the extent to which the radioactive tracer method would provide a solution of this problem.

Indirect method: In rivers where the alluvium is formed mainly of coarse gravel and pebbles, labelling of the alluvium (previously classified according to diameter) makes it possible to determine the hydraulic parameters for the onset of entrainment as a function of diameter, to derive transport granulometry as a function of liquid discharge and to calculate the sediment discharge by application of a transport formula (e.g. Meyer-Peter). This method is relatively simple to use, but its accuracy is directly related to that of the transport formula used. It has yielded excellent results for the Rhone (experiments at Pierre-Bénite and Vallabrègues).

Direct method: We are studying the adaptation of the dilution methods now used in measuring liquid discharges (continuous dilution method, integration (total count) method) to the measurement of sediment transport. However, the use of these methods requires that the lateral and vertical mixing in the mobile sediment bed be sufficiently great to obtain the necessary "good mixture" conditions.

To check the latter point, we are studying the dispersion mechanism of a tracer. Some tests have been made in laboratory channels (two-dimensional flow). Other tests, in which guidance has been provided by a scale model representing the actual section under study, have been carried out in the Loire near Orleans (full three-dimensional flow).

ETUDE DU DÉBIT SOLIDE DE CHARRIAGE DANS LES RIVIÈRES ALLUVIALES AU MOYEN DE TRACEURS RADIOACTIFS. Depuis quelques années, les auteurs ont développé des techniques et réalisé des appareillages pour l'étude des mouvements de sédiments dans les rivières au moyen de traceurs radioactifs.

Etudes qualitatives: Les premiers essais avaient un but purement qualificatif; il s'agissait principalement d'obtenir des indications sur l'évolution des zones de charriage en fonction des débits liquides, sur la formation des seuils et sur la vitesse d'avancement des dunes, soit pour contrôler la bonne similitude géométrique et cinématique du phénomène de charriage d'un modèle réduit (phase d'étalonnage), soit pour apprécier les possibilités de correction d'un chenal en vue de sa navigabilité. Dans cette optique furent effectuées des mesures sur le Niger (Mali) en nature et sur modèle, ainsi que sur la Loire (France).

Etudes quantitatives: L'absence d'une méthode réellement satisfaîsante pour déterminer les quantités de sédiments transportés par charriage dans les rivières a incité à étudier dans quelle mesure la méthode des traceurs radioactifs permettait de résoudre ce problème.
Méthode indirecte: Dans les rivières dont les alluvions sont formées principalement de gros graviers et de galets, le marquage des alluvions, préalablement triées par gammes de diamètres, permet de déterminer les paramètres hydrauliques correspondant au début d'entraînement en fonction du diamètre, d'en déduire la granulométrie de charriage en fonction du débit liquide et de calculer le débit solide par application d'une formule de charriage (Meyer-Peter par exemple). Cette méthode est d'un emploi relativement simple, mais sa précision est directement liée à celle de la formule de charriage utilisée; elle a donné d'excellents résultats pour le Rhône (expériences de Pierre-Bénite et Vallabrègues).

Méthode directe: Les auteurs étudient l'adaptation à la mesure des débits solides de charriage des méthodes de dilution, utilisées couramment pour la mesure des débits liquides: méthode du palier et méthode d'intégration (comptage total). Mais l'utilisation de ces méthodes implique que les brassages latéral et vertical dans la couche des sédiments mobiles soient suffisamment grands pour obtenir les conditions de «bon mélange» nécessaires.

Pour vérifier ce dernier point, ils étudient le mécanisme de dispersion d'un traceur. Des essais ont été effectués dans des canaux du laboratoire (écoulement à deux dimensions). D'autres essais, pilotés par un modèle réduit représentant le secteur d'étude naturel, sont entrepris dans la Loire près d'Orléans (écoulement réel à trois dimensions).

ИССЛЕДОВАНИЕ НАНОСОВ АЛЛУВИАЛЬНЫХ РЕК С ПОМОЩЬЮ РАДИОАКТИВНЫХ ИНДИКАТОРОВ. В течение нескольких лет разрабатываются методы и аппаратура для изучения наносов в реках с помощью радиоактивных индикаторов.

Исследования качественных сторон: Цель первых опытов заключалась главным образом в том чтобы выявить признаки эволюции зон выноса в зависимости от размера стока, признаки образования порогов и скорости движения дна с целью проконтролировать хорошее геометрическое и кинематическое подобие явлений выноса в уменьшенной модели (контрольная фаза) и установить возможности улучшения судоходности-канала. В этой связи были поставлены опыты на Нигере (Мали) и на Луаре (Франция). Опыты ставились в естественных условиях и на моделях.

Исследования количественных сторон: Отсутствие удовлетворительного метода для изучения количества наносов в реках побудило заняться изучением того, в какой степени метод радиоактивных индикаторов позволяет решить эту проблему.

Косвенный метод: В реках, где наносы образуются главным образом из песка и гальки, мечение этих наносов, предварительно классифицированных по величине диаметра твердых частиц, позволяет определять гидравлические параметры, соответствующие началу переноса песка и гальки в зависимости от диаметра твердых частиц, взвешив из этого гранулометрического состав наносов в зависимости от жидкого стока и рассчитать вынос по формуле выноса (например, по формуле Мейер-Петера).

Таким метод довольно прост, однако точность получаемых данных прямо связана с точностью примененной формулы выноса. На Роне данный метод (опыты Пьер-Менита и Валлабрега) дал прекрасные результаты.

Прямой метод: Для измерения дебита твердого стока при выносях мы исследуем возможность применения методов разбавления, часто используемых при измерении дебитов жидких стоков: метод лестницы и метод интегрирования (общий счет). Однако применение этих методов требует достаточно интенсивного бокового и вертикального перемешивания в слое подвижных наносов с целью подачи необходиным условий "хорошей смеси".

Для проверки этого последнего обстоятельства изучается механизм диспергирования индикатора. Соответствующие опыты были проведены на каналах лабораторий (сток в двух измерениях). Другие опыты, управляемые уменьшенной моделью, которая воспроизводит исследуемый участок в его естественном виде, проводятся на Луаре, около Орлеана (естественные стоки в трех измерениях).

ESTUDIO DEL ACARREO DE SÓLIDOS EN LOS RÍOS ALUVIALES POR MEDIO DE INDICADORES RADIOACTIVOS. En el curso de los últimos años, los autores elaboraron técnicas y construyeron aparatos para estudiar el movimiento de sedimentos fluviales con ayuda de indicadores radioactivos.

Estudios cualitativos: Los primeros ensayos fueron de sifón cualitativa; se trataba principalmente de obtener datos sobre la evolución de las zonas de acarreo en función del caudal, sobre la formación de los umbrales y sobre la velocidad de avance de las dunas, sea para comprobar si la geometría y la cinemática del fenómeno de arrastre, en un modelo a escala reducida, eran similares a las reales (fase de calibrado), sea para apreciar las posibilidades de modificar un canal para hacerlo más navegable. Con este fin se efectuaron mediciones en el río Níger (Mali), en la naturaleza y en modelo, y en el Loira (Francia).
Estudios cuantitativos: Al no disponerse de un método verdaderamente satisfactorio para determinar la cantidad de sedimentos que acarrean los ríos, los autores trataron de averiguar si el método de los indicadores radiactivos permite resolver el problema.

Método indirecto: En los ríos en que los aluviones consisten principalmente en grava y guijarros, la marcaición de dichos aluviones, separados previamente según los diámetros, permite determinar los parámetros higráulicos del acarreo en función del diámetro, deducir la granulometría de las sustancias arrastradas en función del caudal, y calcular el acarreo de sólidos con ayuda de una fórmula (por ejemplo) la de Meyer-Peter. La aplicación de este método no ofrece mayores dificultades, pero su exactitud depende directamente de la que corresponde a la fórmula de acarreo utilizada; ha dado excelentes resultados para el Ródano (experimentos de Pierre-Bénite y Vallabrègues).

Método directo: Los autores estudian la adaptación de los métodos de dilución que generalmente se emplean para medir los caudales de líquidos (método de la meseta y método de integración (recuento total)). Pero, para poder emplearlos, es necesario que los brazajes lateral y vertical de la capa de sedimentos móviles sean suficientemente intensos para obtener las condiciones necesarias de «bueno mezcla».

Con el fin de comprobar este punto, los autores estudian el mecanismo de dispersión de los indicadores. Han realizado ensayos en canales de laboratorio (flujo bidimensional). En el Loira, cerca de Orleáns (flujo tridimensional real), han efectuado otros ensayos iniciados en un modelo a escala reducida que representaba el sector estudiado.

Depuis quelques années, on a développé des techniques et réalisé des appareillages pour l'étude des mouvements de sédiments dans les rivières au moyen de traceurs radioactifs.

1. Etudes qualitatives

Les premiers essais avaient un but purement qualitatif; il s'agissait principalement d'obtenir des indications sur l'évolution des zones de charriage en fonction des débits liquides, sur la formation des seuils, et sur la vitesse d'avancement des dunes, soit pour contrôler la bonne similitude géométrique et cinématique du phénomène de charriage d'un modèle réduit (phase d'étalonnage), soit pour apprécier les possibilités de correction d'un chenal en vue de sa navigabilité. Dans cette optique furent effectuées des mesures sur le Niger (Mali) en nature et sur modèle, ainsi que sur la Loire (France).

2. Etudes quantitatives

L'absence d'une méthode réellement satisfaisante pour déterminer les quantités de sédiments transportés par charriage dans les rivières nous a incité à étudier dans quelle mesure la méthode des traceurs radioactifs permettait de résoudre ce problème.

2.1. Méthode indirecte

Dans les rivières dont les sédiments sont principalement constitués de gros graviers ou de galets, le marquage de ces matériaux de fond, préalablement triés par gammes de diamètre, permet de déterminer les paramètres hydrauliques correspondant au début d'entraînement des diverses classes de galets et de calculer alors le débit solide par tranches granulométriques à l'aide d'une formule de charriage (de Meyer-Peter par exemple) dans le domaine de charriage partiel, c'est-à-dire quand tous les matériaux constitutifs du fond ne participent pas au charriage.
Cette méthode est d'un emploi relativement simple, mais sa précision est évidemment liée à celle de la formule de charriage utilisée.

Pour le Rhône, en aval de Lyon, cette méthode a donné des résultats encourageants. En particulier, les campagnes de mesure ont permis de mettre en évidence la variation du coefficient d'entraînement des galets avec leur diamètre: pour le cas particulier du Rhône ce coefficient varie sensiblement en fonction inverse du diamètre et tend vers une limite voisine de 0,02 pour les plus gros galets.

Une campagne de mesures analogue sera effectuée prochainement sur le Rhône, en aval d'Avignon.

2.2. Méthode directe

Nous étudions l'adaptation à la mesure du débit solide par charriage des différentes méthodes utilisées couramment pour la mesure des débits liquides à l'aide de traceurs.

Mais l'utilisation de ces méthodes implique que les brassages latéral et vertical dans la couche de matériaux participant au charriage soient suffisamment énergiques pour que l'homogénéité du mélange en traceurs soit satisfaisante en aval de leur point d'immersion.

Pour vérifier les conditions dites de «bon mélange» nous poursuivons, au Laboratoire, des études sur le mécanisme de dispersion de traceurs. Ces sédiments progressent habituellement sur le fond de façon discontinue par déformation de rides ou de dunes. A un instant donné, seules les particules superficielles du lit participent au charriage; les grains de matériau enfouis sous une ondulation du lit restent en place jusqu'à ce que, celle-ci ayant progressé vers l'aval, ils soient mis à découvert. Les brassages longitudinal et vertical ne sont donc pas uniformes.

Des essais de longue durée ont été effectués dans un canal fixe horizontal long de 45 m et large de 1,5 m. Trois catégories de matériau de fond ont été utilisées; des galets naturels concassés de diamètres moyens respectifs 0,52 et 0,3 mm (poids spécifique 2,6 g/cm$^3$) et du noyau d'abricot broyé de diamètre moyen 2,72 mm (poids spécifique 1,33 g/cm$^3$).

Pour ces premiers essais en Laboratoire les traceurs lumineux furent préférés aux traceurs radioactifs en raison de leurs facilités d'utilisation. Le traceur était constitué par un échantillon du matériau de fond utilisé, marqué à la peinture luminescente; ce traitement n'affecte aucune-ment les caractéristiques géométriques et physiques des grains, c'est-à-dire que le traceur se comporte exactement comme le milieu à étudier.

A chaque catégorie de matériau correspondait une série d'essais avec traceurs, les conditions hydrauliques étant inchangées pour une même série. Les caractéristiques d'écoulement étaient choisies de telle sorte que le charriage se fasse sous forme de dunes ou de rides (le débit solide en suspension était négligeable). Des essais préliminaires permettaient de déterminer par approximations successives le débit solide requis pour maintenir l'équilibre des fonds mobiles correspondant au régime hydraulique uniforme.

Divers modes d'immersion des traceurs furent essayés:

a) immersion ponctuelle superficielle dans l'axe du canal en eau, mais sans écoulement,
b) immersion par saignée transversale superficielle, le canal étant à sec,
c) immersion par plaques superficielles dans les creux des rides, le canal étant à sec.

Le premier mode d'immersion conduit à une grande dispersion du traceur en début d'essai et fut abandonné pour cette raison.

Des carottages dans le matériau de fond mobile, de profondeurs suffisantes pour couvrir la hauteur du matériau intéressé par le charriage, permettent de suivre la progression des traceurs. Chaque carottage donne ainsi l'épaisseur de la couche de matériau en mouvement et la concentration moyenne en traceur sur cette verticale, après comptage des grains marqués en lumière ultraviolette.

La hauteur moyenne de matériau en mouvement déterminé par les carottages effectués en fin d'essai à proximité du profil d'immersion des traceurs correspond sensiblement à l'amplitude moyenne des ondulations du lit.

La comparaison des concentrations verticales avec les concentrations superficielles ne fait pas apparaître la moindre corrélation entre la répartition des traceurs en surface et en profondeur.

Les possibilités d'application de la méthode des pics (méthode d'Allen) et de la méthode d'intégration ont été examinées à la suite de ces essais. Rappelons que pour la méthode des pics l'on détermine l'évolution spatiale du nuage de traceurs alors que pour la méthode d'intégration ou de numération totale l'on détermine en un profil donné l'évolution temporelle des concentrations en traceur.

a) Dans la première méthode, les carottages successifs permettent de déterminer la vitesse moyenne de progression du barycentre du nuage de traceurs qui, associée à l'épaisseur moyenne de la couche de matériau en mouvement, permet de calculer le débit solide.

Cette méthode fait apparaître l'importance de la notion d'équilibre du lit correspondant à un régime hydraulique permanent uniforme donné. Cette condition nécessite à elle seule de longs essais préliminaires de réglage. Si, en effet, elle n'est pas satisfaite, le fond moyen évolue en cours d'essais; le fond s'exhaussant en aval par exemple, une part importante de traceurs peut se perdre en profondeur et ne plus participer au charriage.

b) La zone de concentration maximale en traceur se déplace très lentement, si bien que la vérification de la méthode d'intégration conduit à des essais de très longue durée mais n'implique pas l'utilisation de canaux de grande longueur à la différence de la méthode précédente.

La vérification de la méthode d'intégration a été effectuée en plusieurs profils transversaux du canal; mais, du fait de la lenteur de progression du point de concentration maximale, l'on se trouve limité habituellement à des profils proches de la zone d'immersion des traceurs. Dans le cas d'un transport solide avec dunes, l'insuffisance du brassage sur une aussi courte distance explique alors la dispersion obtenue dans les valeurs de débit solide ainsi calculées. Par contre, si le lit est couvert de rides, le brassage du traceur dans le matériau de fond se fait plus rapidement et la méthode d'intégration conduit à des résultats très satisfaisants.

Le problème de la dispersion de traceurs radioactifs dans un écoulement à trois dimensions devait faire l'objet d'une étude en nature sur la Loire en aval de Beaugency, étude contrôlée par un modèle réduit. Les con-
ditions climatiques ayant retardé ces essais, il n'est pas encore possible d'en tirer des conclusions.

Simultanément nous étudions les procédés de marquage et de détection nécessaires à l'emploi des traceurs radioactifs tant pour l'expérimentation sur modèle réduit que pour les mesures en nature. Un dispositif spécial permet d'étudier avec beaucoup de souplesse la réponse d'un détecteur à scintillation à une source ponctuelle immergée dans le sable à différentes distances et profondeurs. Les mesures ainsi effectuées permettront de définir différents types de sondes à scintillateurs multiples adaptés à la mesure quantitative des débits de charriage. Des procédés d'exploration de la distribution verticale des grains actifs dans la couche mobile sont également étudiés, soit par mesure sur carottes ramenées sur le bateau-laboratoire, soit directement in situ par sondage vertical au moyen d'un détecteur collimaté.

Les procédés quantitatifs de mesure du débit de charriage exigeant, pour des raisons de statistique, un nombre de grains actifs beaucoup plus élevé que les simples mesures jusqu'ici pratiquées, de vitesses moyennes et de directions, des perfectionnements ont été apportés aux procédés de marquage en surface du sable naturel au moyen de différents isotopes*.

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G. H. LEAN: The difficulty in Mr. Hours' suggestion for using a few very highly active grains rather than a large number of less active particles is that the high residence time of the particles in the ripples, which is a fairly random quantity, means that the statistics for just a few particles will be unsatisfactory. It is therefore necessary to use a large number of them.

R. HOURS: With regard to this statistical problem, I might add that, for reasons of safety and simplicity of production, we have thus far used radioactive glass almost exclusively in France. However, when the purpose is to make a quantitative study and specifically, to measure vertical tracer distribution, the number of grains used must be very large. In this case it becomes difficult — at least under natural conditions — to use the ground glass method, because the irradiation of tens of kilograms of material in a reactor is no easy matter. We have therefore been trying for nearly a year to improve the methods for the surface labelling of sediments. In some cases, we have obtained comparatively satisfactory results, especially for depositing iridium, with which we have obtained labelled material which is entirely stable over a period of time. We have not yet mastered the procedure sufficiently well to be able to achieve a surface labelling such that the specific activity of the particles will be constant. That is to say at present the specific activity of our particles is no longer proportional to the surface, but it is not yet entirely proportional to the volume. As far as the future of these methods is concerned, I think it is essential to be able to achieve sufficient control over surface labelling to ensure that, when mass transports are involved, the activity deposited on the grains will be proportional to their mass.

P. C. LÉVÊQUE: I would like to draw attention to the fact that, while everything connected with laboratory testing is obviously of importance (and this is especially true of hydraulics laboratories, where even the simplest parameters can be defined with accuracy), in the case of the Rhone and other in situ experiments on the dispersion and movement of pebbles, it would appear necessary to formulate a satisfactory definition of water density. In the case of the Rhone, the floods, which originate in Lake Geneva, consist of water which can be described as "clean". I do not think that the findings for such a river can be properly extrapolated to certain rivers in arid or semi-arid countries, or even to those of the Mediterranean basin, where the load of material in suspension increases the density of the water and imparts a considerable competence to the current, particularly at the onset of the flood. (The competence of such currents with densities of 1.05 to 1.10 is infinitely higher than that of clean water like that of the Rhone). I think that studies with labelled pebbles using slightly higher water densities would yield surprising results with respect to the transport capacity of the streams in question.

R. HOURS: Not being a hydraulic engineer, I have no views on this subject. However, the question brings to my mind the situation in certain African wadis, in which the flow can increase by a factor of 100 within a few hours during periods of flood. At such times, these wadis transport huge amounts of suspended matter and become veritable rivers of mud. We
have been consulted on occasion in connection with the measurement of liquid flow rates in these wadis during floods. It would appear that in such cases methods based on the use of radioactive tracers, as described by Mr. Smith and Mr. Guizerix in the discussion following the paper "Use of radioisotopes for open-channel flow measurements", are much superior to those employing chemical tracers or mechanical means, such as a current meter. The latter instrument practically ceases to operate under these conditions.

M. KATO: You mentioned having done an experiment on the vertical distribution of a radiotracer in sand. Was this a model experiment or a field experiment?

R. HOURS: It was simply a laboratory experiment in which the concentrations were measured by means of fluorescent tracers, vertical cores were taken in the channel bed during transport and the distribution of the fluorescent particles was recorded. Measurement of the vertical distribution of the particles under field conditions is another matter and we have not developed any equipment for such work, although we have considered ways in which it could be done. Among the ideas we have studied is that of immersion of a hollow tube, inside which a scintillation counter with a very narrow crystal of given collimation would be lowered. Another technique now used in the Netherlands and at Berkeley, would be that of taking a core, stabilizing it by means of a gel, and then doing a study — either in the laboratory or on board the laboratory-ship — of the distribution, both vertical and lateral. We are also studying the possibilities of energy discrimination, along the lines proposed by Arlman and Svašek, using the two photoelectric peaks of scandium-46. For this work, we envisage the use of two gamma emitters of very different energies, which could be thulium and irridium, or irridium and tantalum-182. However, I am somewhat pessimistic concerning the future of those methods which, although of great theoretical interest, are rather difficult to implement in practice. I am afraid that the Compton scattering in the water will considerably impair the validity of the measurements.

D. B. SMITH (Chairman): Could you indicate how many pebbles of 500 μc activity were used in your investigation in the Rhone? Are any particular health precautions required by the French authorities for this type of investigation?

R. HOURS: The study is described in detail in reference [9] in the paper. There were about 400 labelled pebbles, comprising some 30 large ones, 250 small ones and 125 of medium size. I am aware that our United Kingdom colleagues generally use much weaker sources, of the order of 15-20 μc, but on the basis of our experience we prefer to use considerably higher activities to facilitate detection. I do not think that such activities can represent a serious danger in a river like the Rhone.

We had no particular difficulties with the French health authorities on this occasion, since it was not considered likely that the pebbles would reach a water area where fishermen, for example, might walk on them. Moreover, the experiments were carried out in December. Nevertheless, since the number and scope of experiments of this type are increasing, Mr. Courtois and I are working at Saclay on a set of regulations relating to the use of radioactive tracers in sediment transport studies. Activities up to 500 μc do not seem unreasonable for pebbles if they are not expected to accumulate
in dry places where people would be present for long periods of time. Moreover the pebbles would generally be widely scattered. As far as the limitation of activity on beaches is concerned, we are proposing a value comparable to that of a pure granite beach. Granite of high concentration is an extremely radioactive substance and it is therefore very comforting to note that the concentrations found for granite are much higher than all the maximum permissible concentrations given in the tables for the materials which we use. On the other hand, limitation of the activity of an individual particle or an individual pebble, as well as that of the entire activity used, is a much more complex problem. This problem takes one form if one is considering, in the case of sand, the danger from consumption of water containing radioactive sand in suspension. It takes another form when one considers the question of sand in suspension in the air upon labelling of beaches. In these circumstances it is difficult to bring the maximum permissible concentrations (MPC) into agreement. One has also to consider the hazard, on the one hand, of gamma radiation (originating with external labelling procedures), and on the other hand, of beta radiation (occurring at the organic level and originating with the ingestion of irradiated water). What Mr. Courtois and I are seeking is a formula which would take account of both types of exposure.

D.B. Smith (Chairman): As you say, we use very weak sources (10-15 μc per pebble) in the United Kingdom. We would like to use 500 μc per pebble, but I am sure that our health authorities would not be very cooperative.

R. Hours: The health authorities must be presented with a project accompanied by figures and full justificatory data, otherwise their invariable response is to divide the MPC's by 10—or by 30!—and to give their consent if the resultant concentration is not exceeded. In other words, they take no risks whatever. This conservative attitude is a normal, human and safe reaction at the start, but I think that in the future we shall need more realistic regulations on the labelling of both water and sediments.

J.F. Honstead: On the basis of experience in France, how does the hazard of using sodium dichromate in flow measurements compare with that of using radioisotopes?

R. Hours: I myself have never gone into this question very thoroughly and do not know what concentration of sodium dichromate is considered dangerous to fish. However, I understand from Mr. Guizerix that the concentrations used by Electricité de France in daily river flow measurements in the Alps and Pyrenees may be 100 times less than this dose, but I am not too sure of this figure. I would assume that the concentrations of Br⁴² which are used in France for the same purpose are less dangerous to fish by several orders of magnitude. I think that, wherever possible, reference to the natural radioactivity of a highly radioactive medium like granite is one of our most telling arguments when we apply to the health authorities for more liberal levels. We have done this in France in connection with labelling techniques to determine blast furnace wear and specific activities present in melt and slag.

J.F. Honstead: It seems to me that the relative hazards of dichromate and radioactive isotopes are not always presented quite fairly. The control limit for dichromate is quoted as 1% of the amount which is con-
sidered "hazardous", i.e. which would prove fatal to fish. On the other hand, the limit selected for radioisotopes is a fraction of that recommended by the International Commission on Radiological Protection for drinking water, which is already far below the level that would result in any detectable biological damage.

R. HOURS: I agree that while it is possible and proper to start work very slowly and conservatively, we should go a little faster now. At the same time we are arguing for more realistic limits. We must try to understand the point of view of the health authorities who, unlike us, bear the ultimate responsibility for anything that may happen.
RADIOTRACER EXPERIMENTS ON LITTORAL DRIFT IN JAPAN

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Abstract — Résumé — Аннотация — Resumen

RADIOTRACER EXPERIMENTS ON LITTORAL DRIFT IN JAPAN. In this country, since the first experiment with radioactive tracers was carried out in 1954 to determine the movement of littoral drift at the beginning of harbour construction at Tomakomai, such tracer techniques have been applied to five or more other places over the past seven years. The present paper summarizes these experiments.

First, the experimental methods of the tracer technique are described: the preparation methods and the properties of the radioactive glass sand, the depositing methods and equipment for depositing the radiotracer at a definite point and as point-like as possible on the sea bed, the kinds and the characteristics of the underwater detectors used, the consideration of radiation protection, and some incidental tracer techniques in the field experiment.

Then, some typical examples of the experimental results obtained are shown and discussed from the following standpoint of coastal engineering: (1) The experiment on the Kashimanada Coast carried out as a preliminary investigation for the construction of the harbour; (2) That on sand movement around the breakwater of the Tomakomai Harbour now under construction; (3) That for investigation of sand deposition in the Niigata Harbour basin; and (4) The investigation of erosion of the coast in the western part of Niigata.

In addition, the experiment for determination of the vertical distribution of radioactive sand in the sea bed, the model experiment in a wave channel in order to make clear modes of sand movement and an unexpected experimental obstacle because of accumulated fall-out on the bed of the estuary of Shinano River located at the Niigata Coast are described.

EMPLOI DES RADIOINDICATEURS AU JAPON POUR L’ÉTUDE DU MOUVEMENT DES SABLES DU LITTORAL. En 1954, les radioindicateurs ont été utilisés pour la première fois au Japon pour déterminer le mouvement des sables du littoral, lors de la construction du port de Tomakomai; cette méthode a été appliquée au cours des sept dernières années dans l’étude de plusieurs emplacements. Le mémoire expose ces expériences.

Il décrit en premier lieu les conditions expérimentales d’utilisation des radioindicateurs: préparation et propriétés du sable de verre radioactif, méthodes et dispositifs de dépôt des radioindicateurs en un point aussi défini que possible du fond de la mer, nature et caractéristiques des détecteurs sous-marins utilisés, problèmes de protection radiologique et utilisations occasionnelles des radioindicateurs au cours des expériences.

Les auteurs passent ensuite en revue les résultats de quelques expériences typiques, qu’ils analysent du point de vue des problèmes techniques de génie portuaire: a) expérience sur la côte de Kashima, entreprise au titre de l’enquête préliminaire à la construction du port; b) étude du mouvement des sables au voisinage de la digue du port de Tomakomai, actuellement en construction; c) étude du dépôt de sables dans la rade de Niigata; d) étude de l’érosion de la côte à l’ouest de Niigata.

Outre les expériences ci-dessus, les auteurs décrivent une expérience qui tendait à déterminer la distribution verticale des sables radioactifs du fond et une expérience sur maquette réalisée dans un canal hydro-
dynamique soumis à l'action des vagues, en vue d'expliquer le mouvement du sable; ils indiquent aussi que l'accumulation de retombées au fond de l'estuaire du Shinano, sur la côte de Niigata, a empêché de poursuivre l'expérience en cours à cet endroit.

ОПЫТЫ С РАДИОИНДИКАТОРАМИ ПО ИЗУЧЕНИЮ ДВИЖЕНИЯ ПРИБРЕЖНЫХ НАНОСОВ В ЯПОНИИ. Со времени первого эксперимента с радиоактивными индикаторами, проведенного в 1954 году с целью определения движения прибрежных наносов перед сооружениями гавани в Томакомаи, за прошедшие семь лет такие индикаторные методы применялись в Японии в пяти или более других местах. Данный доклад обобщает эти эксперименты.

Во-первых, описываются экспериментальные методы использования индикаторов, то есть методы подготовки и свойства радиоактивного кварцевого песка, методы и приборы для помещения радиоиндикатора в определенной точке и, насколько это возможно; в ограниченном месте морского дна, виды и характеристики использовавшихся подводных детекторов, вопросы радиационной защиты и некоторые особые индикаторные методы, использовавшиеся в полевом эксперименте.

Во-вторых, насколько позволяют полученные результаты экспериментов, приводятся некоторые типичные примеры и обсуждается с точки зрения береговой техники следующее:

1) опыт у побережья Касимо, проводившийся в качестве предварительного исследования для строительства гавани; 2) исследование движения песка вокруг волнореза гавани Томакомаи, которая находится в процессе строительства; 3) исследования отложений песка в бассейне гавани Ниигата; 4) исследование эрозии берега в восточной части Ниигата.

Кроме того, описываются опыт по определению вертикального распределения радиоактивного песка на морском дне, опыт по модели каналов с искусственными волнами с целью выяснения движения песка и причины неудачи опыта в связи с большими отложениями песка в ложе устья реки Синано, побережье Ниигата.

EXPERIMENTOS RELATIVOS AL ARRASTRE DEL LITORAL REALIZADOS CON INDICADORES RADIOACTIVOS EN EL JAPON. Desde que se realizó en 1954 el primer experimento con indicadores radiactivos para determinar el arrastre del litoral al iniciarse la construcción del puerto de Tomakomai, se ha aplicado el mismo procedimiento en otros cinco lugares del Japón en el curso de los últimos siete años. En la memoria se examinan dichos experimentos.

En primer lugar se describen los procedimientos seguidos en la aplicación de los indicadores radiactivos, es decir, los métodos de preparación y las propiedades de las arenas radiactivas, los métodos de inyección y los aparatos empleados para depositar el indicador en un punto determinado y con la mínima dispersión posible en el fondo del mar. Asimismo se examinan los tipos y las características de los detectores submarinos utilizados, los problemas relacionados con la protección radiológica y algunas técnicas auxiliares aplicadas en los experimentos in situ.

En lo que respecta a los resultados experimentales obtenidos, se citan a continuación algunos ejemplos típicos que se estudian desde el punto de vista de la ingeniería portuaria, a saber: a) El experimento realizado en la costa de Kashima como investigación preliminar para construir el puerto; b) el experimento efectuado para estudiar el movimiento de las arenas en torno al rompeolas del puerto de Tomakomai, actualmente en construcción; c) el experimento llevado a cabo con objeto de estudiar la sedimentación de arena en el puerto de Niigata; d) investigación de la erosión de la costa en la parte occidental de Niigata.

Asimismo se describen el experimento realizado para determinar la distribución vertical de arenas radiactivas en el fondeo del mar, el experimento ejecutado con maquetas en un canal hidrodinámico que estudia las modalidades del movimiento de la arena, así como las dificultades imprevistas que en la realización de un experimento surgieron a raíz de la acumulación de precipitaciones radiactivas en el lecho del estuario del rio Shinano, situado en la costa de Niigata.

1. INTRODUCTION

In Japan the radiotracer technique proved highly suitable for coastal engineering in the first experiment on the Tomakomai Coast [1, 2] in 1954, it has been applied at several other places the locations of which are shown in Fig. 1. Most of these experiments have been carried out for purposes
of harbour construction on a sandy coast and others on beach erosion and sand deposition. The experimental measures are outlined in Table I. Three research groups, at the University of Tokyo, the Ministry of Transportation and the Japan Atomic Energy Research Institute, in co-operation with the respective offices at the sites of the experiments, have engaged in this research and have obtained useful data for planning and design in coastal engineering.

In the present paper techniques used in these experiments and some representative work in this country are considered briefly.

The radiation hazard problem is not discussed here, because the safety of such experiments is now being accepted generally as a result of extensive experience gained in the world to date.

2. METHODS OF EXPERIMENTS

2.1. Preparation of radiotracers

Until three years ago radioactive glass sand as a tracer had been prepared with such processed radioisotopes as Zn$^{65}$ and Co$^{60}$, which were used many times in experiments at the Tomakomai Harbour and the Irako Harbour. Eventually use was made of the irradiation methods of JRR-1 (Japan Research Reactor No.1, a water-boiler type with 50 kW); thereby either Co$^{60}$ or Sc$^{46}$ glass sand is now being prepared. As is well known, the water-boiler-type
## TABLE I
THE FIELD EXPERIMENTS WITH RADIOISOTOPES PERFORMED IN JAPAN

<table>
<thead>
<tr>
<th>Coast</th>
<th>Date</th>
<th>Isotope</th>
<th>Deposition water depth (m)</th>
<th>Number of depositions</th>
<th>Activity deposited at a point (mc)</th>
<th>Measuring apparatus</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tomakomai</td>
<td>1954-1955</td>
<td>Zn⁶⁵</td>
<td>2, 3, 6, 9 and foreshore</td>
<td>6</td>
<td>14-32</td>
<td>Scintillation</td>
</tr>
<tr>
<td></td>
<td>1956-1961</td>
<td>Co⁶⁰</td>
<td>3-14</td>
<td>17</td>
<td>5-20</td>
<td>G-M</td>
</tr>
<tr>
<td></td>
<td>1962</td>
<td>Sc⁴⁵</td>
<td>7</td>
<td>1</td>
<td>8</td>
<td>Scintillation</td>
</tr>
<tr>
<td>Kaike</td>
<td>1957</td>
<td>Zn⁶⁵</td>
<td>2-6</td>
<td>3</td>
<td>50-60</td>
<td>G-M</td>
</tr>
<tr>
<td></td>
<td>1958-1959</td>
<td>Sc⁴⁶</td>
<td>2-6</td>
<td>7</td>
<td>50-60</td>
<td>G-M</td>
</tr>
<tr>
<td>Irako</td>
<td>1957-1958</td>
<td>Co⁶⁰</td>
<td>2-10</td>
<td>4</td>
<td>2-19</td>
<td>G-M</td>
</tr>
<tr>
<td>Fukue</td>
<td>1959-1960</td>
<td>Co⁶⁰</td>
<td>0-9.5 and foreshore</td>
<td>7</td>
<td>3-6</td>
<td>G-M</td>
</tr>
<tr>
<td></td>
<td>1961</td>
<td>Sc⁴⁶</td>
<td>foreshore</td>
<td>2</td>
<td>5</td>
<td>G-M</td>
</tr>
<tr>
<td>Isohama</td>
<td>1960-1961</td>
<td>Co⁶⁰</td>
<td>4.5 and 10</td>
<td>4</td>
<td>2-35</td>
<td>G-M</td>
</tr>
<tr>
<td>Kashimanada</td>
<td>1962</td>
<td>Co⁶⁰</td>
<td>5-15</td>
<td>13</td>
<td>30-60</td>
<td>G-M</td>
</tr>
<tr>
<td></td>
<td>1962</td>
<td>Sc⁴⁶</td>
<td>2-6 and foreshore</td>
<td>4</td>
<td>10-60</td>
<td>G-M</td>
</tr>
<tr>
<td>Niigata</td>
<td>1959</td>
<td>Zn⁶⁵</td>
<td>6-6.5</td>
<td>2</td>
<td>about 80</td>
<td>G-M</td>
</tr>
<tr>
<td></td>
<td>1961</td>
<td>Co⁶⁰</td>
<td>10-15</td>
<td>8</td>
<td>20-230</td>
<td>G-M</td>
</tr>
<tr>
<td></td>
<td>1962</td>
<td>Sc⁴⁶</td>
<td>3-6</td>
<td>5</td>
<td>80</td>
<td>G-M</td>
</tr>
<tr>
<td></td>
<td>1962</td>
<td>Co⁶⁰</td>
<td>3-15</td>
<td>11</td>
<td>10-60</td>
<td>Scintillation</td>
</tr>
</tbody>
</table>
reactor is not suitable for irradiation over long periods of time or for large amounts of target material. This difficulty will be overcome with the beginning of use of JRR-2 and JRR-3 which is expected in the near future.

The total activity of radiotracer used at a point was of the order of several tens of mc and up to 80 mc, with which the tracings could range up to about 100 m. Hitherto, such a scale of tracings has been enough for the purposes of the experiments shown in Table I, with one exception when as much as 160 mc was used for observation of the sea bed for a long time. Accordingly, the specific activities of the radiotracers used, some examples of which are shown in Table II, were of the order of several tens of mc/kg.

<table>
<thead>
<tr>
<th>Tracers with Co&lt;sup&gt;60&lt;/sup&gt; (mc/kg)</th>
<th>Tracers with Sc&lt;sup&gt;46&lt;/sup&gt; (mc/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tomakomai</td>
<td>20*</td>
</tr>
<tr>
<td>Kashimanada</td>
<td>40</td>
</tr>
<tr>
<td>Niigata</td>
<td>40</td>
</tr>
<tr>
<td></td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>60</td>
</tr>
<tr>
<td></td>
<td>50</td>
</tr>
</tbody>
</table>

* Tagged with processed radioisotope [2]

As a matter of course, the radiotracer must bear a close resemblance to the bed material in grain-size distribution and in specific gravity. The specific gravity of natural sand of any coast investigated was 2.65. On the other hand, soda glass has been used as a base material for incorporation with cobalt or scandium; its specific gravity is about 2.5. Accordingly, the adjustment of the specific gravity was made chiefly in the two following ways: cobalt glass was prepared by balancing the quantity of cobalt and scandium glass by adjustment of the addition of lead; some examples of chemical composition of the glass are shown in Table III.

Then these glasses were ground mechanically to the grain-size distribution which is specified for the particular place of observation. In practice two kinds of representative mean grain size in the inshore and offshore zones are specified, and they are used regardless of depth in the respective zones.

2.2. Method of depositing radiotracer on the sea bed

First, it is necessary to deposit the radioactive glass sand at a definite point on the sea bed to obtain a point source insofar as possible. Also, the procedure must be done safely and at a distance from boats, and hence the depositing equipment suited to above-mentioned purposes must be elaborated.
TABLE III

SOME EXAMPLES OF WEIGHT RATIO OF RAW MATERIALS FOR GLASS SAND TO BE IRRADIATED

<table>
<thead>
<tr>
<th></th>
<th>SiO₂</th>
<th>CaCO₃</th>
<th>Na₂CO₃</th>
<th>NaNO₃</th>
<th>Pb₄O₈</th>
<th>Co₁O₃</th>
<th>Sc₂O₃</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co glass¹ ² ³</td>
<td>56.5</td>
<td>15.1</td>
<td>18.8</td>
<td>1.6</td>
<td>-</td>
<td>7.9</td>
<td>-</td>
</tr>
<tr>
<td>Sc glass¹</td>
<td>57.7</td>
<td>18.2</td>
<td>20.3</td>
<td>1.6</td>
<td>2.0</td>
<td>-</td>
<td>0.2</td>
</tr>
<tr>
<td>&quot; ²</td>
<td>56.0</td>
<td>20.0</td>
<td>19.6</td>
<td>1.6</td>
<td>2.4</td>
<td>-</td>
<td>0.4</td>
</tr>
<tr>
<td>&quot; ³ ⁴</td>
<td>61.6</td>
<td>16.4</td>
<td>20.3</td>
<td>1.6</td>
<td>-</td>
<td>-</td>
<td>0.7</td>
</tr>
</tbody>
</table>

¹ Tomakomai
² Kashimanada
³ Niigata
⁴ 2.5 in specific gravity, prepared intentionally smaller than that of natural sand.

For the first experiment a simple depositing method, by a diver, was adopted on the Tomakomai Coast [1, 2]. Since 1958 this method has been superseded by improved equipment [3], as shown in Figs. 2 and 3. With this equipment radioactive glass sand together with soap suds is placed in a cylinder chamber shielded with lead walls which is then sealed with polyethylene sheet. After it is let down to the sea bed from a boat, a plumb weight is dropped along the messenger wire, activating the cam mechanism. Thereby a spring acts to push a piston, and in that moment the polyethylene sheet is cut by a knife-edge and the piston pushes out the radioactive glass sand.

The equipment used at Kashimanada and Niigata has a bottom wall made of glass. That used at Kashimanada (Figs. 4 and 5) is let down into the sea with a rope. When its bottom touches the sea bed, its upper part containing tracer material drops down by its own weight and the bottom of the beaker or the glass plate is broken by impingement against projections on the base. Thus, the glass sand is discharged and deposited on the sea bed inside the wall of the base cylinder.

The equipment used at Niigata (Figs. 6 and 7) is composed of a cylindrical vessel (A) and its rack (B), both made from vinyl chloride except for the lower semispherical part which is made from glass and holds tracer material. At the injection point, a steel plate is sunk beforehand, then the vessel is let down by a lead wire onto the plate. Thereafter, a plumb weight is dropped onto the vessel to break the bottom glass and to discharge the tracer completely on the plate.

2.3. Tracing method

To measure the radioactivity on the sea bed it is necessary to have some underwater detecting apparatus which is sensitive to gamma rays,
Fig. 2

Depositing equipment used at Tomakomai
(Dimensions in mm)

(1) Messenger wire, (2) hammering plumb, (3) cap,
(4) clamping screw, (5) handle to compress spring,
(6) claw hook, (7) shielding lead, (8) piston,
(9) charging space for radioactive glass sand,
(10) annular cutter, (11) vinyl sheet, (12) leaden
cover for shielding, and (13) stand
Fig. 3

Depositing equipment used at Tomakomai
Fig. 4
Left: Charging radioactive sand in the depositing equipment used at Kashimanada.
Right: Submerging the depositing equipment.

Fig. 5
Depositing equipment used at Kashimanada
(Dimensions in mm)
(a) for small volume
(b) for large volume
Fig. 6
Depositing equipment used at Niigata
(Dimensions in mm)

Fig. 7
Depositing equipment used at Niigata
withstands considerable water pressure, is easy to operate, is stable for work in the sea and has other necessary functions.

In the early stages of the experiment, underwater detectors installed with a bundle of two or three large G-M tubes in the probe case, which was water-tight and made from stainless steel, were used because the G-M counter is robust and its sensitivity increases considerably with its size (Fig. 8).

Pulses were fed to a scaler or ratemeter through a coaxial cable 30 m in length. This allowed a sufficient length of cable for any depth and for the pitching and rolling of the boat.

The background of the G-M counter, however, was considerably higher on the sea bed, owing chiefly to the glass envelope. Therefore, an underwater detector equipped with a bundle of three small G-M tubes composed of metal envelopes was used in the experiment at Niigata. (Fig. 9).

On the other hand, it became clear by some basic research that a scintillation counter is more sensitive than a large G-M counter even taking into account the high background of the sea bed; moreover its robustness is improved by transistorizing. A few years ago a G-M type of underwater detector was replaced with a scintillation one. Especially in Tomakomai, a new type of scintillation detector equipped with an adapter which is conical and hollow was exploited (Fig. 10), thereby improving the detecting sensitivity by a factor of about two. In addition, by assorting the pulse-height analysing circuit with the ratemeter the detector was able to select a certain energy band and to decrease the background.

3. SOME EXAMPLES OF THE RESULTS OF FIELD OBSERVATIONS

3.1. Purpose of the use of radioisotopes in the study of littoral sand drift

The field investigations for the survey of littoral sand drift may be classified as follows:

(1) Presumption of the pattern of sand drift by the observation of wave and longshore currents.
(2) Presumption of the limit of area and degree of sand movement by the periodical surveying of the sea bed profile with a certain area.

(3) Study of the source of bottom sand and the direction of longshore movement by the analysis of the grain size, uniformity and mineral content of the sand taken from the shoreline.
(4) Study of the actual movement of the sand grains by radioisotopes. Observation by radioisotopes is a more direct method as compared with other procedures. It gives a record of actual sand movement, especially the movement under local disturbances. It is also expected to be used for the quantitative measure of sand transport. From these points of view some recent examples of observations using radioisotopes carried out in this country are described in this section.

3.2. Field observation on the Kashimanada Coast

3.2.1. Purpose

Field investigations have been performed by S. Sato, one of the authors, since 1961 on the construction of the Kashima Industrial Harbour on the Kashimanada Coast which faces the rough sea area of the Pacific Ocean [4, 5]. This coast is, as shown in Fig. 11, a monotonous sandy beach stretching...
about 70 km from Isohama to Choshi and the sand movement in this area needs clarification for the proper planning of the harbour. Therefore, the sand drift on the sea bottom as well as waves, currents and bottom profiles using radioactive glass sand have been observed. The primary aims of the investigations with radioisotopes are, in this case, to find the limit of water depth for the sand movement in the offshore zone and the difference in the rate of sand transport for the various depths of water. The used tracer was radioactive glass sand containing Co\(^{60}\). It was 40 mc/kg in specific activity, 2.65 in specific gravity and 0.15 mm in mean diameter, including grains from 0.06 to 0.25 mm.

3.2.2. An example of the results of observations

The field observations were carried out at various water depths up to 15 m. An example of the results of a series of observations of the patterns of sand movement at 5- and 11-m depths follows.

Glass sand was deposited at point No. 1 (Fig. 11, depth of 11 m and 1500 m from the shoreline) at about 10 o'clock on 28 April 1962 and at point No. 2 (depth of 5 m and 600 m from the shoreline) at nearly the same time the next day. Activity of the deposited sand was 60 mc for each point.

Figs. 12 and 13 show the time-change of measured equi-count lines. The inscribed numbers on the lines are the values of the counting rate in counts per second which were measured by the underwater detector and subtracted from the background, 16 cps. The intersection of the NS and EW axes in the figure shows the depositing point. The characteristics of significant wave measurement by an underwater wavemeter installed on the sea bed at a 10-m depth are shown in Fig. 14. The following conclusions are derived from these figures.

(1) The advancing direction of the waves on the two depositing days, 28 and 29 April, were nearly the same, while the equi-count lines spread towards NW at No. 1 and SSE at No. 2. This must result from the fact that the sand was disturbed by the deposition flows in the direction of the bottom current which was different between the two points of 11-m and 5-m depths.

(2) On 10 May after the incidence of successive waves which were supposed to be up to 1.4 m in height and of 12 s duration data were obtained. The result showed that the lower count lines extended to WNW but that of 100 cps was nearly similar to that of the depositing day at No. 1, while at No. 2, all count lines extended in the direction normal to the shoreline and the activity decreased remarkably.

(3) At the observation on 19 May after the wave of 1.7 m in height and 6-8 s in period, the pattern of count lines did not change from that of the preceding measurement at No. 1, while the activity could not be counted at No. 2. This shows that the bottom sand is completely scattered by waves of such order in an area shallower than 5 m.

(4) The result of observations on 27 May at No. 1 showed that the bottom sand is completely scattered by a wave 4 m in height and of 8-10 s duration even on a bottom of 11-m depth.

(5) The wave directions shown in Fig. 14 were obtained for the offshore by visual observation from a tower about 26 m in height. From the refraction
Fig. 12

Equi-count lines (cps) at point No. 1 in Fig. 11
Fig. 13

Equi-count lines (cps) at point No. 2 in Fig. 11
diagram for the wave which advanced from ESE it is predicted that this wave takes the direction of E on the contour line of 5-m depth. This property shows that the main directions of the movement of glass sand on 11-m and 5-m depths nearly coincide with those wave directions.

(6) At the observation on 10 May, the shifted distance of the centre of activity at No. 2 measured from the depositing point was twice that at No. 1, and the cumulative amount of activity at No. 2 was about one third of that at No. 1. Therefore, the rate of sand transportation at No. 2 may be roughly estimated to be six times that at No. 1.
3.3. Field observations in Niigata

3.3.1. Introduction

S. Sakagishi, one of the authors, made an investigation of sand transport using radioisotopes in the Niigata district for a year, August 1961 to July 1962 [6, 7]. This investigation includes two series of observations. The first one was carried out at the Niigata Harbour basin situated at the mouth of the Shinano River, and used radioactive glass sand containing Co$^{60}$ to determine from where sand mainly invades the harbour. Another observation was performed on the west coast of Niigata, using glass sand containing Sc$^{46}$; its purpose was to learn the actual direction of transport of bed materials in the offshore or the inshore area (Fig. 15).

The total activities of the deposited sands are listed in Table IV.

### Table IV

<table>
<thead>
<tr>
<th>No. of experiment</th>
<th>Nuclide</th>
<th>Total activity (mc)</th>
<th>Total weight (kg)</th>
<th>Places deposited</th>
<th>Date of deposition</th>
<th>Duration of observation (months)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Co$^{60}$</td>
<td>140</td>
<td>3.5</td>
<td>5 points at river mouth</td>
<td>4-5 Aug. 1961</td>
<td>3</td>
</tr>
<tr>
<td>2</td>
<td>Co$^{60}$</td>
<td>500</td>
<td>12.6</td>
<td>3 points at river mouth</td>
<td>29-30 Oct. 1961</td>
<td>6</td>
</tr>
<tr>
<td>3</td>
<td>Sc$^{46}$</td>
<td>400</td>
<td>8.0</td>
<td>5 points at west coast</td>
<td>7-8 Apr. 1962</td>
<td>2</td>
</tr>
</tbody>
</table>

3.3.2. Results of observations

After depositing the tracer material at a fixed point, an underwater detector measured the activity around the depositing point and from the results of measurements the equi-count lines were drawn as in the case in the preceding section.

To illustrate the shifting of equi-count lines, the contour maps obtained from one series of observation are shown in Fig. 16. This observation was performed at the area surrounding the point $P_4$ at the west coast of Niigata (Fig. 15) during the two months from April to May of 1962. It is seen from these figures that radioactive grains first travelled SSE as a whole and then turned E. Their maximum travelling distance was about 100 m from the depositing point.
Fig. 15
Map of Niigata Coast

- : area investigated
Upper left: Activities from fall-out accumulated at the estuary
Contour: equi-count lines (cpm) measured in April 1962
• : deposition points in Oct. 1961 with Co$^{60}$
□ : sampling points of mud
Contour maps of this kind were drawn and the movement of sand was presumed for each depositing point. The results obtained concerning the direction of sand movement are shown in Figs. 17 and 18 both for the river mouth and the west coast. The arrows in the figures are the main directions of sand movement.

On the west coast of Niigata oceanographical and meteorological observations have been made by the government of the Niigata Prefecture during the past years, including the period of our observation [8].

Sand drift may take place mainly under the action of wave currents at the time of high waves and it must be very remarkable during a storm. From an analysis of the data of the longshore current on the west coast obtained during the three storms in the period of observation, it became clear that the direction of sand transport in the offshore area (Fig. 18) roughly coincided with that of the longshore current. This seems to certify the above mentioned relationship.

The results shown in Figs. 11 to 13 are obtained for sand movement from storms occurring during the warm season, while strong storms frequently pass through the area during the cold season. From studies by the
government of the Niigata Prefecture the following conclusions on the coastal current in the cold season in this area have been obtained [8]:

1) During the growing period of a storm, an eastward current prevails offshore.

2) In the fully grown period of a storm, the current has very small velocity and its direction is normal to the shoreline.

3) In the decaying period of a storm, a westward current prevails and this period is considerably longer than the others.

From these facts it may be presumed that sand is transported, as a whole, westward through the entire period of a storm. On the contrary, the result of our observation for the warm season shows an eastward movement of sand. With regard to the meteorological condition in this region, it is generally calm in the warm season except for stormy days caused by typhoons which pass through the area a few times a year. In the cold season, however, the monsoon prevails almost the entire season and a recession of the coast line takes place in this area. Thus it may be concluded that during a year the sand is transported westward on the west coast of Niigata.
3.4. An investigation of the vertical distribution of radioactive sand

Since 1954, in the course of the construction of the Tomakomai Harbour, routine radiotracer tests were made more than twenty times. The papers on the experiments carried out in the early stage have already been published. After beginning the proper work of construction in 1958, the technique was also used effectively to investigate the change of the pattern of sand movement according to the progressive extension of the breakwater.

Besides such tracings in the plane distribution, experiments on the vertical distribution of radioactive sand in the sea bed were carried out on this coast by M. Kato and his associates. Their intention was to investigate the correlation among the thickness of bed material suffering turbulence, wave height and water depth. Moreover, provided a correlation existed, the radiotracer method might enable a quantitative tracing of sand movement too. Some trials, not yet completed, are mentioned below.

The procedure consisted in measuring the relative activity in sand cores taken from the sea bed by boring. Fig. 19 shows the vertical distributions of radioactivity after six days from depositing radiotracer at a 3-m depth, the locations of boring points inside the equi-count lines, and the wave data during the period of observation. Though not so clear, it may be seen that
An experiment on the vertical distribution of radioactive glass sand in the sea bed at 3 m in depth

(a) Relative radioactivities in vertical direction, from which background is subtracted
(b) Equi-count lines (cpm) and sampling point
- deposition point
Date of deposition: 2 March 1957
- sampling point
Date of sampling: 7 March 1957
(c) Wave data during the experiment at 3 m in depth
Fig. 20

An experiment on vertical distribution of radioactive glass sand in the sea bed at 8 m in depth

(a) Relative radioactivities in vertical direction, from which background is subtracted
(b) Equi-count lines (cpm) and sampling point

*#: deposition point
Date of deposition: 6 July 1957

O: sampling point
Date of sampling: 21 July 1957
the activity, even by waves of 1 m in height, ranges to more than 30 cm from the bottom surface. Another similar experiment at a depth of 3 m suggested the above results also. Fig. 20 shows the results of the same kind of experiment at a different depth. It may be seen from these data that the thickness of bed material disturbed by waves of as much as 2 m in height is at most 20 cm at a depth of 8 m. The results obtained at 7 m and 9 m seem to be similar to that at a 8-m depth.

The phenomena in common in these experiments were the following:

1. The vertical distributions were not always continuous; and
2. The total activity of the sand core was not always proportional to the amount shown at the corresponding point in the equi-count figure of the plane distribution.

It is thought, however, that by using much more radiotracer the phenomenon may be made clearer.
4. A LABORATORY INVESTIGATION ON SAND MOVEMENT IN A WAVE CHANNEL

4.1. Purpose

Although research on equilibrium profiles of a nearshore area and the initiation of sand movement and the development of sand ripple resulting from wave action have already been performed by many researchers, the phenomena of this research have not closely agreed with the movement of individual sand particles. This creates difficulty in tracing the motion of sand particles by the preceding methods. S. Sato, one of the authors, is now making a study of sand movement resulting from waves by using radioactive sand in a wave channel. A brief explanation of the experiment is presented in this section.

4.2. Method

As the nuclide of tracer, Cr⁵¹ was used in this experiment. Its half-life is 27.8 d which is not sufficiently short for laboratory use, but its emitting gamma radiation is only 0.3 MeV and hence safe handling is not difficult. As the radioactivity of the glass sand used at an experimental

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**Fig. 21**

Schematic illustration of apparatus in laboratory investigation

(Dimensions in cm)
run did not exceed a few mc, no special device was necessary in this case. The personal exposure dose was not detected by pocket chambers and film badges.

Radioactive glass sand mixed with natural sand is put on the model sand beach and bottom in sufficient thickness, and its movement due to waves is traced by the water-tight scintillation probe fixed on the carriage travelling over the channel, as shown in Fig. 21. After each test run is finished, contaminated sand is removed into a drum can by a siphon and stocked until the radioactivity decays to a safe level.

4.3. An example of the test

The dimensions of the wave channel are 50 cm wide, 80 cm deep and 40 m long. One end of this channel is separated into two channels; one is 15 cm wide and the other about 25 cm wide. In the part 15 cm wide, a wooden plate slope, the inclination being one to ten, is provided for the experiment.

Before the experiment the sand of 1 to 0.5 mm in diameter is placed on the slope and the equilibrium profile is made by the wave which is 10 cm high, has a 4-s duration and continues for 8 h. Then the radioactive sand is placed on the points of wave breaking and the offshore (Nos. 1 and 2 in Fig. 22).

To describe in detail, 50 g of glass sand of 2 mc/kg is mixed with 200 g of natural sand, about 160 g of which is used to replace the natural sand 5 cm deep, 3 cm wide and 10 cm long at point No. 1 and about 30 g of which is used to replace the sand with a ripple 2 cm wide at point No. 2. After these preparations the wave 10 cm high and 1.5 s duration is generated, and the beach profile and distribution of glass sand are measured at a certain interval of time. The results are shown in Fig. 17. The bottom of the scintillation probe is kept 2 cm above the sand surface during the recording. The count numbers in Fig. 22 include the natural count which is about 170 cps on the offshore and about 240 cps near the water surface.

After the wave attack of 35 min, the beach is eroded in foreshore, especially in the place around point No. 1 where whole glass sand is moved to the offshore. The point of maximum count appeared at a distance of about 70 cm from No. 1.

After 135 min of wave action the trough which was observed around point No. 1 at 35 min was buried and the movement of glass sand into the trough is suggested by the observed distribution of count around the place. Also the lines of count spread down to the offshore nearly 400 cm and the change in profile shows the ascension of sand extending in this distance. At 240 min no remarkable change in profile is found except the elevation of the bottom, in the area from 300 to 400 cm, with an increase of count. After this stage the changes in profile and count are very small for increased wave action.

On the other hand, in the area around point No. 2 the bottom profile and the distribution of count do not show any remarkable change. Only a small decrease of maximum count and a small extension of count lines are found throughout an entire experimental run. The sand of a mound which exists at the distance of 500 cm from the shoreline in the initial stage is gradually scraped and transported shoreward.

From this experiment it is found that when the period of wave changes from 4 s to 1.5 s, the wave height remaining the same, the sand moves sea-
Fig. 22

Time change of profile and travel of tracer

Duration of wave

- Initial profile
- 35 min
- 150 min
- 300 min
ward in the inshore and shoreward in the offshore; these two movements border at the region of nearly 400 cm from the shoreline, being about 150 cm apart from the breaking point of the wave.
5. OBSTRUCTION OF THE OBSERVATION BY FALL-OUT

When the sand drift was investigated by S. Sakagishi at the estuary of the Shinano River, using Co\textsuperscript{60} glass sand, it was found that there had accumulated on the river bed near the estuary, as shown in Fig. 15, other radioactive substances besides Co\textsuperscript{60}. It was affirmed that the substances, which were mainly Zr\textsuperscript{95}-Nb\textsuperscript{95} and Ru\textsuperscript{103}-Rh\textsuperscript{103}, were from the nuclear detonations carried out in the fall of 1961.

For Zr\textsuperscript{95}-Nb\textsuperscript{95}, particularly, the particle size was less than 1 µm, and the substance was relatively stable for both physical and chemical changes, such as dissolution in water and reduction into smaller particles. Thus, it was concluded that part of the fall-out falling along the entire length of the river was carried in the river and further down to the river estuary, accumulating there. It was first thought that such a sea bed condition would not be present along the west coast of Niigata. But precautions were taken in the survey on the spot and in other samplings to prevent inefficient observation.

The results of observation showed that although there was not much obstruction from fall-out, there still existed fall-out such as Zr\textsuperscript{95}-Nb\textsuperscript{95} in the depression on the sea bed, which was not made of sand but of mud. Fig. 23 shows an example of the results of pulse-height analysis of the mud samples, containing not Sc\textsuperscript{46} but Zr\textsuperscript{95}-Nb\textsuperscript{95} having a peak at 0.76 MeV. It was in mud samples, not in sand samples, that large quantities of Zr\textsuperscript{95}-Nb\textsuperscript{95} were found. It may be safely said that fall-out substances gather in the places where fine-particle mud accumulates.

On the whole, the investigation of sand drift at the estuary was affected to a large extent by the unexpected fall-out. In the same observation on the coastline, however, no appreciable effect from fall-out was found.

6. CONCLUSIONS

We have now many coastal problems to be investigated, these are, storm-surge protection, beach erosion, harbour construction on a sandy coast and so on. The history of coastal investigation in this country is not long and even the routine observations of waves along the main coasts are still not sufficient. Concerning the investigation of sand drifts by means of radioisotopes, we have quite a few examples, some of which are described in this paper, but there still remain problems to be studied in an over-all coastal investigation as well as in the development of the method and apparatus.

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REFERENCES


DISCUSSION

R. HOURS: In connection with section 3.9 of the paper, describing the investigation of the vertical distribution of radioactive sand, I have a few questions regarding the procedures used for taking the cores of sand and measuring the vertical distribution of the radioactive grains. For example I should be interested in knowing how much time was needed for taking the cores; whether they could be taken in any weather or only when it was calm; whether the distribution of sand was disturbed during the process; what was the maximum depth at which a core could be taken with your equipment; and what was the cost of taking and studying a core?

M. KATO: Our method was as follows: after the radioactive glass sand had been allowed to disperse to some extent, a diver was sent down to the sea bed (during calm weather) to take cores at as many points as possible in the radioactive area (see Fig. 15 of this paper). About 30 min were required to get a sample at any one point. The sand core samplers were simple cylinders having internal diameters of 5 cm and 8 cm and were pushed into the bed by the diver, to a maximum depth of about 80 cm. Upon removal from the sampler, the sand core was cut into discs about 2 cm thick. Each disc was dried and then measured with a G-M or scintillation counter in a definite geometry. Our figures relate to experiments which were carried out 5-6 yr ago, when the Tomakomai coastal area was not contaminated by fall-out and the natural activity was quite low. Unfortunately, I do not know the cost per core.

R. HOURS: I asked about cost because I feel it must have a fairly important bearing on the situation; to get statistically significant results a certain number of cores will clearly have to be taken.

D. B. SMITH (Chairman): Does geographical or geological evidence indicate that the depth of sand movement in the area you investigated was likely to be as great as 50 cm? Is it possible that some of the radioactive particles found at considerable depths could have been carried there during the drilling or coring operation?
M. KATO: Where wave heights were up to 4 m the depth was in fact about 50 cm. As you suggest, there probably was some disturbance of the seabed by the drilling operations and this would have affected the distribution of the radioactive sand. To this extent, therefore, our results are somewhat approximate.

H. KAUTSKY: In connection with Mr. Kato's discovery of certain spots in estuaries with accumulations of fall-out products, we have made an observation in the Federal Republic of Germany that might confirm this. I am referring to the discovery in the Elbe estuary of a locality with an accumulation of fall-out products in a fine, brownish mud. The isotopes were identified as barium-140, zirconium-95, strontium-89, strontium-90 and caesium-137. This accumulation may have been a result of precipitation of suspended particles when the river water mixed with the salt water of the sea.

D. B. SMITH: We have also had confirmation of this effect in the United Kingdom.

It is stated in section 2.1 of the paper that the glasses were ground mechanically until the grain size distribution agreed with that of the natural sand on the site investigated. How did you match the size of the ground glass with the natural sand?

M. KATO: After mechanical grinding the glass was size-matched by sieving.

D. B. SMITH: An alternative technique is to match the glass and sand by sedimentation. We have used this method in many of our investigations, and feel that it can be expected to produce glass which has similar hydraulic properties to the natural sand.

G. H. LEAN: In view of the great variability of wave height, period and direction in nature, would you not get a better average value for transport direction by making a large number of small injections over a long period rather than making a few intensive ones over a short one?

M. HOMMA: We have not yet reached any definite conclusions on this subject because we are still in the experimental stage. More information is needed on the pattern of sand movement under the action of waves of different characteristics.

R. HOURS: I would like to comment briefly on the question whether a single, large-scale immersion of the tracer on the bottom is preferable to several small immersions, distributed in both time and space. In France, where we have hitherto made use mainly of ground glass, the glass is generally irradiated in standard aluminium containers having a capacity of 40–50 g. On the whole, we found it preferable to immerse the containers on the bottom separately, along a given profile for example, rather than to group them at a single immersion point. In this way we get more information on the movement of the sediment, for equivalent immersed activity.

Similarly, we have often spaced out immersions in time, to take account of the results obtained from earlier immersions, or to replace previously immersed tracers which have been dispersed by storms and can no longer be found.

In certain cases, however, it would obviously be worth while to combine all the available tracer in a single immersion.
THE PLACE OF ISOTOPE METHODS IN GROUNDWATER RESEARCH

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Abstract — Résumé — Аннотация — Resumen

THE PLACE OF ISOTOPE METHODS IN GROUNDWATER RESEARCH. The hydrologist's approach to groundwater utilization is described in terms of a few basic hydrological characteristics, namely the occurrence and origin of water, the physical and chemical properties of the aquifer and the fluid, such as the permeability, porosity, etc., the direction and velocity of water flow and its dispersion and mixing patterns.

The methods used for the determination of the basic hydrological characteristics are then surveyed. A new classification of isotope tracing methods, more suitable for comparison with the classical methods of hydrogeology, is suggested. The place of various isotopic tracing methods in this scheme is analysed. Their present and potential capacities and shortcomings are discussed. Their usefulness in providing "short-cut" solutions is stressed, and some examples are given. The isotope methods discussed are the use of stable isotope (hydrogen and oxygen) variations in water, the use of natural and bomb-produced tritium and C\(^{14}\), as well as the use of locally injected radioisotopes.

The summary emphasizes the destructive features of isotope methods: their variety and sensitivity of detection, the time scale inherent in radioactive decay, the existence of isotopes of the elements of water and their measurable separation in natural processes.

The contribution to the determination of the hydraulic parameters was found to be rather limited but of more direct value for the measurement of movement characteristics. The characteristics related to the bodily movement of water particles can be evaluated only with the aid of isotopes.
МЕСТО ИЗОТОПНЫХ МЕТОДОВ В ИССЛЕДОВАНИИ ГРУНТОВЫХ ВОД. Подход гидрологов к использованию грунтовых вод описывается в пределах некоторых основных гидрологических характеристик, а именно: источники и места залегания водоносного пласта, физические и химические свойства водоносного пласта и жидкой среды, например проницаемость, пористость и т.д., направление и скорость течения грунтовых вод, виды дисперсии и смешивания.

Рассматриваются методы, применяемые для определения основных гидрологических характеристик, предлагается новая классификация методов изотопных индикаторов, более удобная для сравнения с классическими методами. Анализируется место различных методов изотопных индикаторов в этой схеме и обсуждаются их современные и потенциальные возможности и недостатки. Подчеркивается полезность этих методов при составлении быстро распадающихся растворов, даются некоторые примеры.

Рассматриваемые изотопные методы заключаются в использовании вариаций стабильного изотопа (водорода и кислорода) в воде, в использовании естественных и образовавшихся в результате ядерных взрывов трития и углерода-14, а также в использовании локально введенных радиоизотопов.

В выводах подчеркиваются деструктивные черты изотопных методов: их разнообразие и чувствительность при обнаружении, период времени, свойственный для каждого процесса радиоактивного распада, существование изотопов элементов, входящих в состав воды, и разделение их в естественных процессах, которое может быть измерено.

Было установлено, что вклад в методику определения гидравлических параметров сравнительно ограничен, за исключением определения непосредственной величины при измерении характеристик движения. Оценка характеристик, связанных с движением водных частиц, может быть проведена только с помощью изотопов.

1. INTRODUCTION

The growing need and continuing emphasis on the development of groundwater resources has stimulated the advent of several useful techniques in groundwater investigations based on the application of isotopes. The first steps in this direction were rather random, determined partly by available techniques and prompted by acute hydrological problems. There is now a clear need to evaluate more critically the contribution of the isotope tech-
niques to groundwater investigations so as to be able to decide when and to what extent these methods should be used in any given hydrological problem.

During the last years several thorough attempts have been made in this direction [1, 2, 3], without however considering the isotope methods within the framework of the general methodology of groundwater research. In the present paper a classification of the isotope methods, that are suitable for comparison with accepted hydrological methodology, is presented. Their present advantages and limitations compared with conventional methods are indicated. In the final section it is shown by way of some concrete examples that in certain cases the isotope techniques can not only replace and improve an existing method but that they may provide more immediate answers to some hydrological problems. Since the art is in a rather early stage of development only preliminary ideas and a general approach can be given. A more quantitative and authoritative evaluation will have to be left to the future.

2. BASIC PROBLEMS OF GROUNDWATER HYDROLOGY, HYDROLOGICAL CHARACTERISTICS AND RESEARCH METHODS

2.1. Groundwater hydrologists are called upon to solve problems of ever-increasing complexity which usually arise in the context of some engineering scheme and which, therefore, are couched in practical terms. To evaluate the methods proposed for the solution of these problems it is first necessary to translate the problems themselves into the proper scientific terms and to analyse the components involved.

An aquifer and the water contained in it represent a physical and chemical system, the future behaviour of which under the action of certain influences must be predicted for the solution of hydrological problems. From a strictly scientific point of view a rather small number of basic parameters and boundary conditions should suffice to define the system completely and to deduce its future behaviour. In actual practice it is more convenient to use varied information, for which in the following the broad term "characteristics" is used. Knowing these characteristics and the basic physico-chemical laws involved, one should be able to arrive at a complete solution of hydrological behaviour of a groundwater system. In actual practice difficulties are encountered which result from the fact that these "characteristics" can be known only partially owing to practical limitations, and that some future influences (e.g. precipitation) cannot fully be known even in principle. On the other hand, the required accuracy usually falls short of the ideal scientific goal. Predictions within comparatively large margins of error are acceptable in actual practice, provided they can be achieved with a reasonable amount of work and expense.

2.2. It was found convenient to describe the groundwater system in terms of the hydrological characteristics (shown in Table I) which can be grouped into the occurrence and origin of water, the physical and chemical properties of the medium and of the fluid and characteristics of its movement. The terms which are listed are not covered by exact scientific definition but are those commonly used in groundwater hydrology.
2.3. The known methods and techniques employed by the hydrogeologists for groundwater research and development are grouped in a way convenient for later evaluation (see second footnote to Table I). The isotope methods are also included either separately or as part of other methods. They are described in more detail in section 3.

3. THE ISOTOPE METHODS

3.1. Isotopes used

A fairly complete description and evaluation of presently available isotope methods have been given by several authors [1, 2, 3, 4]. In all of these descriptions the methods are classified according to the isotopes used, rather than according to the principles of their applications. Generally the following isotopes have been considered:

(a) Stable isotope tracers of water, i.e. deuterium and O\(^{18}\);
(b) Tritiated water resulting from natural (cosmic ray), thermonuclear or artificial operations;
(c) Other radioisotopes produced by cosmic rays in the atmosphere and introduced into the water through rain scavenging or through the decay of organic matter;
(d) Naturally occurring radioisotopes of the uranium or thorium series; and
(e) Artificially injected radioisotopes (other than tritium) in a chemical form compatible with the geological environment.

In order to evaluate the relative merits of the isotope and conventional hydrological methods a different classification, i.e. into dating, pulse tracing and isotope analysis, is suggested. More direct comparison with the other methods within the framework of Table I is then possible.

3.2. Dating methods

These are specific radioisotope methods which rely on the unique property of radioactive decay. Every radioisotope defines a time scale according to its decay rate. Dating of any system involves a comparison to this scale by measuring the change of the specific radioactivity between an input point to that at the point of analysis. The input can be the rain recharge areas or the point of transition from an adjacent groundwater system. In order to apply the method unambiguously all processes which change the radioisotope concentration other than decay (such as exchange, dilution or removal) must be taken into account. Thus far, the most useful isotope for groundwater dating has been the cosmic-ray-produced tritium, which labels rain with a tritium content characteristic of the locality. The use of tritium as a pure dating tool has, however, been considerably curtailed lately by the thermonuclear production of tritium.

C\(^{14}\), members of the natural radioactive series and other cosmic-ray-produced isotopes, such as Si\(^{32}\), have been proposed as dating tools for groundwater, for time scales of more than tens of years. However, prob-
<table>
<thead>
<tr>
<th>Characteristics</th>
<th>Methods (**)</th>
<th>Evaluation of isotope methods' contribution (***)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Occurrence and origin of groundwater</td>
<td>(A) Existence and extent of the groundwater body 5, (1, 2, 3, 4), 6, 14 [3, 10 or 9 (ii)], 16</td>
<td>S</td>
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<td></td>
<td>(B) Type of water (of meteoric origin, connate, juvenile, etc.) 10, 8(i), 8(ii)</td>
<td>D</td>
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<td></td>
<td>(C) Connection to and relationship with water sources and adjacent aquifers 5, (1, 2, 3, 4), 6, 14, 8(ii), 15</td>
<td>A</td>
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<tr>
<td>Physical and chemical properties of the medium</td>
<td>(D) Internal structure (shape, size and distribution of pores) 7</td>
<td></td>
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<td></td>
<td>(E) Porosity 7, 9[A, I]</td>
<td>A</td>
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<tr>
<td></td>
<td>(F) Effective porosity (coincides in a phreatic aquifer with storage) 7, 11, 12, 13 [A, G, Q, R], 9(i) [A, I]</td>
<td>A</td>
</tr>
<tr>
<td></td>
<td>(G) Permeability 7, 11, 12, 13 [A, F, Q, R]</td>
<td>S</td>
</tr>
<tr>
<td>Characteristics</td>
<td>Methods (*) (**)</td>
<td>Evaluation of isotope methods' contribution (*) (**)</td>
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<td>(H) Elasticity (related to storativity in confined aquifer)</td>
<td>11, 12</td>
<td>-</td>
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<tr>
<td>(I) Dispersivity</td>
<td>7, 9(i)</td>
<td>D, P</td>
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<td>(J) Solubility (leachability)</td>
<td>7, 15</td>
<td>-</td>
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<tr>
<td>(K) Ion-exchange capacity</td>
<td>7, 9(i)</td>
<td>A</td>
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<tr>
<td>(L) Adsorption capacity</td>
<td>7, 9(i)</td>
<td>A</td>
</tr>
<tr>
<td>Physical and chemical properties of the fluid</td>
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<tr>
<td>(M) Temperature</td>
<td>6</td>
<td>-</td>
</tr>
<tr>
<td>(N) Chemical composition</td>
<td>8(i), 4, 6</td>
<td>-</td>
</tr>
<tr>
<td>(O) Density</td>
<td>8(i)</td>
<td>-</td>
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<tr>
<td>Movement of groundwater</td>
<td></td>
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<tr>
<td>(P) Direction of flow</td>
<td>13 [R], 9, 10, 1, 2, 3</td>
<td>A</td>
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<tr>
<td>(Q) Velocity of flow</td>
<td>13 [F, G, R], 9, 10, 15-?</td>
<td>D</td>
</tr>
<tr>
<td>(R) Hydraulic head distribution</td>
<td>5, 13 [A, G, O]</td>
<td>D, P</td>
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<tr>
<td>(S) Mixing and dispersion patterns</td>
<td>9, (13, I), (8, C)</td>
<td></td>
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</table>

Definitions in Table I above

(*) 5 - Method 5 can be applied directly.
(1, 2, 3, 4) - A combination of the methods 1, 2, 3 and 4 furnishes the desired solution.
9 [A, I] - The determination of method 9 is conditional upon the knowledge of characteristics A and I.
TABLE I (cont.)

Concise list of research methods

Surface surveys, also known as preliminary surveys:
1. Topographical and morphological
2. Geological
3. Hydrological survey of replenishment areas and of water basin outlets
4. Surface geophysical exploration (electrical resistivity, seismic, gravimetric, etc.)

Subsurface surveys:
5. Well drilling and testing (including rock sampling, water sampling, water-level measuring and discharge measurements.)
6. Formation logging (drilling rates, electrical, nuclear, caliper, temperature, etc.)

Laboratory analysis:
7. Rock sample analysis
8. Water analysis (i) chemical identifications and (ii) trace analysis (including isotopic tracers)

Dynamic methods, associated with the so-called field methods:
9. Pulse-tracing, divided into (i) controlled injections and (ii) uncontrolled (and natural) injections
   (This method is mainly an isotopic one.)
10. Dating, an exclusive isotope method
11. Pumping tests
12. Analysis of water-level fluctuation (The method refers to fluctuations which are induced by tides, earthquakes, surface reservoir operation and meteorological cycles.)

Computation methods
13. Flow equations
14. Continuity equations
15. Geochemical balances
16. Analysis of spring flow

Underlined method numbers indicate that isotope method is included.

Underlined method numbers indicate that isotope method is included.

- D - Isotope method is the most direct way of determination.
- P - Isotope method is the principal available method.
- A - Isotope method is an alternative choice to conventional methods.
- S - Isotope method is of secondary importance.
lems connected with their removal by processes other than decay have not been satisfactorily resolved to date [2].

Dating methods can contribute to the determination of the extent (size) of groundwater reservoirs (A), can be used as a test of the validity of assumptions concerning non-meteoric origin of groundwater systems (B) and for velocity or transit time determinations (Q). In Table I they are designated as method No. 10.

3.3. Pulse-tracing methods

Pulse tracing is here defined as the process of relating the changing concentration of an isotope at the place and time of measurement to that at a known injection point and time. Tracing is, of course, not peculiar to radioisotopes, and materials such as dyes and salts belong to this category; actually corrections have to be applied for the radioactive decay of the isotope tracers before results can be interpreted. The significance of isotopes as tracers is in their extension of the tracer method owing to their better detectability and the variety of novel natural injection mechanisms with which they are associated. Tritiated water, being an isotopic molecule of water, is an ideal tracer insofar as interference of environment is concerned.

A distinction is made between local controlled injections and natural or artificial uncontrolled injections.

3.3.1. Controlled injections

Injection into the water bodies can be made locally using radioisotopes in a chemical form and with half-lives, and at concentrations suitable to the specific problem to be investigated. The tracers are chosen to move as freely as possible with the water into which they have been injected. Because of health-hazard regulations these activities are generally limited to relatively small areas and to short periods of time. Common tracers used, aside from tritiated water, have been Co$^{60}$ or Co$^{58}$ in the form of $K_3Co(CN)_6$, Cr$^{51}$ as the ethylene diaminetetraacetic acid complex and Br$^{81}$, I$^{131}$ and S$^{35}$ in anionic form.

Pulse tracing has been used for the purpose of determining a number of aquifer characteristics such as groundwater velocity (Q) porosity (E and F) and the dispersivity of the medium (I).

3.3.2. Natural and non-controlled artificial injections

Under this category come naturally occurring isotopes which are introduced in a non-steady manner. Examples are the seasonal changes of O$^{18}$ and deuterium concentration in rain-water resulting from the temperature and climatic changes between summer and winter, as well as the pulsed injections of thermonuclear tritium. The injection pattern as a function of time and space is well known from the results of the rain analysis for these components. Applications include the determination of groundwater velocities (Q) and mixing patterns of rain-recharged groundwater (S).

These methods are designated as method 9(i) and 9(ii) respectively.
3.4. Isotope analysis (component tracing)

This method is based on relating the concentration of some identifiable component of water body to the characteristic concentration of this component in some source. In the usual form of its application the relative contribution of the source to a mixed water body is determined. Conversely, because of different composition, the connection between bodies can be excluded. Any identifiable water component such as dissolved salts, trace elements, stable isotopes of oxygen and hydrogen as well as radioactive isotopes can be utilized. Naturally occurring or artificially injected components can be used for this purpose. The use of the method is based on quasi-steady-state conditions and it can be regarded as an extension of the tracer technique to non-transient conditions. In some respects, it can hence be considered an "isotope dilution method" [5]. In hydrology this method is involved in the determination of the origin of some water bodies and of their relationship to adjacent systems (B, C) and of the mixing patterns (S). The method can, of course, be used only in those cases where measurable differences of trace element concentration, of natural isotopic abundances, or of age, exist between the different waters.

This method is part of method 8(ii).

4. EVALUATION OF THE CONTRIBUTION OF ISOTOPE METHODS TO HYDROLOGY

4.1. Nuclear techniques have found their place in hydrological research on three levels:

(a) Nuclear analytical tools such as activation analysis of trace components in water or rocks and neutron or γ-logging methods of boreholes have been useful aids to existing classical research methods Nos. 6, 7, and 8. These applications have recently been reviewed [1] and are not considered further in this paper as not properly belonging to this discussion.

(b) Isotopic materials have added a new method — dating — to the research methods available to the hydrologists, and have added new dimensions to the tracing techniques 8 and 9. These contributions are discussed more fully in 4.3 in the framework of the research scheme described in 4.2.

(c) The three isotope techniques have in many cases enabled a direct solution of engineering problems without delving into a complete hydrodynamic description of the system.

4.2. Table I lists the groundwater characteristics and the research methods used for their characterization, isotope methods being underlined. The numbers within round parentheses show the methods that jointly contribute to the solution, while those in square brackets following the method indicate that the solution is dependent on additional information of the kind described within the brackets.

The last column summarizes the isotope methods' contribution to the hydrological characterization, as evaluated in section 4.3. The letter "D"
indicates that the isotope methods are the most direct way of obtaining the required information. The letter "P" is added whenever these are the principal methods available. "A" indicates the case where isotope methods are important alternatives to conventional ones, while "S" indicates that they are of secondary importance only.

4.3. The following evaluation will use the terminology of Table I; the numbers refer to methods enumerated in the second footnote to Table I.

4.3.1. The existence and extent of groundwater bodies (A)

There is no substitute for finding water in a borehole (5), or for prediction of its occurrence based on detailed geophysical prospecting (1, 2, 3, 4). These methods are, however, costly and slow, so that indirect methods are advantageous at a preliminary survey stage. Such a possibility is provided by isotope methods. Suppose that the possible extent and depth of an aquifer system is known from geo-hydrological reasoning, and that either the inlet or outflow amounts are known. It is then possible to compute the actual water volume in the system (and hence its extent) by means of water balance equations using the transit time determined by dating at the outflow point. Analysis of the transit time of pulsed injections of thermonuclear tritium can be used for the same purpose; however, more than one year's observation is generally needed for this. The same is true for the transit time determination by discharge analysis of natural outflow (16) which gives very similar information to that obtained by pulse tracing of naturally injected tritium. The advantage of the method, when applicable, is that no bodily approach to the area is needed; its limitation is, that unless the mixing pattern in the aquifer is known the transit time as determined by dating will be ambiguous up to a factor of two [3, 6] and that in principle only minimum reservoir sizes are found by this method (slowly moving water pockets are not detected).

4.3.2. Origin and type of water (B)

In the determination of the origin of some water body, i.e. whether it is of meteoric origin, connate or juvenile, the isotope contribution is through dating and isotope analysis. While dating can be decisive in determining positively the meteoric character of water by tritium analysis, the differentiation by age between the other types is beyond the range of the presently available dating methods. There is then a good case for developing dating methods of suitable range for geologic ages of water.

The isotope analysis contribution is based here on the stable isotope composition of water. Any concentration ratio characteristic of sea rather than meteoric waters, of differing significantly from both, would be a strong indicator of the origin. Isotope analysis may be the only means of indication in cases where sea water intrusion or leaching have strongly influenced the original salt content of the water body. They constitute therefore not only an alternative method, but in the above cases a unique method of origin determination.
4.3.3. Connection to and relationship with water sources and adjacent aquifers (C)

While again most answers to this characteristic will have to come from detailed (and costly) geo-hydrological surveys (1, 2, 3, 4, 5, 6), the isotope water analysis for either stable or radioactive components may be used to define boundaries between adjacent aquifers. A boundary will be indicated by a discontinuity in tritium or stable isotope content, while the transport of water through such a boundary might be indicated by common components in two adjacent water bodies. The isotope methods here provide useful contributory information for the definition of the system. They are especially valuable for the establishment of direct connection between a water body and its sources of replenishment in systems with rapid turnover. Experiments of this kind have been conducted successfully with tritium in karstic limestone.

4.3.4. The physical characteristics of the medium (D, E, F, G, H)

The pulse-tracing technique essentially measures the velocity of flow, being the only direct measurement along the flow path. Hence this method is suitable for determination of those properties within the porous medium which govern the flow rate, i.e. permeability, porosity, etc. The isotope methods (tracing) are here important additional research tools, depending however in every case on additional hydrological information.

Among the methods known for measurement of the effective porosity of an aquifer, only tracing and pumping tests yield results under field conditions. The classical pumping test technique for that purpose is of value only in phreatic aquifers, while under confined conditions it would give the storage value which is some function of the porosity and the elasticity of the system. A tracing experiment, although dependent on other parameters, provides adequate values for the effective porosity under both phreatic and confined conditions. A typical injection experiment for this purpose consists of the injection of a tagged volume of water into a well and subsequent observation of the tracers' concentration in an adjacent pumping well. Using equations of water balance, the product of porosity and aquifer thickness is determined [7]. Alternatively the permeability (G) is determined using the Thies equation [8].

The porosity measured by the isotope tracer method is an intermediate value between porosity and effective porosity, wherever these differ considerably. The actual results depend on how fast diffusion of the tracer into stagnant pores dilutes the tracer concentration in the main water stream.

4.3.5. Dispersivity (I)

The direct measurement of the pore size distribution cannot at present be correlated with dispersivity. Tracer applications are then the only possible methods available for following the dispersion of a moving body of water through a porous medium, and for measurement of the dispersivity as represented by the hydrodynamic dispersion coefficient. This can be
carried out satisfactorily by tracers of type 9(i). On the other hand, the dispersion pattern obtained from naturally injected isotopes 9(ii) generally reflects inhomogeneities of the tracer concentration at the input areas, and interpretation in terms of the dispersion coefficient is difficult. While laboratory experiments on soil samples can be performed and interpreted easily, the extrapolation of these results to field conditions is often questionable, so that in situ tracer experiments have to be carried out [10]. At present only radial geometry field experiments with full penetration of the injection well in the aquifer can be treated in terms of a dispersion coefficient [9]. In principle, however, there is no reason why the mathematical tools for other cases should not be developed. Non-isotope tracers can be used in these experiments as well, and isotopes will be chosen whenever their sensitivity and ease of handling warrant it.

4.3.6. The chemical properties of the medium (J,K,L)

These are best measured in the laboratory using either classical or isotope techniques. An isotope of the element investigated is usually preferred. The single-well technique [10] with such an isotope provides the best means for an ion exchange and adsorptivity determination in situ. This method seems to be the most reliable one to use whenever problems of contamination of water sources from disposal of sewage effluent or of radioactive wastes are envisaged.

Isotope methods discussed here do not contribute directly to the measurement of physical or chemical properties of the fluid (M,N,O). On the other hand, in the determination of the movement of water through porous media the tracing and dating methods are unique, since they measure the characteristics of the flow, such as velocity, direction and the mixing directly. Conventionally, available methods for these characteristics are based on indirect calculations and require a knowledge of parameters of the medium as well as detailed mapping of the head distribution (R).

4.3.7. Direction (P)

The direction of flow in an aquifer is usually inferred from the water-level contours. This is a costly and time consuming procedure and depends on assumptions concerning the isotropy of the aquifer. It is therefore suitable for thoroughly explored aquifers but will not be used at an early exploratory stage.

In principle this characteristic should be amenable to tracer study. Tracing methods are the most direct methods for the determination of flow direction at a given point. They are the only valid ones to use in anisotropic fields where the flow is not perpendicular to equi-potential lines.

Unfortunately, however, several attempts made so far to develop directionally sensitive devices using the tracing method have not resulted in a routinely available isotopic tool. A central injection well surrounded by observation wells at a distance of several meters is the only one yielding results undistorted by local effects. It is however a rather costly device. The single-well technique, based either on multiple counters or directional
adsorption [2] may be strongly influenced by local distortions of the natural gradients around the well. This field offers scope for new approaches and solutions.

The directional adsorption method which is based on $\gamma$-emitting isotopes is specifically an isotopic one. The other methods can also be used with non-isotope tracers. The general flow direction in some aquifers, especially in confined aquifers, can be also determined by the dating method. In this case the characteristic to be determined would be the relative distance of the wells from the recharge areas.

4.3.8. Velocity (Q)

Distinction should be made between velocity determinations which give a very localized characteristic, and transit-time determination from a recharge area. While isotope methods have been firmly established in the determination of surface flow velocities, the groundwater velocity measurements are still in the evaluation stage. The recent results of MOSER and MAIERHOFER [2] indicate that the single-well dilution method is a promising one for the measurement of local velocity, provided that distortion and vertical currents around a borehole are accounted for. This technique is based on tracing methods 9(i) and can be applied for isotopes and other tracers.

The usefulness of naturally injected tracers (9(ii)) to determine velocities has also been demonstrated in several cases. So far only isotope tracers have been used, if only for reasons of lack of other tracer elements suitable for this investigation. These methods measure the mean transit times rather than the velocity at any given point.

The transit time can also be estimated with the aid of the dating method. Tritium is presently the only isotope capable of producing fairly precise data under simple geological conditions, i.e. whenever mixing of water bodies of different ages is negligible or whenever the aquifer boundaries and the dispersivity is known [3, 6]. In other cases the transit time found is ambiguous due to the unknown mixing pattern of flow lines.

Groundwater velocities can also be estimated under favourable conditions from geochemical considerations (15). The reasoning would be based on the concentration in water of some slowly leachable trace elements, whose leaching rate is known or can be computed. The method will be of value mainly for radioactive trace elements which decay at a rate comparable to the transport rate. The isotopes to be utilized would be the natural radioactive elements of the uranium and thorium series. The method is then in a way a dating method, based on an isotope introduced into the water in situ [3].

4.3.9. Mixing and dispersion patterns (S)

Only a method which measures the bodily movement of water can be used for determining this characteristic. Hence pulse tracing for determination of the dispersion and trace element dilution analysis for determining the degree of mixing of different waters, have no competing methods in this
case. The isotopic water molecules, both stable and tritiated, play a specially important role, because they are in principle the best tracers for observing the true movement of the water bodies with which they are associated. In the case of other tracers in solution, care must be taken to eliminate errors resulting from the effects of exchange and adsorption on the aquifer medium.

The determination of the mixing patterns with the aid of tritium has been discussed in detail in a working paper of GAT and NIR [2]. Differences in tritium content of various water streams which result from the difference in age are the distinctive feature used for identification of water from these streams in a mixed water body. The large concentration change induced by the injections of thermonuclear tritium is here used to advantage; mixing can be determined to much higher accuracies nowadays than was possible during the cosmic-ray tritium period. An important result of such mixing studies has been the age stratification which was found in phreatic aquifers [11].

There still remains scope for much additional research to find distinctive tracers to be used for mixing studies. Trace elements and isotope composition peculiarities of dissolved tracers (such as carbonates, sulphates, etc.) may here be used advantageously under certain circumstances.

4.4. The distinctive features associated especially with the natural isotope tracers enable a significant contribution of the isotope methods in the early exploratory stage of groundwater utilization [4] (characteristics A, B, and C of Table 1). As a result of their use, engineering decisions can sometimes be made without resort to a complete hydrodynamical description of the system. These "short cut" solutions may be the only available ones, wherever the hydrological system is complicated, so that a full description of the system cannot be aimed at. Two possible situations, typical of arid zones, are described to illustrate this point.

Example 1: Exploitation of groundwater in very arid regions

Experience shows that in these regions two classes of groundwater resources may be distinguished:

(a) Large reservoirs which are replenished by slow migration of groundwater from very distant catchment areas or were replenished, slowly, during long periods, and
(b) "Pockets" of groundwater of limited size, adjacent to seasonal rivers and flood paths and replenished by them.

A reliable supply can be obtained, of course, only from large reservoirs, whereas exploitation of the smaller pockets is important because water of better quality is usually found here.

Dating by tritium gives in this case an indication which of the two situations is encountered. It is then the only research method by which results may be obtained without very prolonged and costly field investigations.
Example 2: Full exploitation of groundwater resources in an arid plain

An arid plain composed of alluvial fill is surrounded by hills. Storm runoff and snow melt from the slopes of these hills infiltrate into the alluvium and feed a groundwater horizon which is exploited by wells. Evapotranspiration from saline swamps forms the only natural outlet.

For a full exploitation of the groundwater resources it is necessary to find out whether only this alluvial aquifer exists or whether some hypothetical, deeper-lying aquifer conveys groundwater from a replenishment area in higher and more distant mountains to the salt swamps.

The problem can be investigated with the aid of tritium dating and O$_{18}$ analyses of water samples from the swamps, the alluvial aquifer, flood waters and precipitations in the hills and mountains. Oxygen would immediately indicate the origin of the water, i.e. whether it comes from hills or high mountains (altitude effect). Tritium is an indicator of infiltration of recent waters from runoff. Here the advantages of isotope techniques stem from their ease of application in a region of difficult access.

All such solutions will, however, only be temporary ones. As an aquifer system becomes more developed and known, research and planning will tend to proceed along more systematic lines. The isotope contribution will then be within the scheme described in 4.3.

4.5. Summarizing, it is found that almost all isotope applications in hydrology are tracer studies in some sense, i.e. the behaviour of a material added to or being a minor constituent of normal water is being studied.

There are some distinctive features of isotopes which determine advantages and limitations of their use as tracers. Among them are:

(a) The variety of isotopes of different chemical and radioactive properties and the sensitive detection methods available for their identification. This enables appropriate choice of a tracer for almost any situation. Health hazards are here the main limitations.

(b) The inherent time factor of decay associated with the use of radioactive isotopes. As a result isotopic materials have a memory so that one momentary measurement can replace extended periods of hydrological observations. On the other hand the half-life of the tracer must be chosen in accordance with the time scale of the system under investigation.

(c) The fact that tritium, deuterium and oxygen-18 are isotopes of the water elements and hence ideal tracers for the unperturbed identification of water bodies.

(d) The fact that the stable isotopic abundance is changed measurably by the basic physico-chemical processes of the natural water cycle, such as condensation and evaporation. This results in a unique characterization of water from different sources.

As a result of these and other features one can evaluate the distinctive role played by isotope methods within the framework of conventional methods. This role reveals that:

(a) The isotope methods' contribution to the determination of hydraulic parameters is limited;
However, all characteristics related to the movement of water are most directly measured by tracing methods in general and isotope methods in particular;

Those characteristics that are related to the actual travel path of individual water particles, can only be evaluated with the aid of isotopes of water elements; and

The flexibility and relative ease of obtaining isotope data provide short-cut methods, in comparison with the common hydrogeological procedures, for the solution of certain general hydrological problems.

REFERENCES


DISCUSSION

R. AMBROGGI: Can you give us some data on the actual application of these methods in Israel?

Y. HARPZ: Isotope techniques are being used in a variety of ways. We are carrying out a fairly comprehensive tritium survey which covers all the natural water resources of the country. Deuterium and oxygen-18 measurements are being made to study the natural and artificial recharging of groundwater, as well as to check the water balance of a hydrological basin. Finally, intensive use is being made of pulse-tracing methods in an attempt to determine the mixing and dispersion patterns in groundwater that are brought about by underground storage and waste-disposal operations.

R.W. NELSON: Have you had any experience with in situ measurements of hydrodynamic dispersivity parameters?

Y. HARPZ: We have been carrying out tests in single-well and two-well systems to determine the coefficient of dispersion. The technique
used is a new one and is based on the analysis of radial-flow dispersion and the use of a number of simplified assumptions. A report on this work will be appearing shortly.

Y. MIYAKE: You mentioned the possibility of using O$^{18}$ for groundwater investigations. I wonder whether difficulties do not arise as a result of the isotopic exchange of this isotope between water molecules and bicarbonate ions.

A. NIR (on behalf of Y. HARPAZ et al.): Our experience is that the isotopic differentiation found in rain-water is fairly exactly reflected in the corresponding groundwaters. We have not found that any exchange takes place with carbonates. Perhaps Professor Tongiorgi could comment on this?

E. TONGIORGI: Practically no exchange occurs at low temperatures. This is shown by the concordant O$^{18}$-O$^{16}$ and D-H data obtained in groundwater analyses, which reflect the mean isotope composition and, in some cases, the seasonal variation in isotope composition of rain-water also.

C.V. THEIS: In section 3.2 of your paper you mention the possibility of using dating methods for testing the validity of assumptions concerning the non-meteoric origin of groundwater systems. Have you actually found any important groundwater systems that are not meteoric?

Y. HARPAZ: To the best of my knowledge, no groundwater reservoir or system of non-meteoric origin has yet been found. The statement is made in the paper because the origin of water is so often the subject of controversy and we feel that dating and isotope analysis methods can be of decisive importance in definitely establishing whether a given system of water is of meteoric origin or not.

Y. INOUE: In the Table on groundwater system characteristics and methods for their determination you indicate under item (E) that pulse-tracing techniques can be used for porosity measurements. How is this done?

Y. HARPAZ: Briefly what happens is that a pulse is injected into one well and measured at another so as to provide information on the rate of flow. As the thickness of the aquifer is known — it has to be known, as we point out in the Table — it is possible to deduce the effective porosity. Details of the technique can be found in reference [7].
HYDROLOGIC PHENOMENA AFFECTING THE USE OF TRACERS IN TIMING GROUNDWATER FLOW

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Abstract — Résumé — Аннотация — Resumen

HYDROLOGIC PHENOMENA AFFECTING THE USE OF TRACERS IN TIMING GROUNDWATER FLOW. The phenomena of field-scale dispersion, through poorly understood at present, are among the most important affecting the use of tracers in groundwater hydrology, and in particular in timing movement by tracing peak concentrations.

The dispersion in uniform media observed in the laboratory is of such magnitude that, when it is extrapolated to lengths of travel found in the field, it is for most considerations negligible. However, dispersion in the field is of a much greater magnitude, amounting in a test at Hanford to a lateral spread of at least 30' and longitudinally to several times the distance of travel, about 3.5 km. This dispersion is several orders of magnitude greater than that observed in the laboratory.

The reason for the increased longitudinal dispersion must lie in the wide distribution of permeabilities in any suite of sedimentary beds, which appears to be roughly exponential in character. This in itself would give a large dispersion. Part of these effects are probably statistical in nature, but probably some are caused by lenses of such extent that they cannot be treated statistically.

Dispersion of this type is believed to explain best the previously published data of von Buttlar on bomb-produced tritium in groundwater at Carrizozo, New Mexico. By using the times of peak concentrations, von Buttlar computed times of travel for groundwater in an alluvial fan. The concentrations consistently rise during summer rains and fall during winter rainless periods, and all wells are at the sides of arroyos which furnish most of the recharge of the area. The data probably represent peak concentrations during flow in the arroyos followed by a decline in concentration as the tracer spreads by largely lateral dispersion.

On one hand, tracers give a direct approach to the problems of movement of contaminants in groundwater and probably the most useful approach to problems of dispersion in the field. On the other hand, they cannot be used a short cut in the absence of other hydrologic data.

PHÉNOMÈNES HYDROLOGIQUES INFLUENÇANT L’EMPLOI DES RADIOINDICATEURS POUR LA DÉTERMINATION DE LA VITESSE DE DÉPLACEMENT DES EAUX SOUTERRAINES. Les phénomènes de diffusion dans les sols en place, bien qu’encore mal connus, sont entre ceux qui revêt la plus grande importance pour l’emploi des radioindicateurs en hydrogéologie, notamment lorsqu’il s’agit de déterminer la vitesse de déplacement des eaux en suivant le déplacement des concentrations maximums des indicateurs.

La diffusion dans des milieux uniformes, observée en laboratoire, est d’un ordre de grandeur qui, appliqué aux parcours constatés dans les sols en place, peut être considéré comme négligeable dans la plupart des cas. Or, dans les sols en place, la diffusion est beaucoup plus importante; au cours d’un essai fait à Hanford, elle s’est étendue latéralement sous un angle d’au moins 30° et longitudinalement sur 3,5 km environ, soit plusieurs fois la longueur du parcours des eaux. Cette diffusion dépasse donc de plusieurs ordres de grandeur celle qui est observée en laboratoire.

L’ampleur de la diffusion longitudinale s’explique certainement par le fait que dans toute succession de couches sédimentaires, la perméabilité prend des valeurs très différentes qui se répartissent grossièrement selon une courbe exponentielle. Ce phénomène assure à lui seul une diffusion étendue. Certains de ces effets doivent être de nature statistique, mais d’autres sont probablement dus à des loupes de dimensions telles que la méthode statistique ne leur est pas applicable.

On estime qu’une diffusion de ce type fournit la meilleure explication des données publiées par von Buttlar sur la présence, dans les eaux souterraines de Carrizozo (Nouveau-Mexique), de tritium provenant d’explosions d’armes nucléaires. En se fondant sur les valeurs du temps correspondant au passage des concentrations maximums, von Buttlar a calculé la vitesse du mouvement des eaux souterraines dans un cône de déjection. Les concentrations augmentent régulièrement durant l’été à la faveur des pluies, et diminuent

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предиким канальем и с одной стороны, меченые атомы дают прямой подход к проблемам изучения движения загрязняющих веществ в грунтовых водах и по всей вероятности дают наиболее подходящее решение проблемы дисперсии в полевых условиях, а с другой — их нельзя использовать без других гидрологических данных.

МЕДИЦИНОН ДЕЛБИ ФЛЮДОГ АГУАС СУБТЕРРАНЕАС МЕДИАНТЕ ИНДИКАРОРС: ИНФЛУЕНСА ОФ ЛОС ФЕНОМЬЮН ХИДРОЛОГИЧЕСКИХ. Проблеме, вьг unprotected="""" в науке и в особенности когда смещения получены определяя максимальные концентрации, то вероятность в области применения индикаторов в отдельных точках.

ГИДРОЛОГИЧЕСКИЕ ЯВЛЕНИЯ, ВЛИЯЮЩИЕ НА ИСПОЛЬЗОВАНИЕ МЕЧЕНОХ АТОМОВ ПРИ ХРОНОМЕТРИРОВАНИИ СТОКА ГРУНТОВЫХ ВОД. Явления дисперсии в полевых масштабах в настоящее время изучены недоста- точно и являются наиболее важными среди процессов, влияющих на использование меченых атомов в изучении грунтовых вод, в частности при определении времени прохождения потоков посредством введения индикаторов в отдельных точках.

Дисперсия в однородной среде, наблюдаемая в лаборатории, представляет собой величину, кото- рая, будучи эстимована на большие расстояния в полевых условиях, в большинстве случаев оказывается незначительной. В полевых условиях дисперсия имеет гораздо большую величину; при испытаниях в Хэнфорде распространение индикатора доходило в горизонтальном направлении почти до 30°, а в продольном направлении в несколько раз превышало продольный путь — около 3,5 км. Эта дисперсия на несколько порядков больше дисперсии, наблюдаемой в лаборатории.

Основная причина увеличения продольной дисперсии должна лежать в широком диапазоне прови- ци-цемостей серии осадочных лож, которые по всей вероятности носят экспоненциальный характер. Это само по себе должно давать большую дисперсию. Часть этих эффектов является, вероятно, статисти- ческой по своей природе, а некоторый, вероятно, вызываются чечевицообразными залежами, и их не- возможно обработать статистически.

Можно полагать, что дисперсия этого вида наиболее приближает опытные данные фон Бутлера о тритии, попавшем в грунтовые воды в Карризозо, Нью-Мексико, после взрыва бомбы. Используя время прохождения пиковых концентраций, фон Буттлер вычислил время прохождения индикатора в грунтовых водах в аллювиальном веерообразном конусе выноса. Концентрации экскономерно увеличи- ваются во время летних дождей и уменьшаются в зимнее время, когда нет осадков; все колодцы на- ходятся в местах расположения арройо, составляющих большую часть питания грунтового горизонта района. Данные по всей вероятности отражают пиковые концентрации во время прохождения потока в арройо и сопровождаются уменьшением концентраций, поскольку меченые атомы распространяются на большое расстояние в горизонтальной плоскости.

С одной стороны, меченые атомы дают прямой подход к проблемам изучения перемещения загрязняющих веществ в грунтовых водах и по всей вероятности дают наиболее подходящее решение проблемы дисперсии в полевых условиях, а с другой — их нельзя использовать без других гидрологических данных.

MEDICION DEL FLUJO DE AGUAS SUBTERRANEAS MEDIANTE INDICADORES; INFLUENCIA DE LOS FENÓMENOS HIDROLÓGICOS. Probablemente, el más importante de los fenómenos que ejercen influencia sobre los resultados obtenidos cuando se emplean indicadores en el estudio hidrologico de aguas subterráneas, y en particular cuando se miden sus desplazamientos determinando las concentraciones máximas del indicador, es la dispersión que se produce en la naturaleza y que todavía no se conoce satisfactoriamente.

La dispersión que se observa en el laboratorio en un medio uniforme es tal que cuando se extrae a las longitudes de recorrido que se presentan en la naturaleza, se podría despreciar en casi todos los casos. Sin embargo, es mucho mayor sobre el terreno; por ejemplo, en un ensayo realizado en Hanford, alcanzó una amplitud lateral de por lo menos 30° y, longitudinalmente, varias veces la distancia recorrida, a saber, unos 3,5 km. Esta dispersión es varios órdenes de magnitud mayor que la observada en el laboratorio.

La magnitud de la dispersión longitudinal se atribuye a la extensa gamma de permeabilidades que se encuentra en cualquier secuencia de estratos sedimentarios, que, tiene, al parecer, carácter aproximadamente exponencial. Este hecho daría ya en sí una dispersión muy grande. Probablemente, una parte de estos efectos tiene carácter estadístico, pero es casi seguro que algunos se deben a estratificaciones lenticulares de tal magnitud que no es posible tratarlas estadísticamente.

Se estima que una dispersión de esta clase es la que mejor explica los datos publicados por von Burtlar sobre la presencia de tritio-formado a raíz del ensayo de armas termonucleares— en las aguas subterráneas,
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en Carrizozo, Nuevo México. Ese investigador calculó las velocidades de desplazamiento de las aguas subterráneas en un cono aluvial de deyección a partir de las velocidades de los máximos de concentración. Estas concentraciones aumentan siempre durante las lluvias veraniegas para disminuir en los períodos secos de invierno, y todos los pozos se encuentran cerca de arroyos que suministran la mayor parte de la recarga de la zona. Los datos representan probablemente máximos de concentración durante el flujo en los arroyos, seguidos de una disminución de la concentración conforme el indicador se reparte por dispersión principalmente lateral.

Los indicadores brindan el medio más directo para abordar los problemas del desplazamiento de las materias que contaminan las aguas subterráneas y, probablemente, constituyen el mejor modo de resolver los problemas de dispersión en la naturaleza, pero sólo pueden utilizarse cuando se dispone de otros datos hidrológicos.

INTRODUCTORY STATEMENT

The uses of radioisotopes in the study of hydrology are many. The present conference will doubtless develop new instances and techniques for their use in new subfields of the science. The present paper is intended to call attention to some hydrologic principles affecting the interpretation of data from tracer experiments concerned with groundwater flow. It will reinterpret some published work on tracers for the purpose of pointing out modifying hydrological principles.

FIELD DISPERSION

One of the processes, the knowledge of which is only now being developed, affecting the interpretation of tracer data is that of dispersion on field scale. Just as hydrodynamic dispersion in a homogeneous porous medium observed in the laboratory is much larger than that resulting from molecular or ionic diffusion [1, 2], so that observed in the field is much greater than that observed in the laboratory.

The extent of field dispersion is probably best shown by the observations made at Hanford [3]. In this experiment 100 lb of urine in a concentrated solution was slowly introduced into an unused well over a period of about 24 h. The diagram (Fig. 1) shows the concentration of the dye found in two unused wells about 4 km distant. The lines joining the input well to the two observation wells include an angle of almost 30°. One observation well is almost directly down the regional hydraulic gradient from the pumped well. The peak concentration in the second observation well was about 1/4 of that in the well down-gradient. In both wells measurable concentrations of the dye first appeared about 60 d after injection. The peak concentration occurred 130 to 140 d after injection, and measurable concentrations were still present after 227 d.

This dispersion is on a scale some orders of magnitude greater than that shown in the laboratory. The formation in which the dispersion occurred is unusual. It consists of about 40 ft of extremely permeable, gravelly glacial outwash with permeabilities as high as 2400 m/d. Because of its unusual character, the dispersion observed in it may not be typical of other formations.

The explanation for the dispersion in a longitudinal direction seems to lie in the distribution of permeabilities in most formations carrying water. Most suites of samples taken from formations or parts of formations show an exponential distribution of permeabilities; that is, if the strata are
Dispersion of a slug of fluorescein in coarse sediments at Hanford (from HW-60601)

A. After 227 days concentration was 2.0 parts per $10^9$.
B. After 227 days concentration was 2.2 parts per $10^9$.

arranged in order of increasing permeability, a plot of the logarithms of permeability against accumulated thickness of beds gives points lying approximately on a straight line. Fig. 2 indicates typical plots of several formations. The plots represent a spread of permeabilities through 1 or 2 orders of magnitude plus a small percentage of very low permeabilities that lie below the line joining other points in a suite.

The significance of this distribution of permeabilities seems to be that in itself it would give a wide distribution of a tracer placed in a well. As a first approximation, a tracer put into a well, whether or not by pumping, is distributed among the strata in proportion to their respective permeabilities. The rates of travel are proportional to the permeabilities again as an approximation, and the rate of input into an observation well from the various strata is also in proportion to their respective permeabilities. If the beds were continuous from input to observation wells and there were no other dispersive forces operating, there would still be a wide dispersion because of these differences in permeability. Some characteristics of the observed field dispersion would be reproduced; a rapid rise in concentration would be observed, followed by a much slower fall in concentration.

However, the individual beds in most formations are not generally extensive, and no doubt sedimentary lenses of varying permeabilities act on a larger scale very much as do inequalities of pore size in a uniform medium in the laboratory to produce a statistical type of variation in velocity and therefore dispersion. It is likely, however, that some series of beds of different permeabilities extend through all or most of the distances commonly
involved in tracer tests. Channel fillings extending 50 km or more are known. Such extensive groups of beds may explain some of the steps and other irregularities shown in the Hanford and other tracer studies.
TRANSVERSE DISPERSION

Most sedimentary formations forming aquifers are lenticular in nature, and the lenses have commonly unequal dimensions in the horizontal, as well as vertical, directions. This lenticularity causes erratic movements of water, in travel speed and in direction, that are superimposed upon the general direction of travel predicted by the hydraulic gradient, as determined from water levels in wells spaced at intervals relatively large in comparison with the scale of the lensing.

When water moves from a less permeable material to a more permeable one, it is refracted according to the relation

\[ \tan \alpha_1 / \tan \alpha_2 = p_1 / p_2, \]

in which \( \alpha_1 \) and \( \alpha_2 \) are the angles of incidence and refraction, as measured from the normal to the interface of the materials, and \( p_1 \) and \( p_2 \) are the respective permeabilities.

The width of a band of tagged water is narrowed in consequence of this refraction according to the relation

\[ W_2 / W_1 = \cos \alpha_2 / \cos \alpha_1 = \left[ \frac{1 + \tan^2 \alpha_1}{1 + (p_2 / p_1)^2 \tan^2 \alpha_1} \right]^{1/2}, \]

in which \( W_1 \) and \( W_2 \) are the respective widths.

Lateral dispersion is quite sensitive to the width of band in which a tracer is contained [4]. Tracer put into a well is always introduced into an aquifer in a band of finite width, which will vary as it moves with the permeability of the lenses it encounters and with the angle with which it meets them. This is shown clearly by an experiment reported by HUBBERT [5]. The importance of this intergranular dispersion in the field is that it renders largely irreversible the large-scale spreading of a tracer by lenticular features which might be expected to be largely reconcentrated by similar features oppositely oriented at some other point in its course.

It is probable that movement occurs not only transversely to the general direction of flow of the water but also vertically, causing an irregular and complex spiralling movement of the water down the general hydraulic gradient. In a lenticular aquifer the trend of coarse lenses is probably in different directions in different horizons of the aquifer. The excess head at the downstream end of one coarse lens at one horizon may coincide with the deficient head at the upstream end of a differently oriented coarse bed at another horizon, giving rise to cross-formational flow, some vertical mixing of a tracer, and a further dispersal of an original band of tracer introduced. Such common features as cross-bedding will also tend to give a vertical component to the water velocity and to produce cross-formational movement.

Lenses of relatively low permeability will divert much of the flow of water around them, which will also lead to dispersion in favourable locations by bringing tracer-bearing water in contact with tracer-free or tracer-poor water.

It would appear that the non-homogeneities in the formation on a small scale relative to the distance involved in a field experiment might give a
normal or nearly normal distribution of any tracer and lead to a diffusion or dispersal-type equation with enormously larger constants analogous to diffusivity. However, it would also appear that in most field experiments there may be elements on a scale large enough to give unpredictable movements to the water and of a tracer unless there is rather detailed knowledge of the stratigraphy.

This discussion has been directed primarily toward granular aquifers but applies in principle to limestones, or other aquifers with solution-type openings, and to porous basalts. They are not discussed, because the erratic type of movement in such aquifers has been long recognized.

In passing it may be well to point out that the instance of very slow dispersion comparable with ordinary molecular diffusion cited by ERIKSSON [6] is over distances too small (approximately 3 m) to involve the processes discussed here and is concerned with water moving at an average velocity small enough (approximately 2 m/yr) to be in the range where simple diffusion dominates other dispersive mechanisms in flow through porous media [4].

INTERPRETATION OF TRITIUM DATA AT CARRIZOZO, NEW MEXICO

As an instance that seems to involve dispersion on a large scale, data reported by von BUTTLAR [7] are reviewed here.

In 1956 bomb-produced tritium was used as a tracer for groundwater movement at Carrizozo, New Mexico. Water from a group of five wells and one spring was sampled at intervals of about two months through a period from October 1955 to February 1958 and analysed for tritium content.

The wells are on an alluvial fan on the northwest flank of Sierra Blanca on the eastern edge of the Tularosa Basin, which is a large basin without surface drainage. The sampling locations range in altitude from about 1600 to 2100 m. Sierra Blanca is a mass of intrusive and extrusive igneous rocks reaching an altitude of 3300 m. Surface drainage in the mountain area is eastward; no streams from the mountains go through the area considered. Several low hills of igneous rocks and of tilted Cretaceous shales and sandstones project through the unconsolidated sandy fan material, which may be in other places more than 30 m thick.

The groundwater flows more or less radially down the alluvial fan, as contours on the water table are roughly parallel to the smoothed topographic contours. The fan is scored radially by a great number of arroyos ordinarily dry but flowing after heavy rains. There is no doubt that such flows contribute significant amounts of recharge to the fan and they may contribute the major part. Some water is contributed from Sierra Blanca, but the fact that the surface drainage divide is only about a mile from the northwest edge of the mountain suggests that it is not a large proportion.

All the sampled wells are on the edges of creeks or arroyos and are pumped by windmills, with the exception of well 4, which is the well for municipal supply of Carrizozo, and well 6, which is a shallow pit dug probably as a prospect hole. Source 7 (despite the original paper) is a spring in Cretaceous rocks outcropping under alluvium in an arroyo. Because no logs are available, the aquifers for most of the wells are in doubt. Well 4 is perforated both in the alluvium and in Cretaceous sandstone; well 8 is
deep and probably draws water from Cretaceous rocks but probably also from the alluvium inasmuch as, according to the owner, it is affected greatly by flows in the arroyo; and well 2 is probably in alluvium. Well 1 near the mountain front is probably in slope wash but perhaps in weathered igneous rock. There is little uniformity in the wells used as sampling points, and it is probably to be expected that there should be little uniformity in the tritium content of water found in them.

Fig. 3 C shows the results of tritium analyses as taken from von Buttlar's papers. Fig. 3 B shows the rainfall by 6-d periods at Carrizozo in inches of depth, and Fig. 3 A shows the tritium content of rainfall at Chicago, Illinois, and Socorro, New Mexico (dotted line, left scale), and the product of Carrizozo rainfall and the measured tritium in rainfall, labelled tritium-inches, which represents the dosage of tritium applied to the ground at Carrizozo by 15-d intervals (solid line, right scale). One tritium-inch is about 740 μc of tritium per km².

Von Buttlar in his first paper, dealing with the measurements in 1956, proposed two hypotheses to account for the tritium concentrations in the sampled waters: (1) that the 1956 peaks represent the 1954 peak concentration
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of atmospheric tritium and that the lag in time between the peaks in the other wells, as referred to well 1, indicates the rate of travel of the groundwater which is in excess of 50 ft/d (about 15 m/d); (2) that the peaks represent the tritium after the tests of 1956 and the lags represent the times of travel of the tritiated precipitation through the zone of aeration to the zone of saturation. In his final paper von Buttlar somewhat tentatively concluded that the first hypothesis was probably correct inasmuch as the peaks observed in 1957 were smaller than those in 1956, which he judged to correlate with the low atmospheric tritium of 1955 better than with the intermediate tritium of 1957 itself. He suggested that the data were subject to re-interpretation.

The pore velocity of 50 ft/d suggested is not excessive. The water body in the alluvium rests upon a sloping platform, and the water table slopes nearly 200 ft/mile, or at a slope of about 0.04. The permeability suggested by the rapid movement calculated is rather high but in the absence of better information could be assumed.

However, if we assume with von Buttlar that the tritium peaks represent the passage of a single surge of tritium, it is necessary to assume that the 1954 tritiated rain took 2 yr to pass from its catchment in Sierra Blanca to the alluvial fan and then completed its journey through the fan in about 3 months and that its appearance in the fan happened to coincide with the rainy season and with another thermonuclear explosion and consequent peak in atmospheric tritium. It is also necessary to find an explanation for more peaks in the rainy season of 1957, which, under the assumptions, coincide with the low atmospheric tritium of 1955.

Probably the main reason to question this interpretation lies in the irregularity of the tritium peaks. Well 1 does show the first peak (in July), as pointed out by the author; but all the other wells peak at the same time (in October), with the exception of well 4, the most distant from the mountain front, which peaks 2 months earlier. This thus would indicate that the water moved up the hydraulic gradient and up the slope of the alluvial fan. In 1957 well 1 peaks later than most of the other wells. Well 8 has two peaks each year.

It seems that the inconsistencies in these tritium records should be disregarded because they apparently must be ascribed to individual differences in rainfall in their vicinity, to differences in stratigraphy and perhaps in some cases to contamination by rainfall in open tanks from which some of the samples were obtained. The general characteristics of the wells as a group seem to be the following:

(1) They all rise to their peaks in summer in the rainy season;
(2) They fall to their lowest points in the winter and early spring when precipitation is low and falls as snow on frozen ground; and
(3) The heights of the summer peaks in the 2 yr of record in all but sampling point 8 correspond approximately to the tritium availability.

In 1956 heavy rains at Carrizozo coincided with the rainfall tritium peak (at Chicago) in July and August. All sampling points reached their maxima during this summer and fall for the 2-yr period, except for the consistently anomalous windmill well 8. In 1957 the tritium peak corresponded with a rainless period at Carrizozo, and atmospheric tritium was apparently relatively low when the rains came. The peaks of tritium content are much lower with the exception of those of well 8.
It would seem that a proper interpretation of these tritium data should try first to explain the over-all pattern. The peaks of the wells in October of 1956 can be correlated with the high rainfall of July and August during the high peak in atmospheric tritium and similarly the low peaks in 1957 with the much lower atmospheric tritium of that year. This suggests local sources of recharge, that is, the arroyos.

The general rises in early 1956, before extensive rains and during a time of low atmospheric tritium, but during a time when recharge probably occurred in the form of melted snow, and the ground thawed, seem to indicate storage in the zone of aeration of tritium resulting from the 1954 explosion.

The very rapid decrease in tritium concentration after each peak during each summer dry period and particularly during each winter, characterized by little or no available recharge, needs explanation. The spring and wells 1 and 8, which fluctuate most widely in tritium content, are close to arroyos, which in general flow in the same direction as the groundwater moves. Hence on the basis of the general flow there would be no reason to expect that the concentration of tritium contributed along the length of an arroyo when it is in flood should rapidly diminish. The high-tritium water contributed near a well should be replaced by similar water moving from points somewhat farther up the arroyo.

The rapid decrease seems best explained by the lateral dispersion of the tritium-carrying water under the arroyo. In the alluvial deposits, enough elements of heterogeneity almost certainly occur to cause field-scale dispersion, and in the Cretaceous rocks the tilted bedding is at an angle to the general movement of groundwater. Hence the addition of tritium under the arroyo bed at a time of storm runoff first appears at the edge of the arroyo in high concentration, but in the course of two or three months the tritiated water now passing the sampling point, having entered farther up the arroyo, has had opportunity to disperse and consequently to be diluted. It is also probable that movement away from the groundwater mound built up under an arroyo during storm runoff, in a direction transverse to the general direction of movement of the water, causes an unknown amount of longitudinal dispersion.

The general fluctuations of the tritium content of water in these wells seem therefore to indicate: (1) a rapid dispersion lateral to the general flow once the tritium enters the ground water, as shown by the rapid decrease in concentration near the arroyo when recharge stops; (2) a probable dependence in most of the wells on the availability of the current atmospheric tritium, as shown by the general correspondence of the tritium peaks with the levels of atmospheric tritium during local rainfall; (3) a storage of some tritium in the vadose zone, as shown by the general rises in the tritium levels between January and March 1956, when it may be presumed that recharge was being added during the thawing of the ground but when atmospheric tritium was at a low level.

The erratic changes in tritium content in individual wells relative to each other indicate local sources of recharge to the ground water. As to individual wells, it may be pointed out that well 4, which is not on an arroyo and which pumps the greatest quantity of water, has the lowest tritium peaks (8 to 12 tritium units), has a tendency to peak later, and reaches minima
of pre-bomb tritium levels. In fact, the minima would indicate an age of at least one tritium half-life and, taken in conjunction with the variation indicating some admixture of bomb tritium, seem to indicate a mixture of some water carrying bomb-produced tritium with a larger volume of tritium-free, and therefore quite old, water. It, together with well 8, probably taps water in both the Cretaceous rocks and in the alluvium; well 8 certainly and well 4 probably have a double peak. The higher peaks in well 8 probably result from its position at the edge of an arroyo. It is so situated that it may receive some surface contamination, and contamination is also suggested by its high nitrate content. Source 7, a spring in an arroyo, has the highest peak, probably because it is the most centrally located with respect to the recharge. Well 1 is highest in elevation, is near a permanent mountain brook and is in the foothills of the mountain. The rainfall pattern may be different for this well, and the course of water to it is almost certainly different from that to the others. Its changes relative to the others are therefore an index of its environment rather than a marker of a pulse of tritium passing through an aquifer far from recharge.

If this explanation is correct and the data are correct, we have welcome evidence of the effects of dispersion and some hints about the method of movement of water through the vadose zone.

**MIXING OF TRACER WATER VERTICALLY**

In the preceding discussion the effects of dispersion have been emphasized, both along the direction of movement and transverse to it. The apparent conclusion is that there would be doubt about stratification of hydraulic flow on field scale.

Some of the best evidence for stratification of flow in aquifers is provided by data from the Wharton Tract study in New Jersey [8]. The Wharton Tract is an area lying about 40 km northwest of Atlantic City and about 15 km from Atlantic Ocean waters. Analyses of sampling of water collected in 1958 from various depths at each of 8 well sites in an aquifer about 30 m thick indicated by its tritium content that the age of the water varied with depth from a few months near the water table to pre-bomb old water at the bottom of the water body. Analyses of samples of water in a closely neighbouring stream during a period of low flow, when it was carrying essentially only effluent groundwater, showed that only the topmost layers of the groundwater contributed to its flow. The authors point out that the presence of discontinuous clay lenses in the sand of the aquifer causes the vertical permeability of the sediments to be much smaller than the horizontal permeability.

This anisotropy of sediments is the usual condition in horizontally-bedded sediments, differing only in degree. On a large scale the actual field condition could be represented by an isotropic model with the vertical dimension enlarged to a scale inversely proportional to the square root of the ratio of the permeabilities. In such a model the shallow flow tubes would have a great advantage over the deeper ones in short distances. In such a model the 30-m thickness of the aquifer in the Wharton Tract might very well be equal to the distance from the farthest wells to the river (about 150 m) or even larger.
Most water tables slope at an angle to the trend of a stream, so that there is a downstream, as well as a toward-stream, component of movement of the groundwater. Obviously the water in the deeper stream tubes will travel farthest. In a stream system in humid areas groundwater emerges at various levels, from sea level or some other control in the main stream, to successively higher levels in successively higher, perennially flowing tributaries. The highest outlets in tributary streams discharge the youngest water, which is the topmost water. The tributaries act as skimmers of the groundwater body of an area and by furnishing relatively high-level and relatively closely-spaced outlets accelerate the movement in the upper part of the aquifer. Relatively deep pumping wells accelerate the movement of the deeper water, and in many areas their effects must be considered.

It would appear that stratification of tracers would be favoured by relatively low vertical permeability with respect to the horizontal and by proximity to streams or other outlets, both of which conditions were present in the Wharton Tract; but these conditions may not occur to the same degree in groundwater that has travelled farther or in material with less vertical variation.

GENERAL DISCUSSION

Tracers are a useful addition to the tools of groundwater hydrology; and radioactive tracers, because of their use in minute quantity and therefore without change in the physical character of the liquid, are particularly useful. Tracers in the form of particular ions in groundwater have been used for decades for the quantitative estimation of the sources of groundwater in drains, the amount of brine leaking into a river, the volume of a groundwater reservoir and similar problems. The first quantitative field studies of the movement of groundwater in the United States were made by means of tracers.

However, the use of tracers in quantitative problems in groundwater movement is not as simple as it seems and is an adjunct to, and not a substitute for, other types of groundwater investigations. In the study of waste disposal, either radioactive or other industrial waste, it promises to be the most useful of all methods, for we are interested here in the actual path and time of travel of a contaminant. Even here the geology and the general hydrology of the area must be known to permit the design of an intelligent experiment with tracers. In the study of groundwater production, tracers have a disadvantage compared with pumping-test methods notwithstanding the difficulty of the application of equations based on homogeneity to aquifers which are in varying degree heterogeneous. These at least integrate the effects of heterogeneity over an area and yield direct data on the quantities of chief interest, which are the possible rate of production of water and the effects on the other wells in the vicinity and produce estimates of future trends in these quantities, whereas tracers yield information on travel time along lines. It remains to be seen how elaborate a tracer test is needed to produce a good estimate of these quantities.

Every effort should be made to use tracers in connection with intensive groundwater investigations by governmental agencies or by industries and
their groundwater consultants. The use of tracers in this way will serve a two-fold purpose: it will aid the interpretation of pumping tests, and, conversely, the techniques of using tracers can be evaluated and improved.

Probably the best places for the evaluation of tracers in combination with other hydrologic methods are large nuclear establishments in which very large amounts of tracer-level water has entered the ground. Here the incentives for intensive study are present, and there is the opportunity for a study of the movement of tagged water over a large area. Such intensive studies are being made at the larger nuclear institutions of the United States.

When those unfamiliar with hydrologic techniques use tracers, it would seem obvious that they should seek all the advice they can get from practicing hydrologists as to the general hydrology and as to methods of sampling and the nature of errors inherent to sampling. Although seemingly obvious, this has not always been done in the past.

Finally, it appears that two fields of investigation are particularly suitable for the use of tracers and that studies in them are necessary for the intelligent application of tracers to groundwater studies. The first is their use in the determination of the amount and character of field dispersion in various types of sediments and other rocks and, in many areas, in the hope that some quantitative generalizations can be developed. The second is the nature of movement through the zone of aeration to the water table: how rapidly a particular batch of water moves through this zone under different antecedent conditions, under conditions of simple and of complex stratigraphy, and how much mixing occurs with water already in the ground.

REFERENCES


DISCUSSION

J. F. HONSTEAD: I wonder whether a large-scale divergence of flow lines rather than actual dispersion might not possibly have contributed to the relatively widespread distribution of tracer dye found in the Hanford experiment. I am thinking of the possibility of flow diverging from the input
well owing to the existence of divergent gradients which might not have been recognized because of lack of data.

C.V. THEIS: A cursory examination of the data might certainly suggest an interpretation along those lines. However, the site on which the investigation was carried out was some distance to the south of the centre of a water-table mound, which was fairly low at the time of the test. Any divergence of flow lines from the input well which might have been caused by this mound could hardly have amounted to more than a few degrees. Moreover, in the case of divergent flow lines in a homogeneous material one would expect the concentration of the tracer to have been the same in both wells. The pulsing in the wells points to the interposition of some agency other than ordinary Darcian flow. Finally, it has been established by observation that the glacial outwash in which the test was carried out is extremely heterogeneous. Theoretically, therefore, it must be assumed that an erratic course would be followed by any parcel of water passing through this zone in response to a hydraulic gradient. In other words, the tracer would have an opportunity of dispersing widely.

Y. HARPAZ: Do you think there is any substitute for tracer methods among conventional hydrological techniques when it comes to dealing with problems involving the bodily movement of water underground, e.g. underground storage, underground mixing, waste disposal, etc?

C.V. THEIS: The movement of individual parcels of water underground through most porous formations is so complex and even unpredictable that I am convinced that tracers provide the only means of establishing exactly how such wastes do move. The use of tracers seems to me to be essential for mapping the movement of water in connection with any operation, such as the disposal of waste in the ground, which is liable to give rise to contamination.
APPLICATION OF RADIOACTIVE TRACERS IN SCIENTIFIC GROUNDWATER HYDROLOGY*

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Abstract — Résumé — Аннотация — Resumen

APPLICATION OF RADIOACTIVE TRACERS IN SCIENTIFIC GROUNDWATER HYDROLOGY. Radioactive tracers may serve as a valuable tool in the analysis of groundwater hydrology. An area of major contribution is in the determination of the more fundamental properties of porous flow systems, such as: porosity distributions, stream functions, and components of the permeability tensor. However, before the full capability of radioisotopes can be utilized in studying these fundamental properties, and understanding and description of hydrodynamic dispersion in porous media is needed.

The importance of dispersion and flow-system geometry in the analysis of tracer results is illustrated through the solution of a particular flow system. Equations describing three-dimensional flow through the soil from a pond to an adjacent river are solved. A linear approximate analysis of dispersion is superimposed to provide a method for interpreting tracer results. Some examples of possible misinterpretations by not separating the several independent causations are illustrated. Suggestions are given to assure representative sampling techniques for radioactive tracers.

UTILISATION DES RADIOINDICATEURS EN HYDROGÉOLOGIE. Les radioindicateurs peuvent rendre de précieux services dans les études d’hydrogéologie. Ils sont particulièrement utiles lorsqu’il s’agit de déterminer les propriétés fondamentales de terrains poreux (répartition de la porosité, fonctions de courants, composantes du tenseur de perméabilité, etc.). Cependant, avant de pouvoir tirer pleinement parti des radioisotopes pour l’étude de ces propriétés fondamentales, il est indispensable de comprendre le mécanisme de diffusion hydrodynamique dans les milieux poreux.

Pour illustrer l’importance que la géométrie du système de diffusion et d’écoulement revêt dans l’étude critique des résultats obtenus grâce à un radioindicateur, les auteurs donnent la solution des équations caractérisant l’écoulement à trois dimensions des eaux d’un étang, à travers le sol, vers un cours d’eau voisin. Ils donnent en outre une analyse, par approximation linéaire, de la diffusion, comme méthode d’interprétation des données fournies par le radioindicateur. Ils citent quelques exemples d’interprétations erronées que l’on risque de faire en omettant d’isoler les différentes causes indépendantes en jeu. Ils suggèrent des méthodes pour obtenir des échantillons représentatifs, ce qui est indispensable lorsqu’on emploie des radioindicateurs.

ИСПОЛЬЗОВАНИЕ РАДИОАКТИВНЫХ МЕЧЕНЫХ АТОМОВ В НАУЧНОЙ ГИДРОЛОГИИ ГРУНТОВЫХ ВОД. — Радиоактивные меченые атомы могут служить ценным средством при изучении гидрологии грунтовых вод. Область главного их применения является определение основных характеристик течений в пористых грунтах, как, например: распределение пористости, функции потоков и компоненты тензора проницаемости. Однако до того как все возможности радиоизотопов в изучении этих основных характеристик будут открыты, необходимо иметь представление о гидродинамической дисперсии в пористой среде.

Значение дисперсии и геометрии системы потоков при анализе результатов применения меченных атомов иллюстрируется анализом отдельной системы подземных течений. Рассматриваются уравнения, описывающие трехмерный поток через грунт из бассейна в прилегающую реку. Анализ дисперсии сделан в линейном приближении. Тем самым мы достигаем упрощение интерпретации полученных результатов. Описываются несколько примеров возможных ошибок интерпретации, возникающих вследствие недостаточного учета разных независимых факторов. Даем советы, обеспечивающие правильный отбор проб при использовании радиоактивных индикаторов.

EMPLEO DE INDICADORES RADIACTIVOS EN EL ESTUDIO CIENTÍFICO DE LAS AGUAS SUBTERRÁNEAS. — Los indicadores radiactivos pueden constituir un medio muy valioso para analizar la hidrología de las aguas

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The application of radioactive tracers in making conventional studies of groundwater flow is receiving increased attention; however, not to be overlooked is the possible utility of radioactive materials in making comprehensive, refined analyses of groundwater flow systems. Although the immediate need for such detailed studies appears to be restricted to special situations (e.g. radioactive waste disposal, biochemical degradation of wastes) where a nearly exact knowledge of travel time is desired, it may well be that the future will reveal many other useful applications for such information.

Radioactive tracers do not offer a panacea to groundwater flow studies. Much the same limitations are imposed on the use of radioactive materials as on chemical tracers. Such limitations arise from the vast amount of prior knowledge about the flow system which is required to plan tracer tests and later to accurately interpret test results. Basic problems relative to the use of tracers, which were effectively pointed out by SKIBITZKE [1], must be carefully considered.

The purpose of this paper is to indicate the several independent factors which, in combination, make up the delay time for tracer arrival in an observation well. Also, a method for combining the several independent causations is illustrated through application to a specific flow problem. Areas are suggested where radioactive tracers hold future promise in evaluating the fundamental properties of porous flow systems and in investigating those aspects of flow in porous media which warrant more accurate definition.

The actual selection of isotopes or compounds for tracers based upon detection limits, radioactive half-life, soil adsorption and other chemical or physical characteristics is purposely omitted since it is discussed elsewhere [2]. Discussion is also limited to the field use of radioactive tracers, thereby omitting the whole area of beneficial laboratory applications.

General consideration of tracer tests

Tracer tests generally involve introducing a water-tracer solution, which is easily identifiable and of known concentration, into the flow system of interest. The distribution of the tracer, that is its location with respect to point of injection and time, is determined through periodic or continuous sampling of observation wells. The distribution of the arrival time of the
tracer then is the single dependent variable measured. Accordingly, the lapsed time between injection of the tracer and its detection in an observation well is the single variable from which one may define useful flow parameters.

The lapsed time between injection and detection is the combined result of four independent causations:

1. The macroscopic flow pattern or geometry of the flow paths in the system;
2. The hydrodynamic dispersion which occurs during flow through the soil;
3. The removal and movement retardation of the tracer resulting from chemical and physical properties of the soil environment; and
4. The dilution and distortion of tracer concentration by the observation well and test inflow structure.

The first two items are fundamental to any porous media flow system and must be analysed. One usually attempts, through careful selection of tracers and if possible through careful observation-well design, to reduce or eliminate the difficulties presented by items 3 and 4.

Of the four effects, the largest is that of flow-pattern geometry and the associated variation of velocities along each travel path. If the tracer moves in a straight line of distance \( \sigma \) and at a constant velocity \( v \) from injection to the observation point, then the time analysis will involve only dividing \( \sigma \) by \( v \) to determine the flow-geometry component of time. Yet in actual flow systems the flow path is seldom a straight line nor is the velocity uniform. Therefore, the shape of the flow path must be determined and the velocity integrated along that irregular path to yield the component of tracer time.

The second item requiring consideration in tracer analysis is hydrodynamic dispersion which designates the microscopic aspects of flow in porous media. Hydrodynamic dispersion will be used here to designate the following observed phenomenon. When two miscible fluids, originally separated by a sharp interface, move through an inert porous medium, the mechanisms of molecular diffusion, velocity variations in pores of irregular length, and interphase mass transfer tend to alter the shape of the interface. Such dispersion has been shown capable of at least moderately precise description in terms of soil dispersivity parameters.

The detrimental effects - physical, chemical or biological - of soil environment should be eliminated through careful selection of the tracer. Effects such as ion exchange, complexing with subsequent removal by soil filtering action, precipitation, and removal by bacteria are complex and usually very difficult to evaluate and to integrate into the flow calculations.

The distortion of tracer concentration by the observation-well disturbance can only be minimized and then the remaining effect logically analysed. The strong interaction between flow-system geometry and distortion dictates the need for a thorough analysis of well effects.

A specific flow system for tracer analysis

Steady flow from a pond into a gently sloping permeable bed is approximated by a combined three-dimensional source and sink in a uniform flowing stream. The contour of the pond is allowed to assume the shape necessary
to meet the equipotential boundary condition. The resulting shape for a pond is realistic, so a useful closed-form solution is available. In the solution the soil is somewhat unrealistically assumed to be homogeneous with respect to permeability. Considerable progress has been made and more work is being done by the authors and others [3, 4, 5] on solution methods for flow in heterogeneous media; however, the idealized case is used here to prevent becoming ensnared by computational complexities.

The flow system is illustrated schematically in Fig. 1. Fluid standing in the pond of width $W$ at a potential $H$ seeps outward and downward to join the natural groundwater which has a gradient $S$. At a distance $L$, down-gradient, all of the flow is parallel to the polar axis of coordinates ($\theta = 0$). Two tracer sampling locations are shown in the figure along the polar axis. The equations describing the flow system and derivations required to analyse geometric travel times are presented elsewhere [6] in detail and will be only summarized here. The formulation is in terms of dimensionless variables which offer greater efficiency in computation and tabulation of results. The dimensionless variables are:

$$r = \frac{R}{L} \quad (1)$$

$$\phi = \frac{\phi}{SL} \quad (2)$$
where

\[ r \text{ is the dimensionless radius in spherical polar coordinates} \]
\[ R \text{ is the radius in any consistent set of units} \]
\[ L \text{ is the distance down-gradient from the pond to where the velocity is uniform} \]
\[ \phi \text{ is the dimensionless groundwater potential} \]
\[ \Phi \text{ is the groundwater potential} \]
\[ S \text{ is the uniform gradient of the groundwater flow if no pond were present} \]
\[ \varphi \text{ is the dimensionless stream function} \]
\[ \psi \text{ is the stream function} \]
\[ h \text{ is the dimensionless potential in the pond} \]
\[ H \text{ is the fluid potential in the pond} \]
\[ w \text{ is the dimensionless pond width} \]
\[ W \text{ is the pond width} \]
\[ t \text{ is the dimensionless time variable} \]
\[ T \text{ is the time with units consistent with } K \]
\[ K \text{ is the saturated hydraulic conductivity or permeability} \]
\[ F \text{ is the effective porosity with respect to the macroscopic velocity} \]
\[ v \text{ is the dimensionless macroscopic velocity} \]
\[ V \text{ is the macroscopic velocity through the soil as given by Darcy's Law} \]
\[ A \text{ is the dimensionless discharge rate of seepage leaving the pond} \]
\[ Q \text{ is the dimensional steady discharge rate of seepage leaving the pond.} \]

The equations from the earlier work [6] are:

*See Fig. 1.*
where

\[ A = \frac{\frac{W}{2L} \left( \frac{H}{2L} - 1 \right) \left( \frac{W}{2L} + 2 \right)}{2} \]  \hspace{1cm} (10)

\( \theta \) is the angular coordinate in spherical polar coordinates.

**Equation for the stream function**

\[ \varphi = \frac{1}{2} r^2 \sin^2 \theta + A \left[ \cos \theta - (r \cos \theta + 2) (r^2 + 4r \cos \theta + 4)^{-1} \right] \]  \hspace{1cm} (11)

**Equation for flow length**

\[ \sigma = \int_{r_0}^{r} (1 + I^2)^{\frac{1}{2}} \, dr \]  \hspace{1cm} (12)

where

\( \sigma \) is the dimensionless length of flow along a streamline

\( r_0 \) is the distance from the centre of the pond to the point where the streamline under consideration leaves the pond, and

\[ I = \left[ \frac{r \sin^2 \theta + (r \cos \theta + 2) (r + 2 \cos \theta) A - \frac{A \cos \theta}{\sqrt{r^2 + 4r \cos \theta + 4}}}{\sin \theta \left( \frac{2(r \cos \theta + 2)}{\sqrt{(r^2 + 4r \cos \theta + 4)^3}} - \frac{1}{\sqrt{r^2 + 4r \cos \theta + 4}} \right) A - r^2 \cos \theta + A} \right] \]  \hspace{1cm} (13)

Further \( r \) and \( \theta \) in Eq. (13) must also satisfy the equation for a streamline:

\[ r^5 \cos^5 \theta + \left[ \frac{(r^2 + 4)r - 4A}{4} \right] r^3 \cos^4 \theta + \left[ A^2 + (f - \frac{r^2}{2}) r^2 - \frac{(r^2 + 4) Ar}{4} \right] \left[ r \cos^3 \theta \right] 
\]

\[- \left\{ 8A \left[ f - \frac{r^2}{2} \right] r - \left[ r^2 + 4 \right] \left[ A^2 + (f - \frac{r^2}{2}) r^2 \right] + A^2 r^2 \right\} \cos^2 \theta
\]

\[ + \left[ 2r \left[ (f - \frac{r^2}{2})^2 - A^2 \right] - (r^2 + 4) (f - \frac{r^3}{2}) A \right] 2 \cos \theta + \left[ (r^2 + 4) (f - \frac{r^2}{2})^2 - 4A^2 \right] = 0, \]  \hspace{1cm} (14)

where \( f \) is the value for the stream function \( \varphi \) designating the streamline of interest.

**Equation for travel time along a flow line**

\[ t = \int_{r_0}^{r} \left[ (1 + I^2)/D \right]^{1/2} \, dr \]  \hspace{1cm} (15)
where

\[ I \text{ is defined by Eq. (13), and} \]

\[ D = \left[ \cos^2 \theta - \frac{A}{r^2} + \frac{(r + 2 \cos \theta)A}{\sqrt{(r^2 + 4r \cos \theta + 4)^3}} \right]^2 + \sin^2 \theta \left[ 1 + \frac{2A}{\sqrt{(r^2 + 4r \cos \theta + 4)^3}} \right]^2. \]  

Also, \( r \) and \( \theta \) in \( I \) and \( D \) (Eqs. (13) and (16)) must once again satisfy Eq. (14) identically.

**Equation relating actual velocity to macroscopic velocity**

\[ v^* = \frac{v}{F} \]  

where
- \( v^* \) is the actual velocity or movement rate through the soil
- \( v \) is the macroscopic Darcian velocity, and
- \( F \) is the effective porosity with respect to the macroscopic velocity.

Through use of the above equations the flow system is completely described macroscopically.

A tracer test in the selected flow system

The dimensions of a hypothetical flow system in which a tracer test is to be run are as follows:

- \( L = 200 \) ft
- \( S = 0.001 \) ft/ft
- \( H = 2.14 \) ft
- \( \text{W/2} = 14 \) ft
- \( K = 0.0464 \) ft/min. \hspace{1cm} (18)

Using these conditions and Eqs. (4), (5) and (10) we find that

\[ h = 10.70 \]
\[ w = 0.07 \]
\[ A = 0.70. \]  

These three parameters, which define the flow system, were used in Eqs. (9) through (13) to obtain the flow patterns shown in Fig. 2. The pattern depicts flow in a vertical plane passing through the polar axis (\( \theta = 0 \), see Fig. 1). Although computations were made in spherical polar form, Fig. 2 is shown in rectangular coordinates in the vertical plane for ease in use. The streamlines (\( \varphi = \) constant) are expressed as a fraction of the total flow leaving the pond; for example, one tenth of the pond flow passes between the lines \( \varphi = 0.6 \) and \( \varphi = 0.7 \). The equipotentials (\( \phi = \) constant) are similarly expressed as the fraction of the potential in the pond.

Sufficient information is now available to design a tracer test for the flow system shown in Fig. 2. Needed information is the addition time of tracer to the pond and observation-well specifications, i.e., location, type and approximate sampling duration. Examination of the distortion of tracer concentrations by different well types and locations is now considered.
Observation wells as monitoring structures

Observation wells are so widely used that often little thought is given to the errors that can be introduced by these measurement structures. The results from tracer observation wells usually fall somewhere between the precise measurement in a piezometer and the often erroneous results from a deep well which is perforated over its entire length and intercepts a non-uniform potential field. Such errors may or may not be particularly critical in tracer studies. The effects of the flow system of Fig. 2 on a deep perforated well will be compared to the effects on short piezometer observation structures.

The undesirable characteristics of deep perforated wells are illustrated through considering a well located at \( x = 0.35 \) and penetrating to \( z = 1.4 \) (Fig. 2). Fluid will move into the upper part of the well, where the potential is highest, then down the well and out near the bottom in an area of lower potential. The distribution of relative inflow and outflow with depth is shown in Fig. 3 as obtained from Eq. (9) and the differential form of Eq. (2) from Appendix A. The inflow rate is highest at the top of the well (\( z = 0 \)) gradually diminishing until at \( z = 0.555 \) no flow occurs either in or out of the well. At well depths greater than 0.555, progressively increasing outflow occurs and reaches a maximum at the bottom of the well.
The streamline ($\varphi = 0.36$), which passes through $z = 0.555$, significantly divides the flow system of Fig. 2 into two parts. Any streamlines having values greater than ($\varphi = 0.36$) are not sampled by the deep perforated well since only water outflow occurs below $z = 0.555$. For values of the stream function less than $\varphi = 0.36$, the well intercepts the tracer, but the observed concentration is distorted. The distortion is the combined effect of the inflow gradation and the varying arrival time; both are a function of well depth.

The tracer arrival time variations resulting from different flow lengths from the pond are given in Table I. These tabulated results [6] are obtained from Eq. (15), using the values of Eq. (19). Fig. 4 indicates the tabulated variation of tracer arrival time with well depth at $x = 0.35$. For instance, tracer leaving the pond at $t = 0$ arrives at the top of the well at $t = 0.018$. Similarly, if the tracer is fed continuously to the pond, then by $t = 0.053$ all of the fluid entering the well above $z = 0.35$ will contain tracer.

The inflow variation with depth is related to the macroscopic tracer concentration which would be observed in the well*. The cumulative flow

\* Macroscopic tracer concentration by definition ignores hydrodynamic dispersion by assuming that the arrival of a tracer is instantaneous. Hydrodynamic dispersion will be treated in a later section as a deviation from the macroscopic tracer concentration.
TABLE I

FLUID INFLOW AND TRACER ARRIVAL-TIME VARIATION FOR A DEEP PERFORATED WELL
(length = 1.4, location x = 0.35)

<table>
<thead>
<tr>
<th>Well depth (z)</th>
<th>Cumulative relative flow rate into well (q/q_T)</th>
<th>Tracer arrival time at well (t)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0</td>
<td>0.0180</td>
</tr>
<tr>
<td>0.025</td>
<td>0.084</td>
<td>0.0182</td>
</tr>
<tr>
<td>0.075</td>
<td>0.250</td>
<td>0.0193</td>
</tr>
<tr>
<td>0.125</td>
<td>0.405</td>
<td>0.0220</td>
</tr>
<tr>
<td>0.175</td>
<td>0.544</td>
<td>0.0260</td>
</tr>
<tr>
<td>0.225</td>
<td>0.674</td>
<td>0.0320</td>
</tr>
<tr>
<td>0.275</td>
<td>0.775</td>
<td>0.0382</td>
</tr>
<tr>
<td>0.325</td>
<td>0.856</td>
<td>0.0474</td>
</tr>
<tr>
<td>0.350</td>
<td>0.896</td>
<td>0.0530</td>
</tr>
<tr>
<td>0.375</td>
<td>0.915</td>
<td>0.0590</td>
</tr>
<tr>
<td>0.425</td>
<td>0.958</td>
<td>0.0732</td>
</tr>
<tr>
<td>0.475</td>
<td>0.985</td>
<td>0.0910</td>
</tr>
<tr>
<td>0.525</td>
<td>0.995</td>
<td>0.1120</td>
</tr>
<tr>
<td>0.555</td>
<td>1.0</td>
<td>0.1275</td>
</tr>
</tbody>
</table>

* See Appendix A for definition of q/q_T

entering the well q/q_T is the integral with respect to depth of the function in Fig. 3 [see Eq. (2), Appendix A]. For the case given, the integral to z = 0.35 has a value of q/q_T = 0.896. The other values for q/q_T in Table I are obtained similarly. The well depth is the linking variable in Table I for the time t and relative flow in the well q/q_T which is equivalent to the macroscopic tracer concentration, i.e.,

\[ \frac{C}{C_0} = \frac{q}{q_T}. \]  

(20)

The relationship for the deep perforated well between macroscopic concentration and tracer arrival time is plotted from Table I as curve A in Fig. 5. This curve provides a basis for comparing other observation wells with fewer perforations at the same location or comparing locations in the flow system to simplify tracer analysis. The other curves in Fig. 5 represent piezometers with a perforated length of 0.15, which corresponds to 30 ft in the actual flow system. The short perforated piezometers in
Fig. 4
Variation of tracer arrival time with depth for a deep perforated well located at $x = 0.35$

Well at $x = 0.35$
Perforated $z = 0-1.4$

Fig. 5
Fraction of traced flow entering the well at $x = 0.35$ as a function of time
The arrival-time variation of traced flow entering several sampling structures located at \( x = -0.35 \) in Fig. 5 have steep concentration-arrival curves which are desirable since shorter tracer tests can be used. At greater depths, such as in the piezometers, the slopes of the curves flatten since a wider range of flow-path lengths from the pond are intercepted by the short perforated length.

In Fig. 6 the macroscopic concentrations are shown for different arrival times at a new location, \( x = -0.35 \), and with several different perforation lengths in the observation wells. The cluster of curves starting at \( t = 0.025 \) are useful in examining the effects of perforated length of an observation well and the desirable time for adding tracer to the pond.

Curves such as those in Fig. 6 depict the delayed response at the well when starting a tracer instantaneously from the pond through the soil. At some later time the addition of tracer is stopped and the same shaped response curve, but displaced in time, describes the arrival of pure water at the well. Three displaced tracer and water response curves from Fig. 6 are shown in Fig. 7(a). The dimensionless time displacement, 0.004, represents a tracer addition time of between 3 and 4 d. At any time the tracer concentration which would be observed in the well is the difference in ordinates between the tracer and water arrival curves. For example, on curve A (representing a deep perforated well) at time \( t = 0.05 \), the over-all concentration, \( C/C_0 \), in the well is 0.37 minus 0.10 or 0.27. Values found in a similar manner are plotted in Fig. 7(b) and represent the relative concentration distributions which would be measured in the three observation well types.

Analysis for gradual replacement of pond fluid by tracer

For the analysis just completed the pond water of initially zero tracer concentration was assumed instantaneously changed to a concentration \( C_0 \).
In practice satisfying such an assumption is difficult, if not impossible. Therefore, the case is considered where only the concentration of tracer inflow to the pond is instantaneously increased, resulting in a gradual increase in the tracer concentration of fluid standing in the pond. Accordingly, the tracer concentration leaving the pond starts at zero and increases gradually to $C_0$ rather than abruptly as assumed earlier.

The effect of a gradually increasing concentration at an observation well can be analysed through use of Eq. (6) in Appendix B when combined with the methods already discussed. Fig. 8 shows successive concentration arrival. The base curve $C_0/C_d = 0$ in the figure is curve C from Fig. 6 plotted to an expanded time scale. The remaining curves are the sum of the base curve ($C_0/C_d = 0$) time abscissa, and the value for $t^i$ obtained from Eq. (6), Appendix B, for successive values of $C_0/C_d$. The curves for successively increasing concentration indicate the approach to equilibrium with time at each point along the well depth. However, the over-all concentration at a given time is required, so the combined variation of $q/q_T$ (from inflow to the well) and $C_0/C_d$ (from pond dilution) with time must be obtained.
The fraction of traced flow entering a piezometer as a function of time for a gradually increasing tracer concentration in the pond

The combined in-well effects of concentration and relative inflow rates are obtainable from Fig. 8. At a given value for $t$ the variation of $q/q_T$ (ordinates in Fig. 8) as a function of concentration is known. The known relationship is plotted in Fig. 9 for several values of $t$. The cumulative or gross relative concentration variation with depth and hence time in the well is

$$c_{\text{max}} \frac{C}{C_0} = \int_0^{c_{\text{max}}} \frac{(C/C_0)}{d(C/C_0)} d(C/C_0).$$

(21)

where

$C_0$ is the instantaneous concentration leaving the pond and later arriving at the well, and

$C'_0$ is the steady concentration fed to the pond.

Substituting Eq. (20) into (21) we find that

$$C/C_0' = \int_0^{c_{\text{max}}} \frac{(q/q_T)}{d(C_0/C_0')} d(C_0/C_0').$$

(22)

Equation (22) represents the relative concentration which would be observed in the well as a function of time. The right-hand term is represented by the area under the various time curves in Fig. 9.
The numerically evaluated integral is shown in Fig. 10(a) as curve A. Curve A represents the relative tracer concentration which would be obtained for a very long tracer test. Curve A in Fig. 10(b) is the concentration variation with time in the well if the tracer were added to the pond for a dimensionless time increment of 0.004 (3 - 4 d). In contrast, curve B in Figs. 10(a) and 10(b) from the earlier analysis is for the condition of instantaneously replacing all of the pond water. The comparison is for the best observation piezometer (perforated length 30 ft) at one of the better locations in the flow system. Any other location or perforated length can be analysed similarly. For example, curves C and D in Fig. 10 are for an identical piezometer except that it is located deeper (z = 0.305 to 0.455). The low flat curve in Fig. 10(b) indicates that the tracer test should be run longer (about 5 times) if desirable observations are to be made.

Hydrodynamic dispersion in tracer tests

The significance of hydrodynamic dispersion in tracer tests requires careful consideration. Unfortunately, the complexity in solving Scheidegger's equations [7] combined with the lack of measurements on soils for use in such solutions currently prevents desirably complete analysis. An indication of significance will be found through a linear approximate consideration in which the gently curving stream tubes shown in Fig. 2 will be treated as straight columns of equivalent length and of uniform cross-section. The
Tracer concentration responses in two piezometers for a gradually-increasing pond tracer concentration and for an instantaneous change in pond tracer concentration

analyses of BANKS and JERASATE [8] and OGATA [9] for longitudinal dispersion are then applicable and will be used as an indication of dispersion effects on a tracer test. The columns are sufficiently long to permit use of Eq. 41 from BANKS and JERASATE [8] with the assumption that no ion exchange occurs. This equation in terms of the dimensionless variables given earlier is

\[
\frac{C}{C_0} = \frac{1}{2} \left\{ 1 + \text{erf} \left[ \frac{\sigma}{\sqrt{4\alpha \sigma}} \left( \frac{1-(t'/t_0)}{1-t/t_0} \right) \right] \right\} .
\]

Letting \( M \) be the inverse error function of \((2C/C_0 - 1)\) and solving for \( t'/t_0 \) gives

\[
\frac{t'}{t_0} = 2M^2(\alpha/\sigma) + 1 \pm \sqrt{\left[(2M^2(\alpha/\sigma) + 1)^2 - 1\right]},
\]

where

- \( t' \) is the arrival time for fluid of concentration, \( c \)
- \( t_0 \) is the arrival time for \( C/C_0 = 0.5 \) and is equal to \( t \) as used elsewhere in this report
- \( \alpha \) is the dimensionless dispersivity characteristic value for the soil
- \( \sigma \) is the streamline length found from Eq. (12)
- \( M = \text{erf}^{-1} (2C/C_0 - 1) \)
- \( C \) is the tracer concentration
- \( C_0 \) is the tracer concentration in the pond.
Eq. (24) provides an estimate of dispersion effects for known conditions of $a$ and $\sigma$. The length, $\sigma$, is available from tabulation of Eq. (12) [6]. However, only meager data indicating the range of values for $a$ are available. Among these results are those of DAY and FORSYTH [10] and NELSON [11]. Consideration of these results indicates that an expected range for this problem between $5 \times 10^{-5}$ and $10^{-7}$ is reasonable for $a$. However, in view of the meager data a value of $10^{-4}$ will be used. If the results are still found to be small, even with such a large value, then ignoring the effects may not be hazardous.

![Tracer Arrival Time, $t$](image)

The time variation of tracer concentration reaching the piezometer as a result of hydrodynamic dispersion will be described in connection with the short piezometer shown as Curve D in Fig. 6. The piezometer is located at $x = -0.35$ and is perforated between $z = 0.305$ and $z = 0.455$. Fig. 11 shows the concentration gradation front at the well caused by hydrodynamic dispersion. The curve for $C/C_0 = 0.5$ is curve D from Fig. 6 and is
analogous to the base curve of Fig. 8. In fact the whole dispersion analysis follows the pattern as in the earlier case where the concentration leaving the pond gradually increased. Accordingly, a curve like Fig. 9 can be obtained from Fig. 11 and can be graphically evaluated. The appropriate integral is

\[ \frac{C}{C_0} = \int_0^{c_{\text{max}}} \left( \frac{q}{q_T} \right) d \left( \frac{C^*}{C_0} \right), \]  

(25)

where

\( C/C_0 \) is the concentration observed in the well, and
\( C^*/C_0 \) is the instantaneous concentration at one point along the perforated well depth.

![Fig. 12](image)

The effects of hydrodynamic dispersion on the observed tracer concentration in a piezometer.

The relative concentration due to dispersion, from an evaluation of Eq. (25), is shown in Fig. 12 as the dashed curve. The solid curve represents no dispersion in the flow system. The minor effects of dispersion seen only at the bottom and top of the curve are a direct result of Eq. (23) containing the probability integral. In the region of no effect, the concentration is a normal distribution of \( q/q_T \) (see Fig. 11). However, at the top and bottom of Fig. 11, truncation of the normal curve gives a dispersion effect shown in Fig. 12.
Fig. 13

The combination of all effects on tracer concentration in three observation structures
(with optimum matching)
Combination of individual effects

The combination of techniques already described enables synthesis of expected tracer concentrations at any location and with any well-sampling structure. Certain values of length of tracer addition time provide optimum peaks in the concentration-time curves thereby making the observations more accurate. The improved accuracy is obtained through careful matching of the time for tracer addition to a specific observation-well location. Such matching may enable an otherwise rather undesirable measurement structure to be optimally used. For instance, the results shown as curves C and D in Fig. 10(b) show significant improvement for the longer tracer addition time in Fig. 13(a).

Fig. 13 shows the concentration variations in two piezometers and in a deeper perforated well with ideal matching. Note that the dashed lines, representing only flow geometry effects, peak directly above the gross concentration solid curve. This condition provides an optimum peak and tends to reduce the effects of any approximation errors in dispersion analysis or incomplete mixing effects in the pond concentration treatment.

The effective porosity for a homogeneous soil can be found through use of the concentration peaks in Fig. 13. The dimensionless time value, \( t \), associated with the concentration peak is matched with the real time, \( T \), observed at the peak from the actual tracer test. The two numerical values for the time when combined with the values from Eq. (18) are used in Eq. (6) to calculate the porosity, \( F \). In homogeneous media the permeability can be obtained in a similar manner if the porosity is known independently.

CONCLUSION

The rather detailed discussion of factors to be considered in evaluating tracer arrival time has had three primary purposes:

1. To provide insight into the complexity and interactions occurring in tracer studies which often have been ignored or oversimplified;
2. To provide methods of analysis for combining the several interactions of the physical flow system in tracer use for groundwater studies; and
3. To provide a specific analysis reaffirming Skibitzke's thesis of limited present utility for tracers in field groundwater studies.

The specific flow system used to illustrate the methods is rather idealized as compared to most actual situations. However, the methods for combining individual physical effects are capable of handling complex flow systems providing a detailed basic flow analysis is available. The basic analysis of more complex systems is made possible through use of the continuum mechanics of flow in porous media or through extensions in this field. Therefore, the greatest need for further study and accomplishment lies in extending present knowledge in this area and if necessary, developing new frameworks for theoretical analysis of complex groundwater flow systems. Such developments should be directed toward describing multiphase diffusive flow in anisotropic, heterogeneous porous media. More specifically hydrodynamic dispersion requires considerably more study to enable better de-
scription, adequate measurement and subsequent definition of significance in tracer work.

Today, tracer tests are qualitative and of limited utility for many of the proposed applications. However, as theoretical advances are made to more sophisticated in-place analysis, in-field tracer tests may provide major contributions. Possible areas of future contribution are in the measurement of the more fundamental properties of porous flow systems, such as: porosity distributions, stream functions, and components of the in-place permeability tensor.

ACKNOWLEDGEMENT

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APPENDIX A

AN EQUATION FOR FLOW INTO AN OBSERVATION WELL IN EQUILIBRIUM WITH THE SURROUNDING POTENTIAL FIELD

Consider a plane source or sink of thickness dz providing flux at the rate dq. The potential is

\[ \phi - \phi = \frac{1}{2\pi K} \frac{dq}{dz} \log \frac{r_e}{a}, \]  

where
- \( \phi_w \) is the potential in the observation well
- dz is the elemental well length
- dq is the elemental flux into the well in dz
- z is the well depth having a range \( 0 \leq z \leq z_{max} \)
- \( r_e \) is the effective radius of influence of the well
- a is the radius of the observation well.

Rearranging Eq. (20) and letting \( r_e \) be independent of depth we find that, on integrating,

\[ q = 2\pi K / \log \left( \frac{r_e}{a} \right) \int_0^Z (\phi - \phi_w)dz, \]  

when \( z = z_{max} \) then \( q = 0 \) since inflow to the well must equal outflow. Further Eq. (2) has a maximum when \( \phi - \phi_w = 0 \) as seen from Eq. (1). Let \( Z_T \) be the z associated with \( \phi_w \) when \( \phi = \phi_w \). Equation (1) can be expressed as two integrals and using \( q = 0 \) when \( z = z_{max} \) provides

\[ \int_0^{Z_T} (\phi - \phi_w)dz = -\int_{Z_T}^{z_{max}} (\phi - \phi_w)dz. \]  

Let N be the value of the left side of Eq. (3) then using (2) we find that for
the total flow in the well $q_T$

$$q_T = 2\pi KN / \log (r_e / a). \quad (4)$$

Dividing Eq. (2) by (4) we find that

$$q/q_T = 1/N \int_0^Z (\phi - \phi_w) \, dz. \quad (5)$$

Equation (3) can be used with Eq. (9) in the report to determine $\phi_w$ which is the well potential and the associated depth, $z_T$. The value of $N$ is also obtained in the process. Evaluation of Eq. (5) using $N$ and Eq. (9) for the well location of interest gives the relationship of $q/q_T$ to $z$.

**APPENDIX B**

**EXPRESSION FOR GRADUAL INCREASE OF TRACER CONCENTRATION IN A POND OF CONSTANT INFLOW AND OUTFLOW**

Consider a pond of volume $V_0$ which initially has zero tracer concentration. At $t = 0$ the steady inflow rate, $Q$, instantaneously changed to concentration $C_0$ then for a differential element of time $d\tau$ the differential amount of tracer entering, $dA_{\text{in}}$, is

$$dA_{\text{in}} = C_0 Q \, d\tau. \quad (1)$$

Similarly, the differential amount leaving the pond by seepage, $dA_{\text{out}}$, is

$$dA_{\text{out}} = C_0 Q \, d\tau, \quad (2)$$

where $C_0$ is the instantaneous concentration in the pond. But the change of concentration in the pond is

$$dC_0 = (dA_{\text{in}} - dA_{\text{out}})/V_0. \quad (3)$$

Substituting (1) and (2) into (3) we find that

$$(1/C_0) dC_0 = (1 - C/C_0')(Q/V_0) d$$

or

$$\ln (1 - C_0/C_0') = - Q\tau/V_0. \quad (4)$$

Substituting dimensionless parameters (Eqs. 1 through 8) from the body of the report we find that for Eq. 4

$$\ln (1 - C_0/C_0') = - 2\pi AF t'/ V_0'. \quad (5)$$
where \( V_0^t = \frac{V_0}{L^3} \) is the dimensionless volume of fluid the pond. \( t' \) is the dimensionless time increment due to ponding.

Equation (5) when solved for \( C_0 \) can be substituted into any of the equations obtained for an instantaneous concentration \( C_0 \), leaving the pond to yield conditions for a gradually increasing pond concentration. The equation has one disadvantage in that it contains the effective porosity, \( F \), which in some cases is to be measured by the tracer test. In the results presented in this paper the value of \( F = 0.30 \) was used to simplify description. For an actual situation the simultaneous condition could be met, if necessary, iteratively with successive improvements of the estimated \( F \) value.

REFERENCES


DISCUSSION

Y. HARPAZ: Have you tried to compute a dispersion coefficient by applying the analysis described in the paper to the actual field measurements?

R. W. NELSON: No. At the moment we are working hard on the problem of determining the permeability distribution for the whole project, using the methods described in references [3 and 4]. Once this has been established and once the flow system has been simulated (see reference [5]), it will be possible to make use of the field tracer test - such as those referred to by Dr. Theis - for which data are already available.

Y. INOUE: Are there any impermeable strata in the formations under study?
R. W. NELSON: In the problems we analysed the soil was assumed to be homogeneous. However, at Hanford all the soils are in fact heterogeneous. At present we are concentrating on theoretical calculations and in-place measurements of the heterogeneous permeability distribution.
NOTE SUR L'UTILISATION ÉVENTUELLE DES COMPLEXES DE NITRONITROSYLRUTHÉNIUM COMME TRACEURS EN HYDROLOGIE

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Abstract — Résumé — Аннотация — Resumen

POSSIBLE UTILIZATION OF NITRONITROSYLRUTHENIUM COMPLEXES AS TRACERS IN HYDROLOGY. Ru$^{106}$ might be a useful tracer in hydrology. Its half-life of about one year is in a range in which there are few radioisotopes which can be used as tracers.

There are a great variety of complexes of Ru$^{106}$, the nitro-complexes of nitrosylruthenium being amongst the most stable.

Percolation tests have been made with nitronitrosylruthenium diluted in water from the mains, on columns of aquiferous sand and a very argillaceous soil.

NOTA SOBRE EL POSIBLE EMPLEO DE LOS COMPLEJOS DE NITRONITROSILRUTENIO COMO INDICADORES EN HIDROLOGÍA. El Ru$^{106}$ podría emplearse en calidad de indicador en hidrología. Tiene un periodo de un año que se sitúa en una gama en la que existen muy pocos radioisótopos utilizables como indicadores.

El autor ha realizado ensayos de percolación con nitronitrosilrutenio, restringidos a dos aguas de grifo, en columnas de arena y en un suelo muy arcilloso.

Le W$^{66}$Ru pourrait présenter un intérêt comme traceur en hydrologie pour diverses raisons:

a) sa vie moyenne, environ 1,0 a, le situe dans une gamme où il existe peu de radiotraceurs,
b) les rayonnements β durs et γ de son descendant, le W$^{66}$Rh, le rendent aisément détectable,
c) le W peut, en théorie, être obtenu en grandes quantités à partir de résidus radioactifs.
a) Le ruthénium est un élément de transition, susceptible de subir de nombreuses complexations. Une variété de complexes prédomine dans les eaux résiduaires radioactives, le nitrosylruthénium RuNO dans lequel le ruthénium est en général considéré comme trivalente [1]. Il est également admis que c'est cette variété de complexes qui se déplace à de très grandes distances, à une vitesse assez voisine de celle de l'eau, autour des installations de rejet dans le sol d'eaux résiduaires radioactives. Une étude de la littérature disponible sur ce sujet suggère (ce point n'a pas été, toutefois, établi avec certitude) que les nitrocomplexes de nitrosylruthénium, les plus résistants à l'élévation du pH due à la dilution des résidus liquides acides dans les eaux souterraines, pourraient être la variété de nitrosylruthénium la plus mobile [2, 3, 4].

Par exemple, entrerait dans cette catégorie de complexes le dinitronitrosylruthénium, dont la formule est:

\[ [\text{RuNO(NO}_2\text{)}_2(\text{NO}_3\text{_2})(\text{H}_2\text{O})]\text{]}^+ \text{ Na}^-. \]

b) Du nitronitrosylruthénium fut préparé suivant une méthode indiquée par Fletcher et ses collaborateurs [5]. Du tétrachlorure de ruthénium stable (10 mg) marqué au $^{106}$Ru, dilué dans 100 ml d'acide nitrique 1N, fut soumis pendant 3 h à un barbottage de dioxyde d'azote NO, le mélange étant maintenu à une température de 70°C. Le mélange vira au rouge-brun, puis au jaune, cette dernière coloration étant caractéristique des complexes de nitronitrosylruthénium. Un chromatogramme à la méthylisopropylcétone montra que le mélange contenait, en proportions égales, des complexes mono et dinitro de nitrosylruthénium. Cette préparation fut répétée avec du nitratonitrosylruthénium, de $^{106}$Ru sans entraîneur; les résultats du chromatogramme à la méthylisopropylcétone furent les mêmes.

c) Les nitrocomplexes de nitrosylruthénium ainsi obtenus furent dilués dans de l'eau de ville et le pH réajusté à 7. La solution ainsi obtenue, contenant 0,1 ppm de Ru, fut refroidie au voisinage de 0°C et soumise à une percolation sur une résine échangeuse d'anions Lewatit M 600 sous forme mixte NO$_2^-$-NO$_3^-$. Une quantité correspondant à 93% de la radioactivité fut retenue. Parallèlement, la solution de Ru dans de l'eau de ville neutralisée fut percolée sur une colonne de résine échangeuse de cations Lewatit S 100. Une quantité correspondant à 10,5% de l'activité fut retenue.

De ces résultats, on peut déduire que:

1. Le nitronitrosylruthénium préparé dans ces conditions est un mélange de complexes anioniques et cationiques, avec prédominance évidente de la première de ces formes.
2. Aucune variété neutre de ces complexes n'est mise en évidence par l'addition des pourcentages de Ru retenus respectivement sur les résines anion et cation.

d) Une colonne de sable aquifère de Beauchamp, dont la capacité d'échange de cations est de 4,0 meq/100 g, fut alimentée avec la même solution de nitronitrosylruthénium dilué dans de l'eau de ville à pH 7 (0,1 ppm de Ru). La courbe de traversée du radioisotope est représentative du volume mort de la colonne, mais la concentration en Ru dans l'effluent est, au maximum, 35% de la concentration initiale, après trois volumes morts.
de colonne. La colonne étant de petite taille (20 cm de longueur, 2,5 cm de diamètre), on n'essaia pas de préciser l'allure de la courbe de traversée du Ru par comparaison avec la courbe de traversée de tritium.

Par contre, la solution effluente fut percolée à travers une colonne fraîche de sable après 24 h de séjour dans une étuve à 40-50°C suivi de réfrigération au voisinage de 0°C immédiatement avant percolation. De nouveau, 61% seulement de l'activité introduite furent retrouvés dans l'effluent.

Ce résultat suggère qu'il pourrait exister, au sein d'un mélange de complexes de nitronitrosylruthéniun, des réactions d'équilibre tendant à restaurer les variétés qui pourraient être artificiellement retirées du mélange.

e) La traversée du nitronitrosylruthéniun en solution dans de l'eau de ville (> 10^-6 ppm en Ru) fut comparée à celle du tritium sur une colonne.
de sol prélevé au Centre d’études nucléaires de Saclay. Les dimensions de la colonne étaient:
- longueur: 43 cm,
- diamètre: 11,5 cm.

La fraction argileuse du sol de Saclay est constituée de:
- montmorillonite: 40%,
- kaolinite: 40%,
- illite et divers: 10%.

Autres caractéristiques:
- oxydes de fer: 1%,
- capacité d’échange de cations: 21 meq/100 g.

La figure 1 montre, sur un même diagramme, les courbes de traversée respectives des deux isotopes. On voit que:
Alors que la rétention de l’eau tritiée sur la colonne n’excède pas 5%, celle du nitronitrosylruthénium atteint 95% environ.

L’eau tritiée était mesurée par scintillation liquide avec une précision statistique de ± 5%.

f) En conclusion, notons que:
1. Le nitronitrosylruthénium, tel qu’il a été préparé, ne comporte pas, en proportions décelables, de complexes neutres. On a préparé des complexes où les variétés anioniques prédominent, et qui peuvent être retenus par les minéraux argileux et/ou les oxydes métalliques.
2. Le pourcentage de nitronitrosylruthénium susceptible de se déplacer à grande distance avec les eaux souterraines est très difficile à prévoir, car il dépendrait à la fois des propriétés sorptives des sols traversés et de l’évolution, en fonction du temps, des mélanges de complexes désignés sous l’appellation «nitronitrosylruthénium».

RÉFÉRENCES

DISCUSSION

J. F. HONSTEAD: I am wondering whether this investigation into the possibility of using Ru⁶⁺ as a tracer has not perhaps thrown some light on a rather different problem, namely, whether there are chemical forms of this substance that might be appropriate, for example in waste-disposal operations, for preventing the movement of radioruthenium through the ground.

A. MENOUX (on behalf of C. GAILLEDREAU): Of course, hydrologists and waste-disposal experts approach this problem from diametrically opposed points of view. The former are interested in finding radioactive
substances like tritium that will move through the ground at the same speed as the water in which they are contained, while the latter are anxious to discover ways and means of reducing the migration of radioelements in the soil.

In the case of ruthenium, it was found as the result of waste-disposal operations in the United States that this substance does in fact travel considerable distances through the ground. That is why it seemed reasonable to consider the possibility of using it as a tracer. However, Mr. Gailledreau's paper suggests that ruthenium is retained relatively easily by soils, which would debar its use as a tracer. The fact that the ruthenium travelled a considerable distance from the disposal point in waste-disposal operations seems to suggest that there are other factors involved that are independent of the retention-capacity of the soil. I do not know what the reason is. Perhaps we shall find out eventually. In any case this would seem to be a clear case for co-operation between the hydrologists and the waste-disposal experts.
MEASUREMENT OF RIVER-BED VARIATIONS DURING A FLOOD

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Abstract — Résumé — Аннотация — Resumen

MEASUREMENT OF RIVER-BED VARIATIONS DURING A FLOOD. Despite all efforts to measure the variations of a river bed during a flood, no suitable measuring process has yet been discovered. To arrive at a definite solution to this problem, the authors applied a type of backscattering, gamma-ray densimeter to such measurements. As for the character of such a gamma-ray densimeter, fundamental research was carried out in a laboratory preceding field measurements at Kurihashi, the selected site on the River Tone, where a steel tube had been driven in as an access tube. The densimeter was suspended in this tube, to determine a boundary face between the water and river-bed layers.

Such measurements were carried out several times in 1960 during both median and flood hydro-conditions; variations of the river bed were measured during the inundation of the June, 1961 flood season. As a result, it was found that scarring began as the water level went up, presenting a clear-cut difference in the densities of river-bed and water layers. Piling up began as the water level went down. Although a newly formed layer of such a pile was comparatively low in density, it was stabilized as gradually compressed. The highest water level was 5.5 m during this observation while the deepest scar was 3.3 m.

MESURE DES VARIATIONS DU LIT EN PÉRIODE DE CRUE. Malgré toutes les tentatives qui ont été faites en vue de mesurer les variations du lit en période de crue, il n’existe pas encore de moyen satisfaisant d’y parvenir. Pour aboutir à une solution définitive de ce problème, les auteurs ont eu recours à un densimètre à rétrodiffusion gamma. La mise au point de ce densimètre a fait l’objet de recherches approfondies en laboratoire; l’appareil a ensuite été utilisé pour faire des mesures réelles sur le Toné, à Kurihashi. A l’endroit choisi, on a enfonce un tube d’acier qui devait servir de colonne d’accès. Dans ce tube était suspendu le densimètre en vue de déterminer l’interface de l’eau et des couches constituant le lit.

Les mesures ont été effectuées à plusieurs reprises en 1960, en régime moyen et en régime de crue; les variations du lit ont été mesurées en période d’inondation pendant la saison des crues, en juin 1961. Elles ont permis de déterminer que la formation de trous s’amorce quand le niveau de l’eau s’élève, en repérant la différence très nette de densité entre le lit et les couches d’eau. Des sédiments ont commencé à se déposer quand le niveau de l’eau s’est mis à baisser. Bien que la couche de sédiments nouvellement formée air eu une densité relativement faible, elle s’est stabilisée au fur et à mesure de sa compression. Au cours de ces observations, le plus haut niveau d’eau enregistré atteignait 5,5 m et le trou le plus profond 3,3 m.

ОПРЕДЕЛЕНИЕ ИЗМЕНЕНИЙ ДНА РЕКИ ВО ВРЕМЯ НАВОДНЕНИЯ. Несмотря на настойчивые попытки пока еще не найден подходящий метод определения изменений дна реки во время наводнений. Очередная попытка состояла в применении измерительной плотности обратного рассеяния гамма-излучения для определения границ соприкосновения между водой и слоями дна реки. Измеритель, подвешенный в стальной трубе, был помещен во вспомогательную трубу. Полевые измерения производились на участке у моста Курихаси на реке Тоне. Опыт предшествовал тщательно лабораторная подготовка. Несколько замеров было проведено в течение 1960 года в условиях среднего уровня воды и половодья. В июне 1961 года опыты проводились в период половодья. В результате установлено, что размывание дна начинается во время подъема уровня воды, и это является показателем четкого различия в плотностях дна реки и слоев воды. Наносы образуются во время спада воды. Хотя вновь образовавшийся слой наносов имел сравнительно низкую плотность, под действием постоянного давления он становился устойчивым. Самый высокий уровень воды в период наблюдения составлял 5,5 м, а глубина самой глубокой вымоины - 3,3 м.
MEDICIÓN DE LAS VARIACIONES DE UN LECHO FLUVIAL EN ÉPOCA DE CRECIDA. A pesar de los meticulosos trabajos realizados para determinar las variaciones del lecho de un río en época de crecida, no se ha hallado todavía un método adecuado de medición de esas variaciones. Con objeto de contribuir a la solución de este problema, los autores han utilizado un densitómetro gamma de retrodispersión para efectuar tales mediciones. Las características de dicho densitómetro se estudiaron en el laboratorio antes de proceder a las mediciones en el lugar escogido (Kurihashi, río Tone), en el que se introdujo un tubo de acero que sirvió como vía de acceso. El densitómetro se suspendió en el interior del tubo para localizar la superficie de separación entre el agua y los estratos del lecho del río.

En 1960 se efectuaron varias mediciones de esa índole tanto en período de caudal mediano como de crecida; las variaciones del lecho del río se midieron en época de crecida (junio de 1961). Los resultados hallados fueron los siguientes: el proceso de formación de fisuras comenzó al elevarse el nivel del agua y al aparecer una diferencia acusada entre la densidad de los estratos del lecho y la de las capas de agua. La sedimentación se inició al descender el nivel del agua. Los estratos recién formados presentaban una densidad relativamente escasa, pero se estabilizaron conforme se iban comprimiendo. Durante este observación, el nivel superior del agua alcanzó a 5,5 m y la altura de la fisura más profunda a 3,3 m.

I. FOREWORD

The significance of surveying scouring of a river bed during a flood is well known and calls for no particular explanation now. On the contrary, at present no instance has ever been heard of where scouring of a river bed during a flood was effectively measured. The authors and their co-workers have been studying the process of surveying river-bed scouring by the use of gamma-ray back-scattering for a long time. They finally had an opportunity to employ the field survey installation at the Kurihashi site of the River Tone, by courtesy of the Upper Tone River Construction Branch. At this site four scouring measurements were performed, including those during the two periods of high water in 1960 and one of flood resulting from continuous seasonal rain in 1961. As a result, they sum up their findings in this report and, in addition, touch on significant points of the problem of surveying river-bed scouring by means of gamma-ray back-scattering.

II. MEASURING PROCESS

1. Measuring apparatus

An insertion-type density meter constructed on the basis of gamma-ray back-scattering was the apparatus used for these measurements. By the determination of densities, the boundary face of two different densities is determined and thus the position of the boundary face lying between river-bed material and the water layer.

The density meter applied to this measurement was one of Geiger-Müller tube type made as an experiment by the Institute of Physical and Chemical Research. Its probe was made of a brass casing of 30-mm diameter and 70-cm length.

The gamma-ray source placed at the farthest end of the probe was replaceable. The several lead segments for a shield placed between the source and the G-M tube were constructed so as to be adjustable in their length. The upper end of probe was connected to a rate meter with a 30-m cable. The probe had been tested and found waterproof.
2. The field installation

At the surveying site of river-bed scouring, a pipe was previously installed to facilitate suspension of the probe within it. The inside diameter of the pipe had to be as small as possible, allowing only the probe to pass. The wall thickness of the pipe also had to be as small as possible within limits for withstanding bending or any damage from water current. As for its material, aluminium or duralumin are desirable because of their small density and comparative strength. A steel pipe, however, may be all right for this application.

A steel gas pipe with a 48.5-mm outside diameter, 42.5-mm inside diameter and 3-mm wall thickness was used in this experiment. The length of pipe on the market was found too short for installation at the site for this purpose, so it had to be prolonged by welding; the welded seam was placed not too close to the river bed to be measured. The pipe was driven into the river bed as deep as 15 m under a median water level, and its upper end was fixed to a supporting steel arm extending from a bridge pier. The pipe was installed on the downstream side, 5 m from the first pier toward the second, of the bridge between Kurihashi and Koga, some 40 km northeast of Tokyo.

III. PROBLEMS OF A BACK-SCATTERING TYPE GAMMA-RAY DENSITY METER AS A BOUNDARY FACE METER

1. Density versus cpm curves for calibration

Each density meter probe has different distinctive calibration curves depending on the length of the shield, the radioisotope nuclide and its intensity, the material of the access tube, its inside diameter and wall thickness, etc., so a calibration curve requires a precise knowledge of these. In addition, the water content of the object to be measured causes some error. This error, however, can be eliminated where the river bed only is the object to be measured.

Generally, a calibration curve crosses zero at either zero density or infinite density and has its maximum value at a point from 0.8 to 1.0 density. Therefore, for the measurement of river-bed variations, the curve shows the cpm decreasing as the density increases. Fig. 1 gives the calibration curve for the case in which water filled the space between the probe and the pipe wall. To get this curve either 2 mc Co\(^{60}\) or 25 mc Cs\(^{137}\) was used, and the shield length was adjusted to 10 cm, 20 cm or 30 cm. The inserted pipe could not be kept free from water leakage in this attempt, so the measurement was performed with the pipe filled with water.

A river-bed density was around 1.8 to 2.1 in the gravel layer while water density was 1.0, so a larger cpm variation per unit variation of density within the range from 1.0 to 2.1 density was desirable for a calibration curve. Under such conditions Cs\(^{137}\) is most desirable as the gamma-ray source with a 10-cm shield length for surveying variation in a river bed.
The density difference between water and a gravel river bed is 1.0, with the result that, if a probe is lowered down in a pipe driven into the river bed, it gives a large variation in the counting rate of scattered gamma-rays across the boundary face of the river bed. Assuming that the river bed is quite flat, we find that the counting rate decreases not radically but gradually with some transient length because there is a shield with a certain length between the gamma-ray source and the G-M tube and that the G-M tube has a length.

In the case where the depth of the probe is measured from the lower end of a G-M tube of the probe, the behaviour of counting-rate variations for the probe entering a gravel layer from the water is as shown, by the gamma-ray sources applied, in Fig. 2. From this chart the length of the cpm variation zone in both the water and the gravel layers is found and set out in Table I. This table reveals the advantages of each condition of measuring as follows:

(a) The longer the shield, the larger the ratio of cpm in water to cpm in gravel. This trend was more noticeable in Cs\textsuperscript{137} than in Co\textsuperscript{60}. 

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{calibration_curve.png}
\caption{Calibration curve of density meter}
\end{figure}

<table>
<thead>
<tr>
<th>Shield Length</th>
<th>Source</th>
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<tbody>
<tr>
<td>A: 10 cm</td>
<td>Co\textsuperscript{60} 2 mc</td>
</tr>
<tr>
<td>B: 20 cm</td>
<td>Co\textsuperscript{60} 25 mc</td>
</tr>
<tr>
<td>C: 30 cm</td>
<td>Cs\textsuperscript{137} 25 mc</td>
</tr>
<tr>
<td>A': 10 cm</td>
<td>Cs\textsuperscript{137} 25 mc</td>
</tr>
<tr>
<td>B': 20 cm</td>
<td>Co\textsuperscript{60} 25 mc</td>
</tr>
<tr>
<td>C': 30 cm</td>
<td>Cs\textsuperscript{137} 25 mc</td>
</tr>
</tbody>
</table>

steel pipe (O.D. 48 mm, I.D. 42 mm)
<table>
<thead>
<tr>
<th>Nuclides</th>
<th>Shield length (cm)</th>
<th>A (cpm)</th>
<th>B (cpm)</th>
<th>B/A</th>
<th>C (cpm)</th>
<th>C/A</th>
<th>(C-A)/(B-A)</th>
<th>a (cm)</th>
<th>b (cm)</th>
<th>a/a+b</th>
<th>D (cpm/cm)</th>
<th>C²/D (cm)</th>
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</thead>
<tbody>
<tr>
<td>Co⁶⁰⁺⁺ (2 mc)</td>
<td>10</td>
<td>3500</td>
<td>6300</td>
<td>1.80</td>
<td>4800</td>
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<td>15</td>
<td>20</td>
<td>43</td>
<td>140</td>
<td>0.49</td>
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<td></td>
<td>20</td>
<td>850</td>
<td>2600</td>
<td>3.06</td>
<td>1300</td>
<td>1.53</td>
<td>25.7</td>
<td>25</td>
<td>20</td>
<td>56</td>
<td>50</td>
<td>0.72</td>
</tr>
<tr>
<td></td>
<td>30</td>
<td>280</td>
<td>1100</td>
<td>3.93</td>
<td>400</td>
<td>1.43</td>
<td>14.6</td>
<td>35</td>
<td>10</td>
<td>78</td>
<td>13</td>
<td>1.54</td>
</tr>
<tr>
<td>Cs¹³⁷⁺⁺ (25 mc)</td>
<td>10</td>
<td>11800</td>
<td>34000</td>
<td>2.88</td>
<td>19500</td>
<td>1.65</td>
<td>34.7</td>
<td>15</td>
<td>15</td>
<td>50</td>
<td>800</td>
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<td></td>
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<td>10.00</td>
<td>800</td>
<td>1.78</td>
<td>8.6</td>
<td>35</td>
<td>10</td>
<td>78</td>
<td>40</td>
<td>0.71</td>
</tr>
</tbody>
</table>

Remarks:

A: cpm in gravel
B: cpm in water
C: cpm in the boundary face between gravel and water
D: inclination of the counting rate at the boundary face
a: length of the cpm variation range in water
b: length of the cpm variation range in gravel
Variations of counting rate at the boundary of water and gravel

**Shield length**

- **O-O** 10 cm A \( \text{Co}^{60} \quad \text{A}' \)
- **△-△** 20 cm B \( \{ \text{(2 mc)} \quad \text{B'} \} \quad \text{Cs}^{137} \)
- **×-×** 30 cm C \( \text{C'} \) (25 mc)

Steel pipe (O.D. 48 mm, I.D. 42 mm)

(b) The ratio \( (C-A)/(B-A) \) is a coefficient representing the position of the boundary face in a cpm variation zone. This coefficient is smaller for a longer shield and also smaller for \( \text{Cs}^{137} \) than for \( \text{Co}^{60} \) applied as a gamma-ray source. In other words, with a long shield, a boundary face is located just at the point where cpm variation is about to cease as the probe moves from the water into the gravel layer.

(c) Between the shield length and length "a" of the cpm variation zone in the water layer the following relation exists: \( a = \text{shield length} + 5 \text{ cm} \). This 5 cm corresponds approximately to the length from the shield end to the lower end of the gamma-ray source.

(d) The length "b" of the cpm variation zone in the gravel layer tends to become shorter with increasing shield length. This fact indicates that the upper part of the G-M tube with a long shield has little effect on the counting rate in the gravel layer.

The above statement is characteristic of a flat river bed. Actually, however, scouring in a river bed does not necessarily produce a plane surface. Especially in this test the pipe driven into the bottom, down which a probe is inserted, might cause uneven scouring.

3. Different variation curves of cpm resulting from a rough surface

Various causes may account for a rough surface of a river bed. Here, however, we consider at what cpm change there occurs a boundary face of the river bed with conical scouring around the pipe. First, holes of 30 cm,
Various counting rate profiles for different shapes of the simulated river bed

- Flat surface
- Conical hole: 10 cm depth, 30 cm diameter
- Conical hole: 10 cm depth, 50 cm diameter
- Conical hole: 40 cm depth, 70 cm diameter
- Casing: 20 cm depth, 50 cm diameter

50 cm or 70 cm in diameter and of either 10 cm or 20 cm in depth, were made artificially around the pipe, and the cpm variation in the boundary face was surveyed by changing of the gamma-ray source and the shield length, which resulted in measurement data some of which were as shown in Fig. 3. For convenience the depth of insertion for the probe in the pipe was taken as for the boundary face of a river bed with a flat surface.

As clearly seen from Fig. 3, the counting rate curve for a flat river bed differs from that for a river bed with a conical cavity around the casing in having a peak before declining when the probe entered the gravel layer from the water layer. The most favourable condition for production of this peak was that of a conical hole of large diameter, deep, and with a long shield. Also Co$^{60}$ produced such a peak more readily than did Cs$^{137}$. This phenomenon was characteristic of a river bed which sloped down toward the pipe all around; so that, if measurements had previously been made under many geometrical conditions, the general condition of the pipe's surroundings could have been presumed from the findings thus made.

Where conical holes were produced, the position of the boundary face on the curve of the counting rate variations was different from that in the case of a flat river bed; i.e. it was closer to the gravel layer than in the case of a flat river bed. The greater the inclination of the hole and the longer
the shield, the more the boundary face position tends to shift toward the gravel layer. This trend is, in addition, more conspicuous with Co\(^{60}\) than with Cs\(^{137}\) as a gamma-ray source.

4. The accuracy of the gamma-ray back-scattering type density meter for analysis of a river-bed boundary face and its range of influence in the horizontal direction

The accuracy of a density meter for analysis in a vertical direction of the density of a boundary face is naturally so conspicuously affected by the configuration of the river bed that it requires too much experimental data for presentation here. The limits of accuracy, under the assumption that the river bed is flat, may be represented by the following expression: error = (standard deviation of counting rate)/(counting-rate gradient in boundary face).

The error for one-minute measurement is shown in c/b in Table I. The error was less than 1 cm for all cases except that of Co\(^{60}\) gamma-ray source with 30-cm shield. It is seen from Table I that a high degree of accuracy is obtainable with a Cs\(^{137}\) gamma-ray source and a 10-cm long shield. Such a high degree of accuracy, however, is not actually required. The one-minute measuring time is enough for good accuracy.

The approximate horizontal range will be discussed below. The counting rate represented in Table I must be in a geometrical series. For Co\(^{60}\), from three counting rates with different lengths of shield, deduct 80 each; this gives a geometrical series with a common ratio of 2.47. For convenience 80 may be taken as natural counts. If the shield length is increased by 10 cm, the length of path of gamma-rays emitted from the source reaching the G-M tube after scattering would be the distance corresponding to the product of 10 cm and a coefficient k, resulting in the following equation:

\[
1/2.47 = e^{-10k\rho\mu}.
\]

\(\rho\) represents the density of water. \(\mu\) are the equivalent mean mass-absorption coefficients of gamma-rays and the primary scattered rays from water. From this we find k\(\mu\) = 0.0905.

For Cs\(^{137}\), deduct 600 from the counting rate by different shield lengths and find a geometrical series of the common ratio 2.92 having k\(\mu\) = 0.107.

For Co\(^{60}\) and Cs\(^{137}\), \(\mu\) are approximately 0.065 and 0.086, so k is 1.39 for Co\(^{60}\) while it is 1.24 for Cs\(^{137}\). Assuming "a" as the distance from a gamma-ray source to the centre of the sensitive zone of a G-M tube, and c and b as the length of path of gamma-ray and primary scattering beam respectively, k = (b + c)/a. In this case, the point perpendicularly separated by \(r_{\text{max}}\) from the centre line of the shield is the maximum range, giving \(r_{\text{max}} = (a/2)(k^2 - 1)^{1/2}\). Therefore, \(r_{\text{max}} = 0.48a\) in water for Co\(^{60}\) while \(r_{\text{max}} = 0.37a\) for Cs\(^{137}\). However, \(\mu\) may be a little larger than the value quoted here. For "a", it is sufficient to take into account 10 cm in the side of a G-M tube in addition to the shield length and 3 cm in the side of the source, so that a 20-cm shield length would give \(r_{\text{max}} = 15.8\) cm for Co\(^{60}\) or \(r_{\text{max}} = 12.2\) cm for Cs\(^{137}\).
By the same calculation method, the counting-rate values in the gravel layer for a 20-cm shield length are $r_{\text{max}} = 13.7 \text{ cm}$ for Co$^{60}$ while $r_{\text{max}} = 5.8 \text{ cm}$ for Cs$^{137}$. At a boundary face these would be intermediate values. The $r_{\text{max}}$ thus found should be taken a little smaller in consideration of the $\mu$ correction for energy loss in scattering. The maximum allowable radius of measurement would be approximately 14 to 15 cm for Co$^{60}$ and 8 to 10 cm for Cs$^{137}$.

IV. FINDINGS AND EVALUATION OF FIELD MEASUREMENT

In view of the fact that the effectiveness of a gamma-ray density meter was brought to light in investigations of variations of the river bed, field measurements were carried out four times in medium-water and high-water conditions, including one preliminary measurement performed in 1960. These measurements revealed the expected effectiveness of the gamma-ray density meter. There was a chance to see a flood of a medium scale in 1961 although there was none in later 1960. In this flood, investigation of river-bed variations was carried out for several days, so several typical measurements made on those occasions will be cited here.

1. Findings of measurement during medium-water conditions

Measurement was carried out on medium water (120 cm deep) on 7 May 1960. With Co$^{60}$ (different from the gamma-ray source shown in Table I) and a 10-cm shield length, density distributions discovered in the river bed gave the data shown in Fig. 4. The density, 1.86, found at the river-bed surface increased gradually as the depth increased to reach a constant value of 2.16. Under this gravel layer was a mud layer mixed with plant remains, giving a density of 1.30 as a minimum value.

The density profiles similarly found with Cs$^{137}$ as the source with a 20-cm shield length were those shown in Fig. 4. In this instance, the length of the varying zone of the counting rate was 60 cm, while it was 40 cm with Co$^{60}$ in place of Cs$^{137}$. The peaks of the counting rate before decreasing differed by 10 cm for Co$^{60}$ and Cs$^{137}$ because of the difference between shield lengths. Assuming that the boundary face is based on the peak of the counting rate, the boundary face is located 5 cm down plus the shield length, so it is at a 13.65 m position for either Co$^{60}$ or Cs$^{137}$. Again, assuming the boundary face from rate of decrease of these counting rates, as seen in the column $(C-A)/(B-A)$ in Table I, 13.7 m is found for either Co$^{60}$ or Cs$^{137}$. The difference between 13.7 m and 13.65 m may be attributable to the fact that the measurements of the probe position were performed every 10 cm.

Taking 13.7 m as the river-bed surface, the variation zone of counting rate in the gravel layer is 20 cm for either Co$^{60}$ or Cs$^{137}$. For a flat river bed this value agrees with the value for Co$^{60}$ of Table I while not agreeing with the one for Cs$^{137}$, being too long by 5 cm. This 5 cm too can be explained by the fact that the measurements of the probe position were performed every 10 cm.
2. Findings of measurements during high water

The River Tone had high water on 12 and 13 August 1960, reaching a depth of some 4.4 m at a gauging site. Findings of the measurement made by means of Co$^{60}$ with a 10-cm shield length at 19.00 on 12 August were as quoted in Fig. 5.

As seen in this chart, fluctuations of the counting rate were large across the river bed. This fact might indicate an unstable river-bed surface with continual movement of gravel. In this chart a peak can be observed before
the counting rate decreases. This means that a conical hole has been made around the pipe because of scouring occurring about it. This hole is presumed to be some 10 cm deep with a 50-cm diameter. With such a configuration taken into account, the boundary face was located at 14.1 m depth. Again, from the decreasing rate of the counting rate, it was also found at 14.1 m depth. The length of the counting rate zone of variation was 20 cm for a conical hole in the gravel layer while in the chart it was as large as 70 cm. In addition, a large fluctuation of the counting rate was observable all over the variation zone. Such a fact is enough to indicate that gravel was moving violently and piling up because of the disturbance to which the river-bed surface was subjected. The disturbance affected the river-bed as deep as 14.9 m.

Similar measurements were carried out 14 h later, resulting in the findings given in Fig. 5. When these are compared with those found in the measurements made on the previous day, it is noticeable that

(a) The later measurements showed a slight decline in water level;
(b) They had a peak on the time versus cpm curve, just as those of the previous day had;
(c) In the later measurements the river bed had deepened a little; and
(d) The variation band of the counting rates in gravel was 20 cm between the two sets of measurements.
It was observable that the violence of river-bed disturbance died down a little and considerably stabilized in comparison with the previous day. There was another high water on 21 August to an extent similar to that of 13 August, giving approximately the same measurements.

3. Findings of measurement during floods

Observation of river-bed scouring was continuously carried out for one week beginning on 27 June 1961 in a flood resulting from heavy rains preceding the seasonal Nyubai. The River Tone approached the high-water stage late at night on 27 June, so observation was begun at 14.00 on 28 June. At that time the river bed was found already to have deepened by 160 cm in comparison with that of median water on 12 June. The measurements were performed every hour except late at night. The observations made up till 1.00 on 30 June from the beginning are shown in Fig. 6. The findings of median water observed on 12 June are also depicted in this chart. The measurements made on 12 June had been resumed after approximately 10 months, since 21 August of the previous year. The river-bed position on 12 June was 13.2 m, showing a 1.28 m rise over that of 21 August of the previous year. A 180-cm-thick layer, from 13.2 m in depth up to 15 m, had less density than did the original river bed. This 180-cm-thick layer decreased in density as the depth increased. The density fell to 1.75 g/cm$^3$, then gradually increased. The river-bed position found at 14.48 m in measurements on 21 August had been scoured up to 15.00 m, and then 180 cm of gravel had piled up, as far as could be ascertained.

The position of the river bed at 14.00 on 28 June was 14.80 m. It deepened to 16.20 m at 1.00 on 30 June. It was conceivable from the configuration of the counting-rate curve that the river bed was approximately flat and that a marked difference of densities existed between water and gravel layers.

In the observation made at 2.30 on 30 June, however, the cpm curve was changing its shape gradually, as indicated in Fig. 7. There was a layer of small density piled up between 15.3 m and 16 m in depth at 2.30. This piled-up layer further developed as high as over 15 m at 7.30 and then to 14.8 m at 15.00. Subsequently it stabilized for a time at 14.8 m. The piled-up layer again developed, reaching the 13.8-m mark and at last went down gradually. In observations made after the high water had subsided, on 20 July, the upper edge of the piled-up layer was located at 14.2 m.

Even after piling up began in the river bed, scouring of the original river bed lasted for 14 h, resulting in some 40 cm of deepening of the river bed during that time. The scouring of the river bed followed by the formation of the piled-up layer might be taken as proof that the gravel layer was subject to disturbances and the piled-up layer itself was remarkably unstable. The fact that a piled-up layer was unstable was obvious from the constant, although small, variations in the perpendicular density distributions as time elapsed. An especially remarkable feature observed was the inversion of density. As shown in Fig. 8, for instance, the density decreased from top to bottom, but this range is short (about 50 cm); i.e. the density inversion was observable from 15.2 m to 15.4 m at 3.00, 7.00 and 11.00 on 2 July. Nevertheless, in the observation made at 15.00 the density inversion disappeared and was followed by the development of the piled-up layer.
Fig. 6

Scouring of the river bed during the flood (June 1961)

- - - 12 June
O - O 28 June, 15.00
X - X 29 June, 7.00
Δ - Δ 29 June, 10.00
□ - □ 29 June, 19.00
☆ - ☆ 29 June, 22.00
○ - ○ 30 June, 1.00
The piled-up layer reached the 280-cm mark on the evening of 3 July. Subsequently its upper end gradually came to have a higher density inside, as shown in Fig. 8. The position of the river bed, i.e. the upper end of the gravel layer with a density of 2.15, presented a gradual rise from that observed on the afternoon of 2 July. There was, however, no marked difference of density between the piled-up layer and the original gravel layer. It was conceivable that increase of density would probably have occurred because of contraction of the lower part of the piled-up layer. In the observation made on 20 July, two weeks having passed since the water subsided, the piled-up layer was found to be 200 cm thick. As seen in Fig. 8, the counting-rate curve reveals that the piled-up layer had been stabilized.
Fig. 8

Counting rate profile of the river bed after the flood (July 1961)

- 2 July, 3.00
- 2 July, 7.00
- 2 July, 11.00
- 2 July, 15.00
- 2 July, 18.00
- 3 July, 15.00
- 4 July
- 8 July
- 20 July
The above statement covers observations made on river-bed variations occurring during a flood. Those variations are compared with those of the water level in Fig. 9. According to this chart, the river bed had been scoured as much as 160 cm before the first observation was made at 14.20 on 28 June. The water level at this moment had not yet reached the first peak. The fact that over half of the whole 3-m depth of scouring had already occurred is very suggestive. Although the river bed gradually deepened, fluctuating repeatedly a little in depth until the water level reached the second peak, it suddenly deepened several hours before the water level reached the second peak, piling-up began and the layer developed as the water level decreased. However, in an earlier period of decrease of the water level, scouring had also taken place in connection with piling-up. As this peak was about to pass, inversion of density was observed. After that, as the water level decreased, the piled-up layer developed, stabilized and contracted.

Further, in the upper part of the newly-formed, piled-up layer, we had to account for the existence of a layer with an especially small density for a river bed. Although the properties of a layer with such small density had not yet been confirmed, when the fact that the mud layer mixed with plant remains had a density of some 1.3 lying under the gravel layer of the river bed, it might be conceivable that the density of river-bed material newly piled-up was low. These data may be analysed further.

V. CONCLUSION

The findings of field measurements and the method for investigation of the scouring of a river bed by means of a density meter of a gamma-ray back-scattering type have been discussed. Many problems are left for future analysis, so that from these findings the analysis of scouring is impossible. However, it is established that this method was effective and suitable for measurement of river-bed variation during a flood. In addition, we have
acquired much useful knowledge of scouring by taking advantage of this method.

A gamma-ray density meter for measurement of a boundary face of a river bed should have a 10-cm shield length with a gamma-ray source of Cs\(^{137}\) for the most desirable measuring conditions. The radius of the measuring range in a horizontal direction across a river-bed boundary face was 14 to 15 cm in the case of Co\(^{60}\) or 8 to 10 cm in the case of Cs\(^{137}\). The counting-rate curve often showed a peak with irregular river-bed surface.

The factors which disclosed the configuration of the river-bed surface were the appearance or non-appearance of a peak, the height and width of the peak, the length of the counting-rate zones of variation in both water and gravel layers, and the mode of counting-rate value fluctuation etc. The boundary face between the river bed and water layer could be found from either the decline in counting-rate values or both the point at which the counting-rate value began to decline in the water layer and the shield length. In such instances, the former in general had a higher degree of accuracy.

If river-bed scouring during a flood is plotted as a function of time, such periods are distinguishable from each other: the period in which only scouring was observable, then the period in which piling-up accompanied by scouring was observable, the one in which a piling-up layer and its stable development were observable, and the one in which the contraction of the piled-up layer was observable. Further, these periods could be seen to correspond with a curve of the water level.

During the flood of the River Tone which occurred in June 1961, a maximum scouring of 3.3 m with a highest-water level of 5.5 m was observed.

In order to make such a density meter an ordinary instrument for field measurements, more fundamental data have to be collected. Such a meter would be useful for investigation of the scouring around a bridge pier, bank revetment, seashore, etc., in addition to the measurement of river-bed scouring.

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DISCUSSION

L.O. TIMBLIN: If you had turned the probe upside down so that the detector had entered the river bed first, do you think that your determinations of the river-bed-water interface would have been more sensitive or less so?

O. KONDO: I should say that the sensitivity would be about the same. There would, however, be difficulties in connection with the arrangement of the electronic parts in the probe. In any case, a lead shield of at least 10 cm in length would have to be placed between the gamma-ray source and the G-M counter to intercept direct gamma radiation from the source.
FLOW MEASUREMENTS CARRIED OUT IN FRANCE WITH RADIOACTIVE TRACERS, USING THE TOTAL COUNT METHOD. The authors describe their experiments with the total count method in measuring the discharges of rivers and of hydroelectric plant penstocks. Their procedure was to sample a portion of the current during the passage of the activity wave by means of gauges and constant-level tanks. The sample and an aliquot part of the injected activity are counted in the same geometry. This method has several advantages over the conventional method using a submerged probe.

First, it avoids the difficulties presented by the counting geometry, both in rivers and in penstocks; it likewise makes it possible to determine the statistical counting accuracy and, by making several samplings in the measurement section, to verify whether good mixing conditions are present.

Furthermore, since it allows the use of a scintillation counter and concentration of the tracer by chemical means, the method, by comparison with the conventional technique, requires a minimal activity to yield a given degree of accuracy.

In conclusion, the authors give the results of measurements of discharges of rivers and penstocks and state their views on the future of the total count method.
MEDICIONES DE CAUDAL EFECTUADAS EN FRANCIA CON AYUDA DE INDICADORES RADIACTIVOS
POR EL MÉTODO DE INTEGRACIÓN. Los autores describen los trabajos que han realizado por el método del número total de impulsos para medir caudales en ríos y tuberías a presión de centrales hidroeléctricas. Operan separando con boquillas de aforo y cubas de nivel constante una parte de la corriente cuando pasa la ola de actividad; la muestra y una parte alícuota de la actividad inyectada se recuentan en iguales condiciones geométricas. Este método ofrece numerosas ventajas sobre el método clásico de la sonda de inmersión.
Entre otras, evita las dificultades que plantean las condiciones geométricas de recuento tanto en ríos como en tuberías a presión, permite establecer la exactitud estadística del recuento y, si se efectúan varias tomas en la sección de medición, comprobar si se respeta la condición de buena mezcla.
Por otra parte, al permitir el empleo de un contador de centelleo y concentrar el indicador por vía química, el método requiere, para una exactitud dada, una actividad pequeñísima en comparación con el método clásico.
Para terminar, los autores citan los resultados de mediciones de caudales de ríos y tuberías a presión y formulan algunas observaciones sobre las perspectivas del método del número total de impulsos.

1. CADRE DE CETTE ÉTUDE ET CHOIX DE LA MÉTHODE

Les études entreprises en France depuis bientôt deux années pour la mesure de débits d'eau à l'aide de traceurs radioactifs avaient pour objet d'évaluer les possibilités de la méthode décrite par Hull [1], connue en France sous le nom de méthode de dilution (procédé par intégration), et qualifiée dans les pays anglo-saxons de «gulp method», «total count method» ou de «sudden injection method».
En même temps était entreprise une recherche de la «méthode d'Allen», connue dans les pays anglo-saxons sous le nom impropre de «peak timing method» ou de «pulse dilution method»; nous ne faisons que la mentionner.
Ces études ont été menées en collaboration étroite par le Commissariat à l'énergie atomique et deux organes de l'Electricité de France dont les objectifs dans cette recherche, s'ils diffèrent dans leur cadre, ont un point commun important: la mesure de débits d'eau importants, supérieurs à 80 ou 50 m$^3$/s.
Le Département des essais du Centre de recherches et d'études de Chatou (CREC) étudie les différentes méthodes de dilution comparées pour la mesure des débits turbinés par une usine hydroélectrique de basse chute, soit directement, soit par l'intermédiaire de son canal d'amenée. La précision souhaitée de ces mesures est voisine du pour cent.
Rappelons qu'actuellement seul le moulinet permet un jaugeage dans ce cas; mais l'on peut reprocher à cette méthode d'être chère, d'une mise en œuvre lourde et de donner lieu à des dépouillements longs et fastidieux. Sa précision est voisine de 2%.
MESURES DE DÉBITS PAR LA MÉTHODE D’INTÉGRATION

Par ailleurs, la Division technique générale (DTG), qui exploite jour-nellement les différentes méthodes chimiques de dilutions comparées pour le jaugeage de rivières de montagne, espère trouver dans la méthode radioactive un moyen de mesurer des débits importants de rivières et des débits de fleuves. Les moyens classiques dont elle dispose ont le caractère commun d'être lourds, et c'est sur l'allègement du procédé de mesure que laissent espérer les traceurs radioactifs et surtout le tritium que reposent ses espoirs.

En effet le moulinet, qui est la méthode classique pour ce type de mesure, présente ici aussi l'inconvénient d'être cher et très lourd, voire dangereux par exemple dans le cas de crues.

Les méthodes de dilutions comparées avec utilisation des sels classiques en 1963 tels que le bichromate de soude ou les nitrites (méthode classique - régime permanent, et méthode d'intégration) seraient très lourdes et chères. Chères, car à l'encontre des traceurs radioactifs le prix du traceur croît linéairement avec le débit à mesurer, très lourdes, car le poids de sel (bichromate de soude) à injecter varie de 0,2 à 2 kg/m³s. Compte tenu de l'eau de dilution, il faut injecter, par exemple, 4 t de solution pour un fleuve d'un débit de 1000 m³/s, opération qui l'on peut qualifier de malaisée et de très lourde. On ne peut dans ces mesures destinées à déterminer le régime d'un fleuve ou au tarage d'appareils de jaugeage tolérer une erreur systématique, la précision souhaitée étant par ailleurs moins critique que pour les grands canaux et voisine de quelques pour cents. Un des caractères recherchés est, enfin, la légèreté du procédé, légèreté qui doit favoriser la multiplicité des mesures.

Avant d'aborder des essais en vraie grandeur, nous avons fait choix, pour mettre au point les différentes parties technologiques de la méthode, qui constituent, en notre sens, une phase essentielle de cette étude, de terrains simples où nous disposions de moyens classiques de référence.

Ainsi, dans la voie des mesures de débits de grands canaux, nous avons effectué un certain nombre de jaugeages de débits en conduites forcées d'usines hydroélectriques, tandis que, dans le but de mesurer ultérieurement des débits de fleuves, nous avons rodé la méthode radioactive sur les bords de rivières dont les débits étaient de l'ordre de quelques mètres cubes par seconde.

La méthode d'intégration a été préférée à la méthode de dilution per-manente pour la simplicité du matériel d'injection, en particulier dans le cas des courants de grande largeur. De plus elle nécessite de deux à trois fois moins de traceur que la méthode de dilution classique.

2. LES MÉTHODES D’INTÉGRATION EN COMPÉTITION

Nous avons jugé utile de rappeler les principaux avantages et inconvé-nients des différentes méthodes d'intégration dont nous avons déjà esquissé le principe général et que nous préciserons plus loin. Assurément, et ce sera l'idée qui se dégagera de cette analyse, seule la rhodamine B semble être actuellement en compétition avec la radioactivité pour la mesure de forts débits.
2.1. Le bichromate de sodium

Les méthodes de dilution comparées de bichromate de sodium ont fait en France, où elles ont vu le jour, l'objet d'un très grand nombre d'essais, et sont aujourd'hui utilisées systématiquement pour le jaugeage de rivières. Elles présentent donc un premier avantage, celui d'être bien connues. Le temps ayant contribué à les parfaire, elles sont actuellement les plus légères en matériel pour la mesure de débits de rivières inférieurs à 20 - 50 m³/s. La très grande solubilité du bichromate (600 g/l) peut être considérée comme un avantage de ce sel relativement à la rhodamine B, ainsi que nous le montrerons plus bas.

La concentration nécessaire pour son dosage est de l'ordre de 2 \cdot 10^{-7} kg/l pour des eaux assez claires, ce qui est souvent le cas des eaux en conduites forcées, et de 5 à 15 \cdot 10^{-7} kg/l en général pour les mesures de débits de rivières. La dilution étant de l'ordre de 10^6 entre le point d'injection et de prélèvement en rivière, ceci explique la masse importante de 2 kg/m³-s déjà mentionnée pour un fleuve. Il en résulte une masse et un prix prohibitif pour les mesures de forts débits.

Le risque de réduction du chrome par les berges ou les particules en suspension présente un inconvénient car il est nécessaire de réoxyder le chrome à sa valence la plus élevée et ceci allonge beaucoup le dépouillement des mesures. Ajoutons enfin comme inconvénient notable, mais mineur, que le chrome nécessite pour son dosage un réactif dont les propriétés évoluent avec le temps, et enfin qu'il est nécessaire de conserver les échantillons dans le noir car ils subissent une transformation sous l'action du soleil.

Nous rappellerons enfin que le bichromate de sodium est maintenant préféré à d'autres sels, toujours dosés par voie chimique, comme le sulfate de manganèse, le nitrite de sodium, le chlorure de sodium, principalement parce qu'il conduit au volume le plus faible de solution injectée, et également parce qu'on ne le rencontre pas à l'état naturel dans les rivières, et que son dosage colorimétrique ne fait appel qu'à un personnel peu initié.

2.2. La rhodamine B

De grands espoirs sont nés avec les premiers essais de la rhodamine B dont l'étude se poursuit actuellement dans de nombreux laboratoires.

Son extraction des solutions, avec par exemple de l'alcool isoamylique, permet de diminuer de deux à trois ordres de grandeur, par rapport au bichromate de sodium, la concentration minimale nécessaire à son dosage (10^{-9}kg/l) et, de là, la masse de produit à injecter.

Cet avantage est cependant moins marqué que semblent le montrer ces chiffres car la rhodamine B est moins soluble dans l'eau que le bichromate, et il en résulte que le gain en masse de solution à injecter n'est que de 20, facteur très intéressant cependant.

Si, par ailleurs, la rhodamine B montre une très grande stabilité chimique, il apparaîtrait qu'elle soit assez absorbée par les matières en suspension, ce qui serait un grave défaut pour son utilisation dans les fleuves en général très chargés.
Notons enfin qu'un spectrophotomètre de précision, matériel assez délicat, est nécessaire pour son dosage.

2.3. Chlorure de sodium et mesure de résistivité

La connaissance de la concentration de NaCl par une mesure de résistivité in situ et en continu représente un double intérêt de cette méthode. Cependant deux reproches principaux peuvent lui être faits. En premier lieu, le niveau souvent assez élevé de la conductivité naturelle des eaux conduit à l'injection de masses de sel trop importantes. En second lieu, les variations dans le temps et dans l'espace de cette résistivité affectent fréquemment ces mesures.

De plus la nécessité de connaître la température de l'eau alourdit quelque peu la méthode; nous savons en effet que la résistivité de l'eau varie environ de -2,5%/°C.

2.4. La méthode radioactive

C'est à Hull que revient le mérite d'avoir le premier présenté la méthode d'intégration avec des traceurs radioactifs. La mesure in situ et en continu est dans ce cas également possible à l'aide d'une sonde immergée et cela peut être considéré comme un avantage pour certaines études, par exemple du temps d'arrivée et du temps de passage de la vague de traceur.

La radioactivité apporte d'autres avantages. Le volume de la solution à injecter est incomparablement plus faible qu'avec les traceurs salins classiques, et le tritium sous forme d'eau tritiée laisse espérer une solution lorsque les traceurs salins actifs ou inactifs subissent une adsorption préjudiciable à la mesure. De surcroît, le prix des traceurs radioactifs, qui croît moins vite que le débit à mesurer, est un avantage intéressant à souligner.

Mais l'on peut reprocher aux traceurs salins radioactifs, utilisés en général en hydrologie, le poids des enceintes de protection destinées à leur transport, et par ailleurs, le phénomène de décroissance d'activité pour les traceurs à courte période, phénomène qui conduit à préparer et à transporter une activité bien supérieure à celle qui est effectivement utilisée.

Le tritium ne présente pas cet inconvénient et l'eau tritiée paraît donc la mieux appropriée aux mesures de débits importants. Cependant, une très grande prudence doit être recommandée, prudence qui doit prescrire toutes les mesures faisant appel au tritium, s'il peut être montré qu'une contamination durable des nappes en aval du point d'injection est possible. Cette contamination pourrait handicaper ou interdire dans ce milieu d'éventuelles études avec le tritium naturel ou d'origine thermonucléaire.

La mesure du tritium impose le prélèvement d'échantillons et leur comptage en laboratoire avec un appareil assez délicat et cher.

Une dernière particularité importante de la méthode d'intégration avec des traceurs radioactifs, à inscrire au chapitre des inconvénients, est la difficulté de connaître le rapport entre l'activité injectée et une activité aliquote destinée à l'étalonnage du système de détection. La détermination de ce rapport, qui varie de $10^9$ à $10^6$, fait appel en général à un dispositif
spécialisé et important, en particulier dans le cas d'émetteurs de rayonnement gamma dur.

3. PRINCIPE DE LA MÉTHODE - CONDITIONS FONDAMENTALES

3.1. Principe de la méthode

Une certaine quantité de traceur radioactif d'activité A est injectée dans la rivière, le canal ou la conduite selon une loi quelconque en fonction du temps.

A une distance suffisamment grande en aval, un comptage de la vague d'activité est réalisé à l'aide d'une sonde placée en regard, soit de l'écoulement (méthode directe), soit d'une déviation de l'écoulement (méthode d'échantillonnage continu). Une troisième méthode consiste à effectuer le comptage d'un échantillon moyen, prélevé à débit constant, pendant le passage de la vague d'activité (méthode de prélèvement ou méthode de l'échantillon moyen).

Nous considérerons en premier lieu la méthode directe.

Soit une sonde S est un volume élémentaire J dans l'eau du volume sensible de cette sonde.

Pendant un temps T, supérieur ou égal au passage de la vague d'activité, passe en J une activité:

\[ \text{d}A_j = \int_T c \text{d}Q \text{ d}t . \]

Le débit élémentaire \( \text{d}Q \) étant supposé constant (régime permanent en moyenne), nous pouvons écrire

\[ \frac{\text{d}A_j}{\text{d}Q} = \int_T c \text{d}t . \]

Nous supposerons l'existence d'une relation linéaire entre le taux de comptage \( n \) enregistré à l'aide de la sonde S et \( \text{d}Q \) à l'activité contenue dans le volume J, et la concentration \( C \) dans ce volume élémentaire:

\[ C = \frac{n}{F_j} , \]

\( F_j \) représentant le taux de comptage pour une concentration unitaire en J. Nous écrirons alors

\[ \frac{\text{d}A_j}{\text{d}Q} = \int_T \frac{n}{F_j} \text{ d}t = \frac{N_j}{F_j} \text{ en posant } N_j = \int_T n \text{ d}t , \]

ou encore

\[ N_j = F_j \frac{\text{d}A_j}{\text{d}Q} \]
Si \( \frac{dA_j}{dQ} \) est constant dans tout le volume \( \omega \) qui est «vu» par le détecteur, c'est à dire si \( \frac{1}{\varepsilon} \frac{cdt}{\omega} \) = constante (et ce sera la condition de «bon mélange»), le nombre total de coups = \( N \) enregistré pendant le passage de la vague active est

\[
N = \int_{\omega} N_j d\omega = \frac{dA_j}{dQ} \int_{\omega} F_j d\omega = \frac{dA_j}{dQ} F
\]
	en posant

\[
F = \int_{\omega} F_j d\omega ,
\]

et comme \( \frac{dA_j}{dQ} \) = constante, \( \frac{dA_j}{dQ} = \frac{A}{Q} \) s'il y a conservation de l'activité et \( Q = F \frac{A}{N} \)

où \( N \) = nombre total de coups enregistrés,
\( A \) = activité injectée
\( Q \) = débit à mesurer,
\( F \) = taux de comptage donné par une activité unitaire dans tout le volume intéressé.

Ce coefficient peut donc être déterminé par un étalonnage dans une géométrie strictement identique à celle dans laquelle est effectuée la mesure.

Une activité \( A_e \) est alors introduite dans une enceinte de volume \( V \) et l'on obtient dans ces conditions un taux de comptage \( f_e \).

On obtient alors

\[
F = f_e \frac{A_e}{A} V
\]

et

\[
Q = \frac{A}{A_e} f_e \frac{V}{N} V \quad \text{(1)}
\]

avec \( A \) = activité injectée durant l'essai,
\( A_e \) = activité injectée lors de l'étalonnage,
\( f_e \) = taux de comptage durant l'étalonnage,
\( N \) = nombre de coups enregistrés pendant l'essai,
\( V \) = volume de la cuve d'étalonnage

Cette identité entre la géométrie de mesure et la géométrie d'étalonnage pose, dans sa réalisation, un problème important et souvent difficile.

Il apparaît que pour une rivière ou un fleuve la meilleure solution est de placer la sonde dans des conditions de «volume infini», c'est-à-dire dans le volume au delà duquel la solution active contribue, par exemple, à moins de 1% du signal.
Ces conditions ne sont pas toujours aîsées à respecter comme nous le montrerons plus loin. De plus, dans le cas de conduites de forts diamètres, on ne dispose pas souvent d'éléments de conduites pour l'opération d'étalonnage, opération qui serait par ailleurs fort lourde de mise en œuvre.

Certains préfèrent donc la méthode d'échantillonnage continue à la méthode directe. Une partie du courant principal est dérivée, en général à l'aide d'une pompe, dans une cuve comprenant la sonde de mesure. L'étalonnage est effectué dans cette même cuve et par conséquent dans une géométrie identique.

Les relations établies plus haut sont valables pour cette méthode et la condition de bon mélange est toujours \( \int \frac{c}{dt} \) dans la section de prélèvement.

Nous avons préféré pour nos essais la méthode de prélèvement, exposée en détail plus bas, dans laquelle on effectue un prélèvement à débit constant \( q \) dans une cuve où est ensuite mesurée la concentration moyenne du traceur.

On fait l'hypothèse que la relation de bon mélange \( \int \frac{c}{dt} \) dans la section de prélèvement est respectée; ceci entraîne

\[
\frac{Q}{q} = \frac{A}{a}
\]

avec \( Q = \) débit à mesurer,
\( q = \) débit de prélèvement,
\( A = \) activité injectée,
\( a = \) activité prélevée.

Au cours de l'étalonnage une activité \( A_e \) donne une fréquence de comptage \( f_e \).

On peut écrire

\[
a = \frac{N}{A_e f_e}
\]

et il vient

\[
Q = \frac{A f_e}{A_e N} q
\]

3.2. Conditions fondamentales

3.2.1. Condition de bon mélange

Les relations (1) et (2) ont été établies en faisant l'hypothèse qu'en tous les points du volume de mesure ou de la section de prélèvement l'expression \( \int \frac{c}{dt} \) est constante. En conséquence, les masses de traceur qui traversent les différents tubes de courant doivent se répartir comme les débits. Il peut paraître à priori étonnant que la nature soit assez complaisante pour qu'il en soit ainsi, mais nous verrons que l'expérience nous en donne une excellente vérification.
3.2.2. Conservation de la matière ou de l'activité

Pour établir les équations (1) et (2), nous avons utilisé la relation \( dA/dQ = A/Q \), que nous avons écrite en faisant l'hypothèse qu'il y avait conservation de l'activité; il est donc nécessaire que toute l'activité injectée passe dans la section de mesure et éventuellement les sections dérivées.

3.3. Remarques sur les conditions d'applications de la méthode

3.3.1. Cas d'un affluent entre la section d'injection et la section de mesure

Il suffit que la distance entre le confluent et la zone de mesure soit suffisante pour que la condition de bon mélange soit respectée.

3.3.2. Cas d'une dérivation du courant principal

Les démonstrations des méthodes de prélèvement faites plus haut montrent que la mesure peut être réalisée sur une partie dérivée du courant principal. Nous rappellerons seulement que l'expression \( I \) doit être constante dans la section de la dérivation. La «distance de bon mélange» concerne ici la distance entre la section d'injection et la section de dérivation.

4. PROCESSUS DE LA MESURE

Dans ce chapitre nous décrivons les différents processus de mesure de débits correspondant à nos propres essais sur les conduites forcées et des rivières de faibles débits. Nous ne ferons que de courtes allusions aux autres méthodes d'intégrations radioactives, et des suggestions pour l'application de la méthode à la mesure de forts débits qui, rappelons le, est notre objectif.

4.1. Prise de partie aléatoire de l'activité principale

Nous insistons dans ce paragraphe sur l'étalonnage des sondes de détection, question qui n'est, à notre sens, pas assez souvent mise en relief.

Dans les équations (1) et (2) apparaît le rapport \( A/A_e \) entre l'activité injectée durant la mesure et l'activité injectée pour l'étalonnage.

Si l'on désire connaître le débit avec une précision de quelques pour cents, ce rapport doit être connu avec une précision de quelques pour milles. Il est important de noter par ailleurs qu'il est difficile de comparer deux activités qui diffèrent de 4 à 6 ordres de grandeur.

Si l'utilisateur peut faire appel à un laboratoire de mesures radio-métriques spécialisé, le problème est simplifié pour lui, car il reçoit des activités étalonnées. Il ne doit alors veiller qu'aux variations des efficacités de ses sondes entre les dates d'étalonnage et de mesure, le mieux étant, bien entendu, que ces dates soient les plus voisines possible.

Nous ne disposons pas à Grenoble de laboratoire spécialisé dans ce type de mesure. La solution que nous avons adopté consiste à préparer
par dilution volumétrique deux solutions dont les concentrations sont connues. Ces opérations doivent être conduites à distance et derrière un mur de protection. Nous disposons aujourd'hui d'une enceinte spécialisée dans laquelle le remplissage, le rinçage et le séchage de pipettes étalonnées sont réalisés à distance. Cette enceinte se trouve dans une pièce thermostatée.

Des précautions sont prises pour éviter l'adsorption du traceur sur la verrerie en la «saturant» au préalable avec une solution concentrée de sel entraîneur.

Les deux échantillons d'activité sont enfin recueillis et transportés dans des bouteilles en verre, fermées avec une bille de verre scellée par une résine. Ces bouteilles, qui seront cassées dans le milieu de l'utilisation, ont été également «saturées» et contiennent les mêmes quantités de sel actif, d'entraîneur et d'eau.

4.2. Détermination de la distance de bon mélange, de la date et de la durée de prélèvement

Il est difficile de prévoir la distance minimale entre la section d'injection et la section de mesure nécessaire pour obtenir un «bon mélange». On peut admettre que cette distance est la même que celle qui est requise pour réaliser un bon mélange dans la méthode d'injection à débit constant.

Dans cette dernière méthode, en effet, la condition de bon mélange est que la concentration du traceur soit la même dans tous les points de la section de mesure : \( C = \text{constante} \). Or on démontre que l'hypothèse \( C = \text{constante} \) entraîne \( \int c \, dt = \text{constante} \). Inversement on montre que si la distance est telle que \( \int c \, dt = \text{constante} \), on a \( C = \text{constante} \) pour une injection continue.

Il est alors possible d'estimer théoriquement la distance que nous appellerons distance de bon mélange.

4.2.1. Distance de bon mélange

a) Mesures de canaux

On suppose que le traceur est injecté à l'aide d'une rampe placée au dessus de la surface de l'eau et perpendiculaire à la direction de l'écoulement afin de faciliter le mélange dans le sens transversal. La longueur de bon mélange \( L \) (telle que les concentrations ne diffèrent pas de plus de 1% pour une injection à débit constant) est donnée par [2] (formule du CREC):

\[
L = 9,5 nh
\]

où \( h \) = le tirant d'eau,
\( n \) = le rapport de la vitesse moyenne à la vitesse de frottement.
(Cette «vitesse» de frottement est définie par : \( U = \sqrt{\frac{g}{R}} \) avec
\( g \) = accélération de la pesanteur,
\( R \) = rayon hydraulique,
\( i \) = perte d'énergie par unité de poids et par unité de longueur du canal.)
On peut prendre pour \( n \):
\[
    n = 0.32 \, KR^\frac{1}{4}
\]
ou
- \( K \) = le coefficient de Strickler,
- \( R \) = le rayon hydraulique,
- Unités du système SI (mètre, seconde).

On a pu obtenir une bonne vérification expérimentale de cette formule théorique.

b) Mesures en rivières et fleuves

Dans ce cas, et pour une injection ponctuelle faite dans l'axe de l'écoulement, la formule établie par RIMMAR [3] permet d'estimer la distance minimale de bon mélange; cette formule ne semble pas encore avoir été systématiquement contrôlée, et il apparaît qu'elle ne devrait donner qu'un ordre de grandeur
\[
    L = 0.13 \, N \, \frac{b^2}{h}
\]
ou
- \( b \) = la largeur moyenne de la section mouillée dans la section de mesure (en mètres),
- \( h \) = la profondeur moyenne de l'eau dans cette section (en mètres),
- \( N \) = un coefficient égal à \( C \left( \frac{0.7 \, C + 6}{g} \right) \)
  - \( c \) = coefficient de Chézy (15 < \( c \) < 50)
  - \( g \) = accélération de la pesanteur (en m/s\(^2\))

c) Mesures en conduite forcée

La condition pour la distance de bon mélange, établie par Dumas, Lièvre et Bonnin de l'université de Grenoble, est
\[
    L > 75 \, d,
\]
avec \( d \) = diamètre de la conduite, et pour une injection dans l'axe de la conduite, ou quatre points d'injection dans une section.

4.2.2. Date du début du prélèvement

Si la connaissance de la date du début de comptage dans le cas d'une sonde immergée ne pose pas de problème, elle demeure une question importante dans le cas d'un prélèvement. En effet, afin de ne diluer qu'au minimum l'activité recueillie, le prélèvement devrait débuter à l'arrivée des premières molécules d'activité au point de prélèvement et cesser lorsque
passent les dernières molécules actives. Dans la pratique de larges tolérances sur ces dates doivent être prises.

a) Mesures en canal et fleuve

Nous ne connaissons pas encore de méthode satisfaisante permettant de déterminer la date de début de prélèvement dans le cas d'un fleuve. Pour les canaux, les caractéristiques hydrauliques de l'usine hydro-électrique permettent d'apprécier le débit, donc la vitesse moyenne. On sait par ailleurs que les premières molécules de traceur arrivent dans un certain temps de 10% inférieur à celui que permet de prédire la vitesse moyenne.

b) Mesures en conduite forçée

Le débit peut être estimé connaissant les conditions hydrauliques; on peut en déduire une vitesse moyenne de la vague d'activité et la date d'arrivée de l'activité selon la règle que nous venons d'indiquer.

c) Mesures de petite rivière

Il semble que la meilleure méthode soit d'effectuer un essai préliminaire avec de la fluorescéine. On peut en effet facilement apprécier à l'œil l'arrivée de la tête de la vague colorée à la zone de mesure.

Une méthode plus lourde, mais plus sûre pour un observateur non expérimenté, utilise l'information délivrée par une sonde nucléaire immergée en amont du point de prélèvement.

4.2.3. Temps de prélèvement

a) Mesures en canal, et en conduite forçée

On montre, pour une injection instantanée, que 99, 9% de la masse du traceur injecté passe en un temps $T$ égal à (formule du CREC)

$$T = 9,3 \sqrt{\frac{m h x}{n u^2}}$$

où $m =$ coefficient adimensionnel de la dispersion longitudinale égal à 7, 3 pour les canaux et 10, 1 pour les conduites,

$h =$ tirant d'eau,

$x =$ distance de la section de mesure à la section d'injection,

$\bar{u} =$ vitesse moyenne,

$n =$ rapport de la vitesse moyenne à la vitesse de frottement.

On peut prendre $0,32 K R^4 = n$

où $K =$ coefficient de Strickler,

$R =$ rayon hydraulique,

Unités du système SI (mètre, seconde)
b) Mesures de fleuve

Nous n'avons pas encore établi de raisonnement systématique qui conduirait à la détermination du temps de passage de la vague d'activité. Nous pensons à ce sujet que cet intervalle de temps, de même aussi que la distance de bon mélange, pourront être déterminés à la suite d'une analyse mathématique basée sur la connaissance expérimentale des coefficients de diffusion turbulente. La mesure effective du débit serait donc précédée d'une mesure des coefficients de diffusion turbulente ce qui alourdirait la méthode. Mais rappelons que la mesure d'un débit de fleuves demeurera toujours une opération assez peu fréquente et onéreuse, pour laquelle on peut donc admettre une certaine lourdeur.

c) Mesures de rivière

Dans la méthode que nous exploitons, le temps de prélèvement doit être fixé avant l'opération elle-même, car nous faisons usage d'orifices qualibrés qui déterminent le temps nécessaire pour remplir un volume donné. La méthode la plus simple semble être alors de procéder à une injection préliminaire de fluorescéine et d'apprécier le temps de passage de la vague colorée à la section de mesure. On a coutume alors de doubler ce temps pour évaluer le temps de passage effectif car l'œil apprécie mal les faibles concentrations de ce traceur coloré au début et surtout à la fin de la vague.

4.3. Injection du traceur

L'injection du traceur ne pose pas de problèmes difficiles pour les faibles débits. Les difficultés apparaissent pour les débits importants.

4.3.1. Cas des conduites forcées

Le dispositif que nous utilisons permet de casser l'ampoule de verre contenant le traceur dans la conduite elle-même. La figure 1 représente un schéma de ce dispositif qui est monté en général dans la chambre des vannes en amont de la conduite.

La pression est de l'ordre de quelques kilogrammes par centimètre carré. La canne étant enlevée, l'ampoule est introduite dans le conduit. Ce conduit est fermé à sa partie inférieure par une soupape qui est maintenue contre son siège par l'effet du contrepoids et de la pression. La canne est ensuite enfoncée brusquement, casse l'ampoule en fin de course, et réalise l'ouverture de la soupape.

4.3.2. Cas d'une rivière

Une canne de 4 m de longueur permet d'extraire l'ampoule de son enceinte de protection en plomb. Son extrémité est ensuite immergée et l'ampoule est brisée à l'aide d'un système du type arbalète et commandé à distance (fig. 2).
Figure 1
Dispositif d'injection en conduites forcées.

Figure 2
Détail de la canne d'injection en rivière.
4.3.3. Cas de fleuves et des canaux

Nous ne disposons pas encore de système d'injection dans le cas de forts débits. Une injection ponctuelle serait assurément la plus simple, mais il est souhaitable, en vue de diminuer la distance de bon mélange, d'injecter l'activité à la surface d'une section transversale. Le temps d'injection peut être théoriquement quelconque, mais l'on a avantage à le minimiser car il détermine en partie le temps de passage de la vague d'activité dans la section de mesure.

L'injection transversale pose un problème important de protection dans le cas d'un traceur émetteur de rayonnement gamma; avec le tritium, une injection à débit constant depuis une embarcation traversant la rivière d'une rive à l'autre pourrait être une solution.

4.4. Détection et comptage du traceur

4.4.1. Comptage avec une sonde immergée

La méthode la plus simple de comptage de l'activité est d'immerger la sonde de détection. Nous avons vu plus haut, au sujet de l'étalonnage des sondes, qu'il doit y avoir identité absolue entre la géométrie de la mesure et celle de l'étalonnage. On peut pour cela placer la sonde dans des conditions de «milieu infini», c'est-à-dire telles que toutes les parties du volume sensible de la sonde se trouvent dans l'eau. Les «rayons infinis» sont respectivement de 50, 70, 100 cm pour $^{131}$I, $^{82}$Br et $^{24}$Na; il faut donc, par exemple dans le cas du brome-82, que la sonde se trouve au moins à 70 cm du fond ou de la surface d'une rivière. Il est prudent d'augmenter ces valeurs de 20%.

Sur la figure 3 nous avons représenté trois dispositifs que nous avons essayés et qui ont été conçus pour faciliter la mise en place de la sonde en milieu infini. En a) figure une sonde suspendue à un flotteur, à une distance de celui-ci supérieure au rayon infini. Ce dispositif est d'une mise en œuvre très malaisée. En b) nous avons représenté un autre dispositif dans lequel la sonde est liée à deux flotteurs par un système rigide. Cette solution est viable, mais comme dans la précédente, on ne possède aucune indication sur la distance de la sonde au fond de la rivière. En c) figure un dispositif un peu plus lourd, mais qui, une fois immergé, donne l'assurance que le détecteur se trouve dans les conditions de volume infini; la sonde se trouve sur l'axe d'un treillis constitué de barreaux d'aluminium représentant un prisme droit dont la base est un triangle équilatéral. Son usage est à recommander pour les rivières à faible tirant d'eau, ce qui est le cas des torrents de montagne.

Nous donnerons cependant plus loin les raisons qui nous font préférer une méthode de prélèvement à cette méthode de la sonde immergée.

4.4.2. Echantillonnage continu

Certains expérimentateurs proposent de dériver une faible partie du courant de la rivière à travers une cuve dans laquelle plonge la sonde de détection. Ce procédé résoud le problème de la géométrie d'étalonnage
et libère des conditions de volume infini. Nous n'avons pas d'expérience de cette méthode mais lui préférons la suivante.

4.4.3. Comptage d'un échantillon moyen

a) Système de prélèvement

La figure 4 représente l'ensemble du système de prélèvement que nous utilisons pour les mesures en rivière. Ce système comprend une crépine et une pompe volumétrique entraînée par un moteur à courant continu alimenté par une batterie qui assure un débit légèrement supérieur au débit q désiré. Ce débit q est déterminé par une cuve à niveau constant. Cette cuve est munie d'un ajutage calibré, dont l'interchangeabilité permet de choisir la valeur de q. Une partie de l'eau entrant dans la cuve à niveau constant est renvoyée à la rivière.

Le débit de la pompe, voisin de 50 cm$^3$/s, varie approximativement de 1%/h; la hauteur du liquide dans la cuve à niveau constant est de 100 mm. On est donc assuré que le débit de prélèvement est constant à mieux que 0, 1%.
Figure 4
a) Schéma général du dispositif de prélèvement d'un échantillon moyen.
b) Détail de la cuve à niveau constant.

Le taux de renouvellement de la solution dans cette cuve est de $\frac{1}{3}$ s$^{-1}$; on peut donc négliger «l'effet de capacité» dans le volume de cette cuve, étant donné les temps de passage des vagues d'activité.

Dans le cas des conduites forcées la pompe est supprimée; en général, un prélèvement est fait au centre et sur la périphérie de la conduite et les cuves à niveau constant sont directement alimentées à travers deux vannes dont l'une sert au réglage du premier débit de prélèvement.

**b) Mesure du débit de prélèvement**

La cuve de prélèvement est munie de deux fenêtres opposées sur lesquelles est tracé un trait de repère définissant un volume de 18 l.

Le débit $q$ est déterminé en mesurant le temps nécessaire pour recueillir un volume donné, voisin de 18 l. On arrête le prélèvement avant que l'eau ait atteint le repère de 18 l, car les cuves ne sont en général pas horizontales et plusieurs prélèvements sont effectués simultanément par un seul opérateur.
Le temps de prélèvement est mesuré. Puis, après avoir établi l'horizontalité de la cuve à l'aide d'un système tripode à vis calantes et un niveau, on ajuste le niveau au trait de repère en versant un volume connu d'eau. On en déduit le volume effectivement prélevé.

Le volume optimum à prélever devrait avoir un « rayon infini » pour le rayonnement gamma utilisé. Nous avons adopté un volume de 18 l pour que les cuves soient faciles à transporter; de plus, le gain sur l'écart type relatif de la mesure en fonction de l'accroissement de volume est faible au delà de 18 l.

Par ailleurs il ne semble pas que l'on ait intérêt à réduire ce volume pour le prélèvement d'eau tritée dans le cas du jaugeage de grands canaux et fleuves; il est en effet certainement plus facile d'obtenir une bonne stabilité du débit de prélèvement si celui-ci n'est pas trop faible.

c) Cuves et sondes de comptage

La cuve qui a servi au prélèvement de l'échantillon est également utilisée pour son comptage ainsi que pour le comptage de la prise aliquote. La sonde de détection est immergée dans la cuve, maintenue dans une géométrie reproductible à l'aide d'un système approprié. Son cristal se trouve au milieu du volume d'eau, dont le diamètre est de 30 cm.

La plupart de nos essais ont été effectués avec une sonde constituée d'un compteur à haute efficacité LCT du type 13G14 dont la cathode à une surface de 180 cm$^2$.

Nous utilisons de plus en plus une sonde à scintillations avec un cristal en NaI (TI) de 1" X 1 1/2 dont la stabilité est très satisfaisante à condition de prendre des précautions pour la maintenir à température constante. Le seuil de discrimination est voisin de 30 keV.

Les caractéristiques de ces sondes avec les cuves de 18 l et pour $^{82}$Br sont:

| Sonde 13G14 | bruit de fond : 725 cpm |
|            | sensibilité : 150 cpm/μc.m$^3$ |
| Sonde à scintillations | bruit de fond : 4620 cpm |
|                | sensibilité : 1160 cpm/μc.m$^3$ |

Des essais nous ont montré qu'un facteur 10 peut être gagné sur la sensibilité de la sonde à scintillation en concentrant l'activité contenue dans les 18 l d'eau sur une résine que l'on dispose contre le cristal pour la mesure.

d) Activité à injecter

L'activité à injecter est fonction du temps de passage T de la vague d'activité, du débit Q qu'il faut évaluer approximativement, et de l'activité a, qu'il est nécessaire de prélever pour obtenir la précision statistique désirée.

Pour un temps de comptage de 30 min de l'échantillon, de 15 min pour le bruit de fond, et un écart type relatif de 0,2%, cette activité est $a = 0,23 \mu c$ de $^{82}$Br (13 μc/m$^3$) avec notre sonde à scintillation.
L'activité à injecter est

\[ A = a \frac{Q}{q} \], q étant déterminé par T.

L'évaluation de T n'est pas très difficile dans le cas des petites rivières et des conduites forcées.

La relation donnée plus haut pour les grands canaux donne des résultats satisfaisants, mais de sérieuses difficultés demeurent pour les fleuves.

Ces considérations et les essais préliminaires à la fluorescéine nous ont conduit à utiliser des activités de l'ordre de quelques dizaines de milli-curies par mètre cube par seconde dans les jaugeages de torrents alpestres dont les débits étaient inférieurs à 10 m³/s.

4.5. Avantages de la méthode de l'échantillon moyen

La méthode d'intégration que nous venons d'exposer présente sur les deux autres procédés classiques d'intégration des avantages sur certains points importants que nous rappelons ou examinons ci-dessous :

1° On évite avec cette méthode de placer une sonde en «milieu infini» dans le courant à mesurer, opération difficile ou impossible dans les rivières à faibles tirants d'eau.

2° Elle rend possible la prise d'un échantillon moyen au centre et sur les bords de l'écoulement afin de vérifier que la condition de bon mélange est satisfaite. Il ne serait pas possible d'immerger une sonde en milieu infini près des bords d'une rivière.

3° Dans le comptage d'un échantillon moyen on peut se fixer a priori l'erreur statistique.

4° Le comptage d'un échantillon moyen permet éventuellement d'effectuer des corrections de perte de comptage. Ces corrections sont possibles avec une sonde immergée au prix d'un enregistrement des variations de la fréquence de comptage.

5° L'activité à injecter est assez critique avec une sonde immergée à compteurs G-M comme le montrent les courbes de la figure 5 qui représentent, pour différents types de détecteurs, les variations de l'erreur statistique et de l'erreur due aux pertes de comptage.

Les sondes dont les données ont constitué la base expérimentale de ce calcul avaient les caractéristiques suivantes :

Sonde (S₁) : 2 compteurs G-M «13G14» à haute efficacité, \( \theta = 200 \mu s \),
Sonde (S₂) : 6 compteurs à halogènes « 3G3 », \( \theta = 120 \mu s \),
Sonde (S₃) : 1 crystal Nal (Tl) \( 1" \times 1" \ 1/2 \), \( \theta = 5 \mu s \).

Les surfaces des cathodes des compteurs G-M étaient par compteur :

13G14 : 180 cm², \hspace{1cm} 3G3 : 105 cm²,

et les rendements relatifs de ces sondes, en milieu infini, avec \(^{82}\text{Br}):

\[ \frac{\varepsilon (S₂)}{\varepsilon (S₁)} = 0,81 \], \hspace{0.5cm} \frac{\varepsilon (S₃)}{\varepsilon (S₁)} = 2,35 \]
En prenant la sonde (S₁) comme référence, nous avons calculé l'erreur due aux fluctuations statistiques et aux pertes de comptage pour les trois sondes envisagées. Nous avons pour cela admis que le temps de passage de la vague d'activité était de 15 min et que les pertes de comptage étaient de 5% au maximum de cette vague pour (S₁). Nous avons représenté en traits discontinus les fonctions relatives à des sondes qui comprendraient six compteurs 13G14, douze compteurs 3G3, et une sonde à scintillation dont l'électronique associée présenterait un temps mort de 0,5 µs.

6° Il est avantageux d'effectuer le comptage de l'échantillon et de l'activité de référence à des dates voisines étant donné les changements de gain de l'ensemble électronique, les corrections de décroissance d'activité, et éventuellement la décroissance d'activités parasites.

7° La méthode de prélèvement autorise l'utilisation d'une sonde à scintillation.

8° Cette méthode permet de concentrer l'activité afin d'accroître l'efficacité de comptage. C'est de plus la seule qui soit utilisable avec le tritium.

9° Les points 3°, 7°, 8° conduisent à une réduction au minimum de l'activité injectée.
5. QUESTION DE SÉCURITÉ

Un règlement régit en France l'application des radioéléments dans le domaine de l'hydrologie. Sont autorisées les applications de traceurs de période inférieure à huit jours à condition de montrer qu'en aucun point de l'écoulement où l'on puisse capter de l'eau pour la boisson ou l'arrosage, la concentration du traceur ne dépasse la concentration maximale admissible.

Ce règlement qui pourrait paraître exagéré par nous semble l'expression de la sagesse. Son premier mérite est celui d'exister; il est possible qu'après un examen approfondi de cette question, il soit quelque peu modifié dans le sens d'une plus grande clémence pour les hydrologistes.

Une autorisation soumise à un examen spécial est nécessaire pour les traceurs dont la période excède huit jours.

Cette clause, qui ne vise pas à éliminer des traceurs comme le tritium, met l'accent sur l'imperatif d'un examen strict des conséquences de l'introduction dans un milieu naturel d'éléments à longues périodes, tant du point de vue de la sécurité que de la contamination d'expériences ultérieures.

6. RÉSULTATS EXPÉRIMENTAUX

Nous avons réalisé sept campagnes de mesures de débits de rivières et trois campagnes de mesures de débits en conduites forcées; nous ne donnerons les résultats que de deux des dernières campagnes.

Le bromé-82 a été adopté pour ces essais car ce traceur à la réputation d'être peu adsorbé sur les terrains comme l'ont montré certaines études sur la sélection de traceurs pour l'hydrologie souterraine. De plus le bromé-82 a l'une des concentrations maximales admissibles les plus élevées.

Nous pouvons distinguer deux périodes dans nos essais. Dans la première, la prise de partie aliquote du traceur se faisait sur le terrain, dans la seconde, nous utilisions au laboratoire une cellule spécialisée pour cette opération. Par une ironie du sort, les débits obtenus après utilisation de cette cellule sont incomparablement plus mauvais que ceux de la première période. Nos ennuis venaient des produits de décomposition de la molécule BrNH₄ au cours de l'irradiation; ces produits contaminaient les prises aliquotes.

Pour les mesures de débits en conduites forcées d'usines hydro-électriques, les valeurs de référence étaient fournies par des moulinets, tandis qu'en rivières nous utilisons les résultats de mesures de dilution de bichromate de sodium. La précision de cette dernière méthode est de l'ordre de 2 à 3%.

Les distances entre les points d'injection et de mesures dans le cas des rivières de débit inférieur à 10 m³/s étaient comprises entre 200 et 600 m, et les temps de prélèvement ne dépassaient pas 30 min.

6.1. Mesures sur une conduite expérimentale – Chatou, 18-23 Juin 1962

Le tableau I résume les mesures effectuées sur une conduite expérimentale de 30 cm de diamètre et dont le débit, déterminé à l'aide d'une bâche de pesée, était connu avec une précision de 0,2%.
RÉSULTATS DES ESSAIS EFFECTUÉS A CHATOU DU 18 AU 23 JUIN 1962 SUR UNE CONDUITE D'EAU EXPÉRIMENTALE

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<th>Débit vrai (l/s)</th>
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<th>$\epsilon_2$ (%)</th>
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</table>
Cinq sections de prélèvements étaient aménagées dans la conduite, comportant chacune deux tubes, l'un prélevant au centre de la section, l'autre contre la paroi. Les distances de ces sections à la section d'injection étaient de 16, 22, 28, 35 et 43 m pour les sections 1, 2, 3, 4 et 5 respectivement.

A chaque injection quatre prélèvements étaient réalisés avec un débit de 0,04 l/s. Les temps de prélèvements variaient de 120 à 300 s.

Pour les injections 1 à 6, le débit de trop plein des bacs à niveau constant étaient trop faibles et a pu même s'annuler. Les prélèvements des mesures 7 et 8 ont été faits sans cuves à niveau constant.

Les valeurs $\epsilon_1$, $\epsilon_2$, $\epsilon_3$, portées dans le tableau, sont définies comme suit:

$\epsilon_1 = 100 \left( \frac{\text{débit mesuré} - \text{débit vrai}}{\text{débit vrai}} \right)$ pour chaque mesure,

$\epsilon_2 = \frac{\sum \epsilon_i}{4}$ pour chaque injection,

$\epsilon_3$ = écarts relatifs entre les deux débits mesurés dans une section.

Cette valeur représente les variations de l'expression $\int f_c \, dt$.

On peut attribuer à une prise défectueuse d'aliquote les divergences représentées par $\epsilon_2$, pour les injections 2, 3, 5, 7 et 11. Les divergences sur $\epsilon_3$, c'est-à-dire sur $\int f_c \, dt$ aux injections 1, 3, 5, 6, 8 peuvent être attribuées aux variations de débit de prélèvement.

Il est important de remarquer cependant que:
- sur 44 valeurs de $\epsilon_1$, 16 sont inférieures à 1%,
- sur 11 valeurs de $\epsilon_2$, 5 sont inférieures à 1%,
- sur 22 valeurs de $\epsilon_3$, 13 sont inférieures à 1%.

On peut espérer à la suite de ces essais et de jaugeages que nous avons effectués ou de conduites forcées, qu'il sera possible d'obtenir pour ce type de mesure une précision voisine, sinon meilleure, de 1% par la méthode d'intégration radioactive.


Au cours des essais précédents en rivière, on a pu mettre en évidence une divergence au plus de 3 à 4% entre nos résultats et ceux de la méthode de dilution de bichromate.

Le tableau II donne les résultats des essais effectués à Roselend en septembre 1962 au cours desquels cinq injections ont été faites; quatre prélèvements dans une même section de la rivière correspondaient à chaque injection.

Les débits mesurés sont erronés de 20% environ, car les ampoules des prises aliquotes étaient contaminées.

Un examen de la dispersion entre les quatre résultats d'une même mesure apporte une excellente vérification de la relation $\int f_c \, dt = \text{constante}$.

Les valeurs de la dernière colonne qui représentent les écarts de chaque mesure par rapport à la valeur moyenne excèdent rarement 1% excepté dans les cas suivants :

a) Injection n° 1 – prélèvements 1 et 4: Les écarts de +2, 8% et -4% montrent que la distance d'injection est trop faible. Cette distance, après
RÉSULTATS DES ESSAIS DE MESURE DE DÉBITS DE RIVIÈRE EFFECTUÉS À ROSELEND DU 26 AU 28 SEPTEMBRE 1962

<table>
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<th>Injection</th>
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<th>Prélèvement</th>
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<th>Qm - Qi (m³/s)</th>
<th>100(Qm - Qi) Qm (%)</th>
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avoir été portée de 300 à 400 m, s'est montrée suffisante pour donner un bon mélange.

b) Injection n° 3 - prélèvement 4 : Nous n'avons pas trouvé de causes à cet écart.

c) Injection n° 5 - prélèvement 2 : La valeur 2,9% peut s'expliquer par une perte d'activité au prélèvement. En effet ce dernier a été réalisé dans un courant secondaire à faible renouvellement. Il est très probable que la valeur d'activité en ce point a été plus longue que le temps de prélèvement.

Nous pouvons dégager de cet essai, et de nos expériences précédentes, une première conclusion. Etant donné, d'une part, la précision de la méthode de référence et par ailleurs, l'amélioration prévue du procédé de prise aliquote, on peut situer sans être optimiste entre 2 et 4% la précision des jaugeages de torrents alpestres par la méthode d'intégration radioactive.

7. CONCLUSION

Nous avons montré qu'un pas important a été fait, à l'aide de la méthode d'intégration radioactive, vers les mesures de débits de grands canaux et fleuves pour lesquelles le tritium semble le traceur le plus favorisé. Si
MESURES DE DÉBITS PAR LA MÉTHODE D'INTÉGRATION

pour la mesure des débits en conduites forcées et de rivières d'autres méthodes sont en compétition, complémentaires ou préférables, on dispose maintenant d'un nouveau procédé qui ne demande pour être bien assis qu'un complément d'étude sur le phénomène d'adsorption du traceur et le verdict de centaines d'essais.

REMERCIEMENTS

Que Messieurs B. Gaillard et H. Santos-Cottin trouvent ici l'expression de nos remerciements pour l'aide très efficace qu'ils ont apportée à cette étude.

RÉFÉRENCE


DISCUSSION

D. B. SMITH: I think that the authors have developed a useful instrument for groundwater measurement. As regards the value of ε, the calibration factor; will this not be critically dependent both on your instrument and on the nature of the strata in the immediate vicinity of the tube? Since these strata will be disturbed during insertion of the tube or production of the borehole, will the accuracy of the method not be limited rather extensively by such factors?

J. GUIZERIX: It can, of course, be assumed that the value of ε is dependent on conditions in the immediate surroundings, especially if the tube is inserted into the terrain by force. The technology of inserting the tube, including the effects of the soil in the vicinity of the tube, will be one of the main points of our study. For example, we plan to use the electric analogue method to examine the effect of the crumbling of the terrain close to the slits and also the effect of having some filtering material around the piezometric tube. It may be that our problem would be solved by the regular use of a filter around the tube. This would have the added effect of "adapting" transmissivities - in other words adapting the resistance, or the permeability of the medium, to that of the tube.

R. W. NELSON: I should first like to comment on Mr. Smith's question. About 15 years ago, Kirkham, Frevert and Luthin published some theoretical and experimental studies in the Proceedings of the Soil Science Society of America, in which they used three-dimensional conducting analogues and which indicated that there were only very minor disturbances or errors (probably of the order of 2 - 5%) immediately outside the piezometer. I therefore think that Mr. Guizerix's experiments are unlikely to be affected by the possible errors to which Mr. Smith refers.

In connection with Fig. 1 of the paper presented by Mr. Guizerix, showing the flow pattern into the piezometer, could he tell us what is the difference
in potential (say, in centimetres of water) between the point OP, at the centre of the piezometer tube, and a point some 10 piezometer diameters out in the soil, in the plane of symmetry immediately above that point? In other words, what is the difference between the potential in the piezometer and that in the natural flow system? Would the difference be of the order of 0.5 cm H2O, or something like 3 - 4 cm H2O?

J. GUIZERIX: Thank you very much for your information. The differences in potential which come under consideration vary over a wide range. Although I cannot give a quantitative reply to your question, I can say that in our laboratory experiments, these differences were of the order of 0.5 cm H2O.

R. W. NELSON: Our experience at Hanford has been that reproducibility was difficult when differences in head amounted to only 0.5 cm, and that was the figure usually obtained. In other words, the small difference in potential makes it difficult to assess the significance of the velocities.

J. F. HONSTEAD: I would like to emphasize, in this connection, that it is not always appropriate to judge the success of an experiment by the exponential nature of the results obtained. A few years ago, we had some experience with the application of this technique, although on a much cruder basis than that described in the paper. We proceeded on the assumption that if our results gave a nice linear plot on semi-log paper, it was a measure of the soundness of our experimental technique. It turned out, however, that this was not an appropriate procedure, because it was found that other processes could produce exponential decreases in tracer concentration which, of course, were interpreted as perfectly valid velocities. In our case, for example, we were studying reactions between injected tracer material and tube walls, the results of which sometimes showed an exponential decrease in tracer concentration. In one case, we were embarrassed to find that we were reporting a significant velocity in a well which had never been perforated at all.

J. GUIZERIX: I agree with you that the obtaining of an exponential response is not proof of the validity of a measurement. To make an over-hasty generalization, we know that in nature, if a law is not linear it is exponential. For example, such difficulties are met in measuring the rate of air renewal in rooms by means of radioactive gaseous tracers.

Y. HARPAZ: You described the measures which you took to prevent vertical flow within the casing tube. Have you done anything to prevent vertical flow outside the tube, when the apparatus is being used to measure natural horizontal flow? The latter flow could be quite considerable, although admittedly not so pronounced as that inside the tube.

J. GUIZERIX: You may recall that we made the assumption that the vertical component of the rate of flow was zero. However, I think that the measurement would still be valid with small vertical potential gradients.

D. B. SMITH: There will be increased danger of extensive vertical flow if the observation tube is surrounded by porous packing, as has been suggested in this discussion. I think that you will have to cut it into sections to reduce the vertical component.

J. GUIZERIX: This may be the solution which we shall use. In any event, for the time being, I am much more worried about other factors which might invalidate the method than this particular matter of vertical velocity.
I believe that in this case, slits in the tube would be preferable to circular holes. Before using this method, it will often be necessary to measure the rates of vertical flow in the piezometric tube, using, for example, a device similar to that described in the paper by Mr. Neumaier ("New experiences with the use of radioactive isotopes in hydrology", these Proceedings).
NEW EXPERIENCES WITH THE USE OF RADIOACTIVE ISOTOPES IN HYDROLOGY

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Abstract — Résumé — Аннотация — Resumen

NEW EXPERIENCES WITH THE USE OF RADIOACTIVE ISOTOPES IN HYDROLOGY. In groundwater science the filtration rate, i.e. the flow velocity of the groundwater across the filter tube, is of importance; it can be measured directly in the filter tube. The water in the filter tube is mixed with a radioactive tracer and the initial concentration then determined at various levels of the hydrological profile. Since non-treated water subsequently enters the filter tube, the tracer will undergo a deconcentration, which is a function of the filtration rate. This filtration rate directly furnished the yield of the groundwater current in its undisturbed state. Applying the Law of Darcy, we can calculate the permeability coefficient of the soil from the filtration rate.

So far, the filtration rate can be measured only in a horizontal groundwater current. In deep filter wells and observation gauges vertical currents may occur as a result of a hydraulic short-circuit. They are fed at certain points of the profile by groundwater layers of higher pressure and flow off at another point into low-strain groundwater layers. The recording of these vertical currents, the rate of the vertical flow in the filter tube as well as the location of the affluents and effluents is of prime importance to hydrological experts. The determination of these values is possible under certain conditions.

The practical application of these methods is described. In particular, the author discusses how radioactive tracers can be used to determine the course and extent of seepage flow under, and downstream of, storage dams.

NOUVELLES EXPERIENCES CONCERNANT L'EMPLOI DES RADIOISOTOPE EN HYDROLOGIE. En hydrogéologie, il importe de connaître la vitesse de filtration, c'est-à-dire la vitesse avec laquelle l'eau souterraine traverse un tube filtrant; on peut la mesurer directement dans le tube filtrant. On mêle un radioindicateur à l'eau du tube filtrant et on détermine la concentration initiale à diverses cotes du profil hydrologique. Lorsque de l'eau non traitée pénètre ensuite dans le tube filtrant, la concentration du radioindicateur diminue en fonction de la vitesse de filtration. De cette vitesse de filtration, on déduit directement la vitesse d'écoulement de l'eau souterraine dans les conditions normales. En appliquant la loi de Darcy, on peut calculer le coefficient de perméabilité du sol à partir de la vitesse de filtration.

Jusqu'ici, on ne peut mesurer la vitesse de filtration que dans un courant horizontal d'eau souterraine. Dans des puits filtrants profonds et dans des jauges d'observation, on peut se produire des courants verticaux à la suite d'un «court-circuit» hydraulique. Ils sont alimentés en certains points du profil par des nappes d'eau souterraine à forte pression et se déversent plus haut dans des nappes d'eau souterraine à faible pression. Pour les hydrologistes, il est extrêmement important d'enregistrer ces courants verticaux, de noter le débit dans le tube filtrant et de déterminer l'emplacement des affluents et des effluents; cela est possible dans certains cas.

Les auteurs décrivent les applications pratiques de ces méthodes. En particulier, ils indiquent comment on peut utiliser des radioindicateurs pour déterminer le cheminement et le débit de suintements sous les barrages de retenue et en aval de ces barrages.

НОВЫЕ ОПЫТЫ ПРИМЕНЕНИЯ РАДИОАКТИВНЫХ ИЗОТОПОВ В ГИДРОЛОГИИ. В науке о грунтовых водах важное значение имеет скорость фильтрации, т.е. скорость движения грунтовых вод через фильтрующую трубку; ее можно измерить непосредственно в фильтрующей трубке. Вода в фильтрующей трубке смешивается с радиоактивным индикатором и затем определяет первоначальную концентрацию на разных уровнях гидрологического профиля. Поскольку необработанная вода затем поступает в фильтрующую трубку, концентрация радиоактивного индикатора уменьшается, что является прямой функцией скорости
филтрации. Эта скорость дает возможность непосредственно определить выход потока грунтовых вод в его спокойном состоянии. Применяя закон Дарси, можно по скорости филтрации определить коэффициент водопроницаемости грунта.

До сих пор скорость филтрации можно было измерить только в горизонтальном потоке грунтовых вод. В глубоких филтрающих колодах и контрольно-измерительных приборах вертикальные потоки могут появляться вследствие гидрологического короткого замыкания. В определенных точках профиля в них вводится грунтовая вода из слоев высокого давления, а в другой точке они вытекают в слои грунтовой воды низкого давления. Измерение этих вертикальных потоков, скорости вертикального потока в филтрающей трубке, а также определение местонахождения притоков и истоков имеет особенное важное значение для специалистов гидрологов. Определение этих величин возможно при определенных условиях.

В докладе рассматривается применение этих методов на практике. В частности автор обсуждает вопрос использования радиоактивных индикаторов для определения направления и размера потока филтрающейся воды под водоудерживающими плотинами и вниз от них по течению.

NUEVAS EXPERIENCIAS EN LA APLICACIÓN DE ISOTOPOS RADIACTIVOS EN HIDROLOGIA. El índice de filtración, esto es, la velocidad de flujo del agua subterránea a través de un tubo filtrante, es muy importante para estudiar las capas freáticas. Se puede medir directamente en el tubo filtrante. El agua presente en el tubo se mezcla con un indicador radiactivo y la concentración inicial se determina a diversas profundidades del perfil hidrológico. Como después de añadir el indicador sigue penetrando en el tubo filtrante agua no trata da, el indicador experimenta una dilución que es función del índice de filtración. Este índice proporciona directamente el caudal de agua subterránea en régimen no perturbado, y a partir de él, aplicando la ley de Darcy, se puede calcular el coeficiente de permeabilidad del suelo.

Hasta ahora, sólo se podía medir el índice de filtración en una corriente horizontal de agua subterránea. Sin embargo, en pozos filtrantes profundos y en pozos de observación se pueden producir corrientes verticales debidas a cortocircuitos hidráulicos. Comienzan en determinados puntos del perfil freático, de presión elevada, y fluyen hacia otras capas de agua subterránea en que reina una presión menor. Para los hidrólogos, es muy importante registrar estas corrientes verticales, el caudal del flujo vertical en el tubo filtrante y el lugar donde se encuentran los puntos de entrada y de salida. Hoy, es posible, dadas ciertas condiciones, determinar los valores respectivos.

Los autores describen la aplicación práctica de estos métodos y, en especial, el empleo de los indicadores radiactivos para localizar y determinar el caudal de las filtraciones en el emplazamiento de los embalses y aguas abajo de los mismos.

1. METHODS

1.1. Measurements of filtration velocity in the case of horizontal groundwater flow

The filtration velocity \( v_f \) of a groundwater stream is defined as the quotient of the flow rate \( Q \) and the flow cross-section \( F \) of the aquifer:

\[
\nu_f = \frac{Q}{F}, \text{ in m/d.}
\]

To measure the filtration velocity \( v_f \), the groundwater flowing through a filter gauge tube is marked with a radioactive tracer. Fresh, non-marked water of the groundwater flow enters the filter tube; the initial concentration \( C_0 \) of the indicator suffers a decrease, which can be verified directly in the borehole with the aid of a radiation counter, or by sampling and subsequent measuring:

\[
C = C_0 \exp (-\nu_f F t / V)
\]
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Formula and diagram for the determination of the filter velocity $v_f$

$$C = C_0 \exp\left(-\frac{v_f F t}{V}\right)$$

Fig. 1

$C = \text{tracer concentration at the time, } t$
$C_0 = \text{initial tracer concentration}$
$V = \text{volume of water where the dilution takes place}$
$F = \text{cross-section of } V$

(V = volume of filter gauge tube where the dilution takes place). By plotting the logarithm of the relative decrease of concentration of the radioactive tracer as a function of time a straight line results, the slope of which is proportional to the filtration velocity $v_f$. The validity of the equation has been proved by laboratory tests. (See Fig. 1.)

Advantages of the method are the following:

(a) Determination of the groundwater yield without having to ascertain the effective pore content and the groundwater flow velocity.
(b) Measurement of the filtration velocity without having to apply the only conditionally valid Law of Darcy. Conversely, the Law of Darcy holding true, the permeability coefficient $K$ of any stratum of the aquifer can be determined.
(c) The measurement is performed in a single borehole; downstream gauges or other observation tubes are not needed.
(d) The measurement furnishes the groundwater yield for any stratum in the vertical profile of the aquifer.
(e) A wide measuring range (filtration velocity values were recorded between some cm/d and several tens of m/d).

Difficulties and limitations of the method are the following:

(a) The groundwater yield is covered only in the vertical profile of a single boring. To determine the yield within a large area, a number of measuring boreholes are necessary.
(b) The method is not applicable whenever there are vertical movements of the groundwater, besides the normal horizontal flow, owing, for instance, to connections between confined groundwater stories (see section 1.2).
1.2 Measurement of the vertical flow rate as well as of inflow and outflow to and from filter gauge tubes

A known amount of activity is introduced by inoculation at a certain depth of a filter gauge tube, detection instruments (counter tubes, scintillation counters) being suspended in the filter gauge tube above and below the point where the tracer is added (Fig. 2). The registration of the passage of the radioactively marked water with the mentioned arrangement furnishes the following measured quantities:

(a) The time the marked water takes to travel from one point in the borehole where a measuring device is suspended to the next measuring point; this serves as basis for establishing the vertical velocity $v_v$ of the water flowing in the filter gauge tube.

(b) The pulse sum triggered by the passing tracer cloud at each measuring point.

From each of the two measured quantities, the flow rate $Q_v$ of the vertical flow can be derived:

(a) Proceeding from the continuity equation $Q_v = v_v q$, the vertical flow velocity $v_v$ and the cross-section $q$ of the filter gauge tube clearly define the flow rate $Q_v$. 
(b) The pulse sum $N$ likewise serves to calculate the flow rate $Q_v$:

$$Q_v = \frac{EA}{N}.$$ 

($E =$ detection sensitivity of the measuring arrangement  
$A =$ amount of tracer added)

In this case, however, the detection sensitivity of the measuring arrangement must be known, which, under circumstances, might involve difficulties.

From changes in the flow rate so determined along the vertical profile, it is possible to deduce an affluent from the aquifer into the filter tube or an effluent from the filter tube into the groundwater duct. Under certain conditions, a picture is thus provided of the yield of the individual groundwater strata as well as of the flow patterns within the aquifer.

It is possible, for example, to seal those zones of a groundwater duct struck by a deep boring, where inflowing water with a high hydrostatic pressure flows off through a vertical hydrological short-circuit into strata of lower pressure and thereby is lost to the catchment. This method also permits knowledge of the seepage flow and of the pattern of flow lines in the foundations of storage dams.

It is difficult at this stage to give an outline of the possible applications of the method discussed, and to offer an accurate comment on the results achieved. The experiences gained in field tests so far, which will be mentioned in connection with an example in section 3.2, nevertheless justify further work in this sector.

2. TECHNIQUES

2.1. Radioactive indicators

Up to now, we have used iodine-131 and bromine-82 exclusively.

2.2. Inoculating equipment

The radioactive tracers are introduced into the groundwater through a borehole laid out as a filter gauge well. Fig. 3(a) shows an injection instrument that is lowered into the boring on a cable provided with depth marks. An inserted glass ampulla contains the radioactive solution; a striker, triggered electromagnetically, smashes the ampulla. The apparatus has so far been used down to water depths of 300 m. Fig. 3(b) shows a newly developed inoculating instrument functioning as a remote-controlled injector that adds the tracer only to the groundwater in the borehole. This instrument allows repeated or continuous injections without its having to be removed from the boring.

2.3. Detecting equipment

We used the gamma counter FHZ 24 (Firm Frieseke & Hoepfner, Erlangen, Germany) and a scintillation counter (Firm Berthold, Wildbad,
Germany) with special watertight mountings. For the measurements described in section 1.2 involving vertical movement, up to four counter tubes were arranged at various depths in a 2-in filter gauge tube (see Fig. 2).

2.4. Dosage

Regarding the dosage of the radioactive tracer, a distinction must be made between the measurement of groundwater flow velocity between two boreholes and the measurement in a single boring. In the first case, no generally valid data can be given on the amount of tracer activity required; it depends, inter alia, on the flow velocity and the yield of the groundwater as well as on the grain petrography of the aquifer.

In respect to the second case, which involves measurements in a single borehole, Fig. 4 gives guide values for the detection sensitivities of $^{131}$I and $^{82}$Br as a function of the boring diameter. The values were measured in the laboratory using solid pipe and do not allow for the influence of indicator substance in the surrounding subsoil. From the diagram we can calculate, however, that, for example, about $7 \mu$C of iodine-131 per meter of tube length suffice to measure an initial pulse rate of 1000 impulses/min with a gamma counter tube of 1.4-cm diam. and 18.5-cm sensitive length.
3. PRACTICAL EXAMPLES

3.1. Measurement of filtration velocity in a filter gauge tube with horizontal flow

Figure 5 shows the deconcentration with time of the indicator at various levels of a filter gauge tube with a 2-in diam. The $v_f$-values listed have been derived from the slopes of the corresponding measurement curves. Based on the same series of measurements, the distribution of the pulse rates along the filter gauge tube at different times has been plotted in Fig. 6. It is evident that no vertical water movement in the filter gauge tube affected the initial concentration distribution; here the condition of a purely horizontal water passage through the filter tube has been satisfied.
Fig. 5

Example for measurement of the filter velocity of a horizontal groundwater flow
3.2. Measurement of vertical flow in a filter gauge tube

Figure 7 gives the measurement results in a filter gauge tube on the upstream side of a storage dam. The problem had been to investigate whether or not there existed favoured seepage paths in the curtain under the dam. A further objective was to determine whether the seepage pattern depended on the storage level. In addition to the example given, about 30 boreholes were tested on the same basis.

The passage curve of the vertical flow represented at the left has been directly plotted from the measured vertical velocities. The considerable difference between the water movements measured about a quarter of a year apart results presumably from the variation in storage level of 0.6 m.

The diagram at the right depicts the differentiated curve, thus the variation in water passage. The zones of inflow and outflow to and from the filter gauge tube are readily recognized in the flow observed in the tube; they clearly indicate the seepage pattern existing in the surrounding subsoil. We are trying at present to supplement and intensify the knowledge of the structure of an aquifer gained through indicator tests by employing density and moisture probes (based on the dispersion of gamma and neutron rays).
Fig. 7
Evaluation of the measurement of a vertical flow in a filter tube, caused by seepage flow in a dam

--- 19.6.61-normal storage level, 751.40
measuring distance everywhere, 2 m

--- 15.9.61-normal storage level, 750.85
measuring distance everywhere, 2 m
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DISCUSSION

P. C. LÉVÉQUE: What method did you use for measuring the density of the alluvial layers saturated with water?

F. NEUMAIER: The measurements are based on the scattering of gamma radiation and neutrons. Since this work is only in its initial stage, it is not discussed in the paper.

P. C. LÉVÉQUE: My reason for asking this question is that in France we have made similar measurements of density in boreholes, using back-scattered gamma rays. We found that there were some unforeseen complications connected with both the granulometry and the chemical composition of the materials used. I might point out that as between two materials of equal density but unequal granulometry, the results given by gamma probes or back-scattering methods are rarely the same and we have had some difficulties on this score.

F. NEUMAIER: This has been our experience, too. Moreover, it is not only grain size which is involved; another important factor is the petrography of the aquifer, on which subject we have carried out a number of laboratory tests.

P. C. LÉVÉQUE: I fully agree.

A. NIR: In section 1.2 of the paper, you mention the equation \( Q_v = EA/N \), which you use for determining vertical flow. However, if horizontal flow is present at the same time, there will be a loss of isotope during vertical passage, i.e. \( A \) will not be constant. This will affect the calculation of the vertical flow rate. Do you take this factor into account?

F. NEUMAIER: Yes, we do, at least in cases where we have only very small \( v_v \) values. Where \( v_v \) is high, the loss of isotope during vertical passage can be disregarded. We believe that in many cases \( A \) will be constant.

D. B. SMITH: In section 1.1 of the paper, you mention as an advantage of the method the fact that the groundwater yield is measured without knowledge of the pore content or groundwater flow velocity. Is it not the groundwater velocity which is really determined by this method?
F. NEUMAIER: I disagree. This is not groundwater velocity; it is the so-called filtration velocity, the velocity of the groundwater in the filter gauge tube.

D.B. SMITH: In other words, you are defining the groundwater yield as the amount of water coming into your borehole?

F. NEUMAIER: Yes.

G. KNUTSSON: Have you carried out tests to determine whether the type of filter tube has any bearing on the measurement of filtration velocity? For example, how many holes – and of what size – should there be in the tubes?

F. NEUMAIER: Yes, we have performed such tests and have found that the type of filter tube does not influence filtration velocity. However, we are planning to make measurements with filter tubes over 5 in in diameter and these may show an influence of the kind you mention.

R.W. NELSON: Could your equipment for measuring vertical velocities fit inside a 1.5-in diam. tube?

F. NEUMAIER: No, it could not. The diameters of our injection instruments are 3 cm and 2.5 cm, respectively.

J. GUIZERIX: In my opinion, the particular conditions of vertical velocity measurements in piezometric tubes by the methods which you propose, call for a few comments. It seems to me that these techniques will be especially important in situations where the conventional microcurrent meters can no longer be used, i.e. for velocities under a few centimeters per second. We know what problems are encountered in applying radioactive methods to rivers and channels, especially as far as obtaining a satisfactory mixture is concerned. This difficulty would be increased in the case of the piezometric measurements. Moreover, since the method would be applied to measurements of low velocity, diffusion would become an extremely troublesome phenomenon.

F. NEUMAIER: I share your opinion. I can inform you that, thus far, we have made laboratory measurements of flow velocities ranging from 0.1 to 12 m/d. In almost all cases these measurements were in good agreement with our theoretical curves. I realize, however, that we must carry this work still further.

R. W. NELSON: With regard to the comments of Mr. Guizerix it seems to me that mixing lengths are not likely to be a problem. On the basis of a quick mental calculation of the diffusion rate, one would certainly not expect diffusion to become a problem until a velocity of at least some 2 to 3 cm/h (48 to 72 cm/d) had been reached.

J. GUIZERIX: I think it can be assumed that if diffusion is not important, the length required for obtaining good mixing will be very high – and this is convenient for measurement.

F. NEUMAIER: We have made some tests of the mixing of the radioactive tracers in the filter gauge tube and have found, on the basis of a simple reading arrangement, that mixing does not influence the measurement of the flow velocity.

G.H. LEAN: In connection with the length required for the mixing of the radioactive water in laminar flow through a vertical tube, reference
should be made to the very thorough investigation of this problem by Bailey and Gogerty in the Proceedings of the Royal Society, 1962. If I remember correctly a distance of more than 20 diam. from the point of injection was necessary.

F. NEUMAIER: Thank you very much for this information.
A METHOD FOR THE ESTIMATION OF GROUNDWATER STRATIFICATION

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Abstract — Résumé — Аннотация — Resumen

A METHOD FOR THE ESTIMATION OF GROUNDWATER STRATIFICATION. Many efforts have been made to establish a method for the estimation of groundwater stratification, and yet no simple method has been established. The authors present a method of estimation involving comparison of two breakthrough curves of radioisotopes injected into ground.

This method requires at least two wells, i.e. one for the injection well and the other for the observation well. Before radiotracers are applied to these wells, groundwater velocity and its vertical distribution is determined by the point dilution method. A radioactive tracer is added in the well casing, and its decay by dilution of groundwater is observed. The groundwater velocity at a certain depth can be estimated from the rate of dilution.

Tritiated water and a radio-cation such as Sr\textsuperscript{90} are injected in the injection well, and their breakthrough to the other well is observed for individual radioisotopes. Logs of the wells, vertical distribution of groundwater velocities and tritiated water breakthrough gives the stratification of groundwater at and around the observation well. The relationship between arrival time of the peak of Sr\textsuperscript{90} breakthrough and tritiated water breakthrough is given as

$$t = \left(1 + \frac{1-f}{f} \rho k_d \right) T = K_f T,$$

where \(t\) is the arrival time of the Sr\textsuperscript{90} peak, \(T\) is the arrival time of the HTO peak, \(f\) is porosity of the formation, \(\rho\) is the density of the formation, \(k_d\) is the distribution coefficient and \(K_f\) is the time transformation factor. \(k_d\) may be estimated from the core samples of wells. By comparison of the ratio of these two arrival times of peak and \(K_f\) the stratification of groundwater in the vicinity of the two wells can be estimated. Additional observation wells may be used if necessary in order to obtain more precise estimation of groundwater stratification for a wider area.

MÉTHODE D’ÉVALUATION DE LA VITESSE DE DéPÔT DES NAPPEs D’EAU SOUTERRAINE. En dépit de nombreux efforts, il n’a pas encore été possible d’établir une méthode simple d’évaluation de la vitesse de dépôt des nappes d’eau souterraine. Les auteurs présentent une méthode qui repose sur la comparaison de deux courbes de pénétration de radioisotopes introduits dans le sol.


On introduit de l’eau tritiée et un cation radioactif \(\text{Sr}^{90}\), par exemple, dans le puits d’injection et on observe le cheminement de chaque radioisotope jusqu’à l’autre puits. Le sondage des puits, la répartition verticale des vitesses d’écoulement de l’eau souterraine et le cheminement de l’eau tritiée indiquent la vitesse de dépôt de l’eau souterraine au niveau du puits d’observation et aux alentours. Le rapport entre le moment où arrive la quantité maximum de \(\text{Sr}^{90}\), d’une part, et d’eau tritiée, d’autre part, est donné par la formule

$$t = \left(1 + \frac{1-f}{f} \rho k_d \right) T = K_f T.$$
оù: t est le moment où arrive la quantité maximum de $^{90}\text{Sr}$; T, le moment où arrive la quantité maximum de HTO; $f$, la porosité de la formation; $p$, la densité de la formation; $k_d$, le coefficient de répartition; $K_f$, le facteur de transformation dans le temps. On peut évaluer $k_d$ à l'aide d'échantillons prélevés à l'intérieur des puits. En comparant le rapport entre ces deux moments d'arrivée des quantités maxima et $K_f$, on peut évaluer la vitesse de dépôt de l'eau souterraine au voisinage des deux puits. On peut utiliser, le cas échéant, des puits d'observation supplémentaires pour obtenir une évaluation plus précise de la vitesse de dépôt de l'eau souterraine sur une plus grande surface.

МЕТОД ОЦЕНКИ СТРАТИФИКАЦИИ ГРУНТОВЫХ ВОД. Много усилий затрачено в целях разработки о-метода оценки стратификации грунтовых вод, и тем не менее нет еще ни одного простого метода. Авторы предлагают метод оценки, связанный с проведением сравнения двух кривых прохождения радиоизотопов, введенных в грунт.

При использовании этого метода необходимо иметь по крайней мере две скважины: одну - для введения радиоизотопов, другую - для наблюдения. Прежде чем использовать радиоиндикаторы в этих скважинах, о помощи метода точечного разбавления определяется скорость течения грунтовых вод и их вертикальное распределение. В отвод окажется радиоактивный индикатор, и наблюдается его распад в результате растворения в воде. Скорость течения грунтовых вод на определенной глубине может быть рассчитана по основанию данных о скорости растворения.

Во входную скважину вводится вода, насыщенная тритием, и радиоактивный катион, например $^{90}\text{Sr}$, и по отдельным радиоизотопам наблюдается их перемещение к другому скважине. Данные относительного размеров скважины, вертикального распределения скоростей течения грунтовых вод и прохождения воды, насыщенных тритиом, дают возможность определить стратификацию грунтовых вод на месте скважины наблюдения и вокруг неё. Связь между временем при поступлении $^{90}\text{Sr}$ и воды, насыщенной тритием, выражается формулой:

\[ t = (1 + \frac{1}{f} p k_d) T = K_f T, \]

где t - время при поступлении $^{90}\text{Sr}$, T - время при поступлении HTO, f - пористость формации, $p$ - плотность формации, $k_d$ - коэффициент распределения и $K_f$ - коэффициент изменения времени. $K_d$ может быть вычислен по образцам керна в скважинах. Путем проведения сравнения отношений этих двух моментов пок и коэффициентов $K_f$ может быть определена стратификация грунтовых вод в окрестности обеих скважин. В случае необходимости могут быть использованы дополнительные скважины для проведения более точного определения стратификации грунтовых вод на большей площади.

METODO DE CALCULO DE LA ESTRATIFICACION DE LAS AGUAS SUBTERRANEAS. Son muchos los trabajos realizados para elaborar un método de cálculo de la estratificación de las aguas subterráneas, sin que se haya logrado preparar un método sencillo. Los autores exponen un método de cálculo basado en la comparación de dos curvas de afloramiento de radioisótopos inyectados en el suelo.

Para este método se necesitan por lo menos dos pozos, uno de inyección y otro de observación. Antes de emplear los indicadores radioactivos en estos dos pozos, se determina la velocidad de las aguas subterráneas y su distribución vertical por dilución de una fuente puntiforme. Se introduce un indicador radioactivo en el pozo y se observa la disminución de su actividad por dilución en las aguas subterráneas. La velocidad de éstas, a una profundidad determinada, puede calcularse sobre la base del índice de dilución.

En el pozo de inyección se introducen agua tritiatada y un radioisótopo, tal como el $^{90}\text{Sr}$, y se observa el afloramiento de cada radioisótopo en el otro pozo. Los perfiles de los pozos, la distribución vertical de velocidades del agua subterránea y la afloración del agua tritiatada permiten deducir la estratificación de las aguas en el pozo de observación y en sus proximidades. La relación entre el tiempo que tarda en producirse el afloramiento máximo del $^{90}\text{Sr}$ y el afloramiento máximo del agua tritiatada se expresa por la fórmula:

\[ t = \left( 1 + \frac{1-f}{f} p k_d \right) T = K_f T. \]

En la que t es el tiempo que tarda en aparecer el máximo correspondiente al $^{90}\text{Sr}$, T es el tiempo que tarda en aparecer el máximo correspondiente al agua tritiatada, f la porosidad de la formación, p la densidad de la
formación, $k_d$ el coeficiente de distribución y $K_f$ el factor de transformación de tiempo; $k_d$ puede deducirse de muestras testigo extraídas de los máximos y $K_f$, puede calcularse la estratificación de las aguas subterráneas en las proximidades de los dos pozos. Si es preciso pueden emplearse más pozos de observación para calcular con mayor exactitud la estratificación de las aguas subterráneas en una zona más extensa.

INTRODUCTION

There are many difficulties in estimating the stratification of groundwater flow, although it is important to determine the nature and origin of the groundwater. Many complex factors, including hydraulic gradient, permeability of formation, and density and viscosity of fluid, which vary as a result of chemical component and temperature difference, govern the directions and velocities of groundwater. Besides, heterogeneity of the strata adds to the complexity of the groundwater flow. Therefore, combinations of hydrological investigation methods must be used to estimate the groundwater stratification. In our study, a dilution-velocity method is adopted for the determination of the vertical distribution of groundwater velocities at a well site, and laboratory experiments utilizing tritiated water and radio-cation tracer are applied for the identification of the mutual relations between stratified waters at each of the well sites.

THEORETICAL CONSIDERATIONS

There are many hydrological methods for the investigation of groundwater. They are observation of hydraulic gradient, measurements of transmissibility and permeability, core tests, in-well techniques and so forth. In order to determine vertical distribution of groundwater flow by in-well techniques, Raymond’s dilution-velocity method [1, 2] is useful. This was presented initially by RAYMOND [1] and improved theoretically by KRONE [3]. The velocity of the groundwater may be calculated by Krone’s equation from the dilution rate of tracers introduced into a well casing:

$$v = \pi d \ln(C_0/c)/8 t \phi B, \quad (1)$$

where

- $\bar{v}$ = interstitial average velocity
- $d$ = diameter of the well
- $C_0$ = initial concentration of tracer in the well casing
- $c$ = concentration of tracer in the cell at time $t$
- $\phi$ = porosity of formation
- $B$ = depth of the well casing.

This equation assumes complete mixing of tracer in the cell. To measure the vertical distribution of groundwater velocities, a part of the well casing is shielded and radioactive tracer is introduced. Samples are taken periodically while the water in the cell is mixed. As a tracer, tritiated water is most suitable because it follows water movement. This method gives only the flow velocity. The bulk of the groundwater flows follow the
hydraulic gradient to the maximum and this indicates flow direction. In fact, however, there are many exceptional cases.

Since the above-mentioned method gives only the vertical distribution of groundwater velocities at a well site, stratification of the groundwater flow for a wider area must be analysed on the basis of data obtained from many wells, taking geological formation into account. As a method of relating point stratification to the wider area, a comparison of the water velocity and radioactive tracer velocity may be used effectively.

When a radioactive tracer (ionic) is introduced into the formation, the variation of the liquid concentration \( c \) may be expressed by Eq. (2) [4].

\[
\frac{\partial c}{\partial t} = \left[ \frac{\partial}{\partial x} \left( D_x \frac{\partial c}{\partial x} \right) + \frac{\partial}{\partial y} \left( D_y \frac{\partial c}{\partial y} \right) + \frac{\partial}{\partial z} \left( D_z \frac{\partial c}{\partial z} \right) \right] + \left[ \frac{\partial}{\partial x} (v_x c) + \frac{\partial}{\partial y} (v_y c) + \frac{\partial}{\partial z} (v_z c) \right] - \frac{(1-f)}{f} \rho \frac{\partial q}{\partial t}, \tag{2}
\]

where \( x, y, \) and \( z \) are Cartesian co-ordinates, \( c \) and \( q \) are the radiotracer concentrations in the liquid and solid phases, respectively, \( \rho \) is the density, \( v_x, v_y, \) and \( v_z \) are interstitial average velocities, and \( D_x, D_y, \) and \( D_z \) are the dispersion coefficients along \( x, y, \) and \( z \) directions. The first group of terms on the right-hand side depicts dispersion, the second group the conventional transport, and the third adsorption of the solid phase.

When the concentration of radioactive tracer is chemically negligible compared to that of gross ion in the water, \( c \) and \( q \) have a linear relationship, as expressed by

\[
q = k_d c, \tag{3}
\]

where \( k_d \) is the distribution factor.

If there is an adsorption equilibrium between radioactive ion \( A \) and gross ion \( B \) in the water, such as

\[
bA^{+a} + B_d R \rightleftharpoons aB^{+b} + A_b R, \tag{4}
\]

\( k_d \) may be written as

\[
k_d = (K_{A-B})^{1/b} (q/c)^{a/b}, \tag{5}
\]

where \( a \) and \( b \) are valencies of ion \( A \) and \( B \) respectively, \( R \) denotes soil, \( K_{A-B} \) is the mass-action constant, and \( q_B \) can be substituted for ion-exchange capacity.

For example, when strontium-90 has an equilibrium relationship with calcium in the water, Eq. (4) gives

\[
Sr^{++} + CaR \rightleftharpoons Ca^{++} + SrR. \tag{6}
\]
In this case, the distribution factor may be expressed by

\[ k_d = (K_{Sr-Ca}) \frac{(q/c)}{C_{a}}. \tag{7} \]

Substituting Eq. (3) for Eq. (2) and applying time transformation, we find that

\[ T = (1 + ((1 - f)/f) \rho k_d) t, \tag{8} \]

and Eq. (2) is converted to

\[
\frac{\partial c}{\partial T} = \left[ \frac{\partial}{\partial x} \left( D_x \frac{\partial c}{\partial x} \right) + \frac{\partial}{\partial y} \left( D_y \frac{\partial c}{\partial y} \right) + \frac{\partial}{\partial z} \left( D_z \frac{\partial c}{\partial z} \right) \right] \\
+ \left[ \frac{\partial}{\partial x} \left( v_x \cdot c \right) + \frac{\partial}{\partial y} \left( v_y \cdot c \right) + \frac{\partial}{\partial z} \left( v_z \cdot c \right) \right]. \tag{9} \]

Equation (9) shows the movement of the water, because it includes only hydraulic dispersion and convectional transport terms. This shows that the water moves faster than radioactive tracer by a factor of \( K_f = 1 + [(1 - f)/f] \rho k_d \), which transforms \( t \) to \( T \) by Eq. (8).

Porosity \( f \), density \( \rho \) and distribution factor \( k_d \) may be obtained by testing core samples taken from each of the well logs. On the other hand, if tracers of tritiated water and a radio-cation, such as strontium-90, are introduced in one well and the arrival time or breakthrough of each of the two radiotracers is measured at an individual stratum, the \( K_f \) value of each stratum may be obtained from the ratio of arrival time of these radiotracers.

As the value of \( K_f \) is usually peculiar to a stratified layer, strata which bear the same \( K_f \) values, having been obtained either from the core sample at two well-sites or by the tracer test between the well sites, must pass the same stratified groundwater flow. For the determination of \( K_f \) values, the latter method would be better than the former, because a direct relationship could be revealed between corresponding strata at two well sites, while both methods could be used for mutual check. Even to the former method, however, we can apply the proposed procedure to determine the \( K_f \) value independently for each of the core samples. This can be obtained from a column test with the tracers, instead of conventional chemical determinations of ion-exchange capacity \( q_B \), mass-action constant \( \text{A}_{AB} \) and direct measurements of \( f \) and \( \rho \). At any rate, geological and mineralogical analyses of core samples must be referred to such a study.

**EXPERIMENTAL VERIFICATION OF THE THEORY**

In order to ascertain the validity of the theory, a laboratory test was performed using two kinds of sand layer. The model, 10 cm wide, 30 cm high and 50 cm long, as shown in Fig. 1, was made of polyethylene plates. Two kinds of sand of different size were packed in this model so as to form
two separate layers each 10 cm thick. The size distribution of the sand is shown in Fig. 2. The model had two chambers with paddles, each of which received effluent from each sand layer.

The dilution-velocity method was examined first. Tritiated water was introduced into each chamber, while tap water was passed through the model. The decay of tritiated water concentration was measured by taking samples, while the water in the chambers was mixed gently. The actual flow rate was measured simultaneously, and the tritiated water samples were counted.
TABLE I

COMPONENTS OF LIQUID SCINTILLATOR*

<table>
<thead>
<tr>
<th>Component</th>
<th>Mass</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dioxane</td>
<td>11 g</td>
</tr>
<tr>
<td>PPO</td>
<td>6 g</td>
</tr>
<tr>
<td>POPOP</td>
<td>0.3 g</td>
</tr>
<tr>
<td>Naphthalene</td>
<td>100 g</td>
</tr>
</tbody>
</table>

* 2 ml of sample water and 18 ml of the liquid scintillator were used.

by Packard Tri-Carb Liquid Scintillation Spectrometer Model-500 B. The components of the liquid scintillator are shown in Table I.

Figure 3 shows the decay of tritiated water concentration in the cell. The lower layer showed good linearity of decay on semi-log paper, while the upper layer did not. The calculated flow rate was 86.7 ml/min in the upper layer and 59.3 ml/min in the lower layer. The actual flow rate was kept at 135 ml/min throughout the experiment. Hence, the dilution-velocity method gave a total flow rate of 86.7 + 59.3 = 146.0 ml/min, which was about 8% higher than an actual record of our observation. From such a result, it is seen that the dilution-velocity method is applicable to the determination of vertical velocity distribution, with an acceptable accuracy.

Before applying radioactive tracers to the model, the time transformation factor, $K_f$, was determined for each of the layers. Since the upper layer of sand had a porosity of 42.6%, while the lower layer had a porosity of 41.3%, it was revealed that the latter was packed more tightly than the former. Densities of the sand were almost identical in both layers,
2.53 g/cm$^3$ for the upper and 2.55 g/cm$^3$ for the lower. The distribution coefficient $k_d$ was calculated by Eq.(7), using observed values of $K_{Sr-Ca}=1.2$, and $c_{Ca}=19.2$ ppm as CaCO$_3 = 9.6 \times 10^{-4}$ meq/ml. As stated in the above, $q_{Ca}$ can be substituted by ion-exchange capacity. The exchange capacities of the sand, which had been determined by the ammonium acetate method [5], were 8.2 meq/100 g for the upper sand layer and 6.4 meq/100 g for the lower. The time transformation factor, $K_f$, calculated from the above data was 350 for the upper layer and 290 for the lower.

From the time tap water was initially introduced in the model at a rate of 196.7 ml/min, tritiated water and strontium-90, in concentrations of $10^{-2} \mu$C/ml and $2.5 \times 10^{-3} \mu$C/ml, respectively, were dosed to the water continuously for 80 min. Effluent samples were taken periodically from each layer and prepared for radiological analysis. Other than the analysis of tritiated water as already described, 2 ml of sample waters were taken for the analysis of strontium-90, evaporated in stainless-steel planchets (2.5 cm in diameter and 0.6 cm in depth), then counted by an Aloka Gas-Flow Anti-coincidence Counter.

An observed breakthrough curve of tritiated water is drawn in Fig. 4, where the upper layer shows a 100% and the lower a 90% breakthrough at 70 min after beginning injection.

Strontium breakthrough is represented in Fig. 5. Although time intervals of strontium sampling were too long, the time of peak arrival was approximately 500 min for both layers. It was recognized, however, that the breakthrough concentration of the lower layer was higher than that of the upper.

The time transformation factor, $K_f$, of each layer can be calculated, as described below, from the ratio between the times of peak arrival of these radioactive tracers. As the injection period of strontium is quite short in comparison with its peak-arrival time, the injection may belong to "slug" dose. With the tritiated water, on the other hand, its breakthrough reached almost 100% at the end of its application; hence, the injection of tritiated water may belong to "continuous" dose.

Slug-dose breakthrough can be obtained from derivatives of continuous dose breakthrough. Thus, the peak-arrival time of tritiated water was com-

![Fig. 4](link)

**Fig. 4**

Tritiated water breakthrough curve
ESTIMATION OF GROUNDWATER STRATIFICATION

In Table II, the peak-arrival times of strontium-90 and tritiated water are tabulated, together with the time transformation factors, thus obtained for each layer.

In Table III, moreover, two groups of transformation factors, one from Table II and another obtained from the analysis of sand, were compared. It was observed that between the values of the factors in Table III there was a difference of 12.5% in the upper layer and practically no difference in the lower. In actual cases of various strata at well sites, the value of the time transformation factor varies more widely than in the above case. Such a variation facilitates an identification of corresponding stratified aquifers.

SUMMARY AND CONCLUSIONS

A method for the estimation of groundwater stratifications was proposed, by means of an observation discriminating moving velocities of tritiated

| TABLE II |
|------------------|------------------|------------------|
|                  | Arrival of Sr\(^{90}\) peak (min) | Arrival of HTO peak (min) | \(K_f\) |
| Upper layer      | 1000             | 25               | 400   |
| Lower layer      | 1000             | 34               | 294   |
water and another radiotracer in groundwater through earth formations between well sites, in combination with Raymond's dilution-velocity method which gives the vertical distribution of groundwater velocities at a well site. The new method reveals clearly mutual relationships between stratified waters at each of the well sites.

A model study using sand had been performed and results obtained were considered to support satisfactorily the proposed method. The dilution-velocity method gave the vertical distribution of groundwater velocities with an acceptable accuracy, while the radioactive tracer study showed that a stratified water at a well site can be connected to a wider area. For such an analysis, the theory of time transformation had been applied effectively to the basic differential equation, describing dispersion and chemical adsorption.

Since the verification of the theory was made only with a few series of model tests with sand layers, a further field study must be carried out to gain a more complete support to the theory as well as to develop the experimental techniques.

REFERENCES


DISCUSSION

A. AMBROGGI: Do you think that this laboratory experiment could be applied in the field at reasonable cost?

Y. INOUE: This would depend on the type of formation to be investigated. If the formation was very wide, the cost might be high. However, by comparison with the other hydrological methods used in the investigation of groundwater, I do not think that this method of estimation would be too expensive.
J. GUIZERIX: I would like to comment briefly on the formula put forward by RAYMOND in 1957 [1] in relation to the flow around a perfect piezometer. My reason for stating, in the presentation of my paper, that this is a classical question is that, even before 1945, it was one of the subjects dealt with in the general hydraulics course at the Grenoble école d'ingénieurs hydrauliciens. It can be shown that the figure formed by lines of flow and equipotentials in this case represents the superposition of two simple and well-known analytical systems: a parallel flow; and a force doublet due to a spring and a well merging at the centre of the circle.
THE USE OF TRITIUM IN TRACING KARST GROUNDWATER IN GREECE

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Abstract — Résumé — Аннотация — Resumen

THE USE OF TRITIUM IN TRACING KARST GROUNDWATER IN GREECE. Tritiated water has been used, to determine possible interconnections between two sinkholes on the high plateau of Tripolis and a number of springs near the coast of the Peloponnese. The existence of two interconnections has been demonstrated and the results have been applied to the determination of transit times, flow rates and groundwater storage.

UTILISATION DU TRITIUM POUR L’ÉTUDE DES EAUX SOUTERRAINES DANS UN SOL KARSTIQUE, EN GRECE. Les auteurs ont utilisé de l’eau tritiée pour rechercher s’il existait une communication entre deux entonnoirs du haut-plateau de Tripolis et plusieurs sources situées près de la côte du Péloponnèse. Ils ont prouvé l’existence de deux voies de communication et ont appliqué les résultats pour calculer la vitesse d’écoulement, le débit et la quantité d’eau souterraine accumulée.

ИСПОЛЬЗОВАНИЕ ТРИТИЯ ДЛЯ ПРОСЛЕЖИВАНИЯ КАРСТОВЫХ ГРУНТОВЫХ ВОД В ГРЕЦИИ. Меченая тритием вода использовалась для установления возможной взаимосвязи между двумя карстовыми воронками на плоскогорье Триподис и несколькими ключами у побережья Пелопоннеса. Существование этой взаимосвязи было доказано, и результаты были использованы для того, чтобы определить расход потока и запас грунтовых вод.

EMPLEO DEL TRITIO PARA ESTUDiar AGUAS SUBTERRANEAS EN TERRENOS KÁRSTICOS DE GRECIA. Los autores han empleado agua tritiada para determinar posibles conexiones entre dos dolinas en la meseta de Arcadia y una serie de manantiales próximos a la costa del Peloponeso. Se ha demostrado la existencia de dos interconexiones y los resultados han permitido determinar los tiempos de tránsito, el caudal, y la cantidad de agua almacenada en el subsuelo.

INTRODUCTION

The aim of the United Nations Special Fund Groundwater Project in Greece was to determine, test and demonstrate methods for the successful and efficient development of groundwater resources in limestone terrain. One of the areas under investigation is that between the high plateau of Tripolis and the plain and Gulf of Argos in the Peloponnese (Fig. 1). The alluvial plateau of Tripolis, which is surrounded by mountains, is at an altitude of about 650 m. The surface drainage leaves the plain through a number of sinkholes, of which those at Partheni and Nestani are the most important on the eastern side of the plain. Towards and particularly near the coast
Map of the area under investigation

© Sinkholes
● Springs
about 30 km away are a number of springs, some of which issue at the foot of the cliffs south of Kiveri at Ay. Ghorgios directly into the sea and also under the sea some distance from the shore (Anavalos).

For utilization of the groundwater in the limestone and possible interception of that water which is being lost to sea, information was required on the following points:
1. which sinkhole feeds which spring or group of springs;
2. what is the groundwater residence time between the sinkholes and the springs; and
3. how much groundwater is stored between the sinkholes and the springs.

It was suggested that radioisotopes could be used for such an investigation. In view of the rather long distances involved, tritium was chosen as a tracer rather than other radioisotopes which would be more prone to loss by adsorption. The sinkholes at Partheni and Nestani were investigated in two field experiments during the springs of 1961 and 1962 respectively.

FIRST FIELD EXPERIMENT

At midday on 23 March 1961 1000 c of tritiated water was added over a period of 1 h to the water flowing into the sinkhole at Partheni. The amount of tritium was sufficient to ensure detection even if it was diluted by ten times the annual run-off from the plain of Tripolis. The tritiated water was contained in 20 glass ampoules, each containing 50 c. Each ampoule was crushed under the surface of the water. The injection device consisted of a brass cylinder with a plunger connected to a long handle. By a sharp jerk given to the handle the plunger crushed the ampoule against the bottom of the cylinder, permitting the contents to be washed away by the stream water through a number of holes around the lower part of the cylinder.

On the same day 12.5 kg of fluorescein was added to the water flowing into the sinkhole at Nestani to ascertain if there was a rapid connection to Kefelari, which provides water for the town of Argos. Although it was realized that fluorescein is far from an ideal tracer, it was thought that, if a rapid connection did exist, some of the fluorescein would appear at Kefelari. This information would be useful for the planning of the second experiment.

Samples of water were taken from the springs at Binikovi, Kefelari, Myli, Kiveri and Anavalos. The sampling programme was commenced before the introduction of the tritium for the establishment of a sampling routine for the locally employed personnel and for the provision of samples from which the natural level of tritium could be determined. Samples were taken at close intervals shortly after the injection in case there was a rapid transport of the tracer.

Sampling at the submarine spring of Anavalos proved to be rather difficult. Although from its appearance the spring appeared to be large, samples of water taken in the centre of the upwelling water and halfway to the bottom were highly saline. Later a plastic tube was anchored in the mouth of the spring at a depth of 72 m and connected to a buoy on the sea surface from where samples of water issuing from the spring could be taken. Chemical
analysis of the water indicated, however, that almost no fresh water is coming from the spring.

SECOND FIELD EXPERIMENT

In the second part of the investigation 400 c of tritiated water was added at midday on 5 February 1962 to the water flowing into the sinkhole at Nestani. The same method of injection was used as in the first experiment. Sampling was carried out at the same springs and in addition at Akladokambos and a bore-hole at Skafidaki.

RESULTS

Fluorescein analyses were made at the Institute for Geology and Subsurface Research in Athens and did not indicate the presence of fluorescein in any of the samples taken at the various springs.

The samples of water were analysed for tritium in Vienna at the laboratory of the Agency in a Packard TRI-CARB liquid scintillation spectrometer. Samples were analysed without any pre-treatment except in the case of seawater contamination and where samples were cloudy when they were distilled. The liquid scintillator used was the dioxane-naphthalene-water system. 2, 5-diphenyloxazole (PPO) at a concentration of 6 g/l was used as the phosphor, with 1, 4 bis-2(5-phenyloxazolyl)-benzene (POPOP), at a concentration of 0.3 g/l, as secondary phosphor. The solution prepared for counting consisted of water (20% by volume) and the 1, 4-dioxane solution of naphthalene (100 g/l) and the phosphors (80% by volume). Tritium was counted with an efficiency of 14% in a polyethylene counting vial, the background being 30 cpm.

Tritium appeared at Binikovi 10d after its introduction at Partheni. The maximum concentration was recorded 6 d later. Thirty-four days after the start of the experiment the concentration had decreased to less than 5% of its peak value, while the main portion of the peak passed in about 20 d.

Fig. 2 illustrates the variation of the discharge rate of the Binikovi spring with time reconstructed from gaugings made with a current meter. From this curve the variation of concentration c with accumulated discharge v from the start of the experiment was plotted and is shown in Fig. 3. The integral \( \int c \, dv \) gives the quantity of tritium discharged, is represented by the area under the curve and is found to be 600 c. It should be noted that this method of evaluating the data takes into account the variation of flow rate with time, which is not the case if the concentration is integrated with respect to time.

The volume of water underground between the sinkhole and the spring may be computed by the plotting of the product of the concentration of tritium and the accumulated discharge against the accumulated discharge (Fig. 4). The integral \( \int c \, dv \) is represented by the area under this curve and is found to be \( 1.83 \times 10^7 \) c/m\(^3\). The ratio of this area to that obtained from Fig. 3 gives the volume of water stored, which is found to be about 30 000 m\(^3\).

In the beginning of June 1962, 15 months after the introduction of tritium at Partheni, tritium appeared at Binikovi. The most recent sample available
for analysis still contains the same general level of tritium. The data from the second appearance of tritium at Binikovi cannot be treated in the same way as has been done for the first appearance, because insufficient discharge
data are available for the spring for the time the water has been underground. Also, more samples need to be analysed to disclose whether tritium will continue to appear. However, a rough estimate based on the average discharge rate (4 l/sec) of the spring and the average tritium concentration (4.5 × 10^-12 c/m³) shows that about 180 mc of tritium has emerged from the spring between June and September 1962. This quantity is very small in comparison with the balance still unaccounted for after the first appearance of tritium in 1961. It remains to be seen whether the concentration of tritium in future samples increases, which would then mean that the storage of water underground is far greater than the figure indicated from the first appearance of tritium.

However, from the data obtained from the first appearance of tritium it is possible to draw some conclusions as to the shape of the direct aquifer between Partheni and Binikovi. From the estimated storage and the distance the average cross-sectional area must be about 7.5 m² and the average water velocity 0.2 cm/sec. Since there is an altitude difference of about 360 m between the sinkhole and the spring, the aquifer cannot consist of a straight tube with the above cross-section. From earlier spelaeological investigations it is known that the average slope of the first 100 m is about 45°. Therefore, it is most likely that the water rapidly reaches a level not far above that of Binikovi and that the main reservoir is along the more level part. The rather large mixing indicated by the peak obtained at Binikovi would indicate the existence of a large pool or series of small pools along the path of the water. From the large delay before the second appearance of tritium at Binikovi it may be inferred that part of the flow went into a deeper underground system, the outlet of which joins the main aquifer shortly before it reaches Binikovi.

Seven days after the labelling of the water entering the sinkhole at Nestani when the rate of inflow was of the order of 3 m³/sec, tritium appeared at the coastal spring south of Kiveri. From Fig. 5 it is seen that the maximum concentration was reached 1 d later and no further tritium was detectable 9 d after its first appearance. To date no tritium has been
found at any of the other sampling points. This spring cannot be gauged by any of the conventional methods. However, if one assumes that all of the 400 c of tritium introduced at Nestani has appeared at Kiveri, then it is possible to calculate the discharge of this spring from the following equation:

\[ Q = \frac{M}{\int c \, dt}, \]

where \( Q \) = flow rate, \( M \) = quantity of tritium added at Nestani and \( c \) = concentration of tritium at the spring. The integral \( \int c \, dt \) evaluated from the area under the curve in Fig. 5 is found to be 20.7 c sec/m³. Substituting this value and that for \( M \) in the above equation, we compute \( Q \) to be 19.3 m³/sec. The hydrologists, however, consider this value to be too large by a factor of two to four for the estimated catchment area for this spring. This objection cannot be easily substantiated since cases are known where the water divide does not follow the topography. Also, nothing is known of the possible additions of water to the stream between Nestani and Kiveri. From Fig. 5 it is seen that a second peak occurred 2 d after the main peak. It is conceivable that during the period 13 - 15 February an appreciable addition could have occurred as a result of a rainstorm, which would have had the effect of depressing the descending part of the main
peak. Unfortunately, no records are available for checking of this point. Another possible explanation of this second peak is that part of the flow from Nestani to Kiveri may have branched off from the main stream and then further on joined it again, with a consequent time lag. The flow-rate figure is, of course, based on the assumption that all the 400 c has appeared at Kiveri. If only 200 c has appeared, then the flow rate would be about 10 m$^3$/sec. Analysis of later samples may indicate the appearance of tritium at some of the other springs, which would reduce the estimated discharge of this spring. This would seem to be a distinct possibility if the time taken for the second pulse of tritium to appear at Binikovi is borne in mind.

CONCLUSIONS

The investigation described in this paper has demonstrated the usefulness of tritium as a tracer for water for distances up to 30 km and has indicated the type of information which may be deduced from the data. Experience gained during these experiments has shown the necessity of supporting hydrological data such as the regular gauging of springs. Even more important is the fact that such an experiment may extend over a number of years, and for the maximum amount of information periodic sampling at the various points must be continued.

The planning of such an experiment must take into consideration the fact that addition of artificial tritium may prevent the application of naturally occurring tritium to hydrological problems. The present experiment may be criticized in that a quantity of tritium was introduced in the second stage similar to that which was unaccounted for from the first stage. One may ask whether the tritium at Kiveri originates from that introduced at Partheni or Nestani. In the authors' opinion this tritium must have come from Nestani. The steep rise and fall of the pulse observed at Ay. Ghorgios is not what would be expected if the tritium had spent eleven months underground. Also, the transit time from Nestani to Ay. Ghorgios is more in line with the speed observed over a distance of 40 km in another tracing experiment with tritium, also in a karst limestone terrain, in the region of Trieste.

DISCUSSION

R. HOURS (Chairman): I should like to make one or two comments on this investigation, which, I think, constitutes the first large-scale hydrological experiment in Europe involving the injection of artificial tritium into the ground. This fact makes the experiment an important landmark in the "tritium controversy". As you all know, there is one school of thought which is opposed to the use of artificial tritium injections altogether on the grounds that the purity of underground waters should be preserved and that it is sufficient to rely entirely on measurements with natural tritium. Of course in certain cases - e.g. when measuring the flow rates of large rivers - the problem is not a serious one because the tritium is rapidly washed into the sea.

I think it is important to remember that in this investigation samples of water were taken from the springs before the tritium was injected, that
the concentration at injection was of the order of the maximum permissible concentration (MPC) for drinking water and also that a second appearance of tritium was recorded at Binikovi 15 months after the tritium injection at Partheni. With the possible exception of Co60-labelled cobalticyanide, the use of which also gives rise to certain problems, I cannot see how this latter result could possibly have been obtained with any other tracer, especially as it is planned to carry out further sampling and analyses in the future.

It seems to me that what the International Atomic Energy Agency (IAEA) is doing - i.e. making use of both natural and artificial tritium, depending on circumstances - is the most reasonable course of action. A whole programme has been worked out for sampling and measuring natural tritium and a special laboratory has been equipped for the purpose in Vienna.

On the subject of the fluorescein, I am not particularly surprised that no trace of the 12.5-kg injection at the Nestani sinkhole was found at the Kiveri spring. In the case of the tritium, 400 c was fed into this same sinkhole for one hour. The rate of inflow was 3 m^3/s, so that this would yield an average concentration of 37 mc/m^3. Eight days later a peak of 135/nc/m^3 was recorded at Kiveri. This corresponds to a 275-fold dilution. As the initial concentration of fluorescein - injected within a period of 5-10 min - was of the order of 7 mg/l (7 ppm), a dilution on this scale would give rise to a maximum concentration at Kiveri of about 2.5 µg/l (2.5 pp10^9).

A few years ago a series of experiments were carried out (Houille Blanche, numéro special A, 1955) in the course of which it was found impossible to pass a 10-ppm solution of fluorescein through a 1-m column filled with very slightly argillaceous sand (column length - 1 m, column diam. - 5 cm, fluorescein - 0.2 l at 10 ppm eluted by 5 l pure water). In other words, in the Greek experiment the presence of argillaceous formations along the groundwater route followed would have sufficed to prevent the passage of the tracer.

D. B. SMITH: The outline of the curve in Fig. 2 is somewhat hypothetical since there is only one point at the top of the curve and three points after about 30 d. If one assumes a less rapid drop in the flow rate, a larger amount of activity would be discharged. This could perhaps account for the missing tritium. It might be useful to make observations at Binikovi, say at the beginning of a dry-weather period when the flow rate is decreasing from a level of 110 m^3/h, in order to establish the actual form of the hydrograph.

B. R. PAYNE: I agree that it is unfortunate that no flow rates were measured when the main part of the tritium appeared at Binikovi. However, if one assumes that the decrease in flow rate was linear then this would only account for about an additional 30 c of tritium coming out at Binikovi.

R. AMBROGGI: I should like to make a few general observations on this investigation which, as you may know, is one of the projects which have been or are being implemented in Southern Europe and the Mediterranean under the United Nations Special Fund. This particular project was carried out with the Food and Agriculture Organization of the United Nations (FAO) acting as executing agent and the IAEA providing specialist advice on the use of isotopes.
The first point I want to make concerns the hydrogeology of the region, a knowledge of which is essential to a proper understanding of the results obtained at the Binikovi spring. On top of an autochthonous layer, which is considered practically impermeable, there is an overthrust nappe made up of permeable limestone. The permeable series consists of a very young, imperfectly formed upper karstic layer and an older, well-formed karstic layer at the base, almost in contact with the autochthonous series. Most of the water moving through the karst passes through the older karstic layer, giving rise to the large springs of Kefelari, Myli, Kiveri and Anavalos spaced out along the coast. The Binikovi spring, on the other hand, is situated in the younger karstic level, where the movement of water is only slight. Furthermore, the karstic waters move in a general NW-SE direction down from the sinkholes. Binikovi, however, lies along a WSW-ENE line in relation to the injection point at Partheni. These two features peculiar to Bikinovi may furnish an explanation of the results relating to this spring.

The second point relates to the value of 20 m$^3$/s calculated for the flow rate at the Kiveri spring. This seems to be too high. In the first place, if the flow is observed at low tide - there is a fairly appreciable tide in this area - it does not appear to be more than 5 m$^3$/s. And, furthermore, the flow rates at comparable springs such as Myli and Kefelari, which have been fairly well gauged, are far below 20 m$^3$/s.

Thirdly, it is clear that two injections of tritium made within the space of one year are liable to produce overlapping results when the samples used in both cases are taken from the same springs. However, it is pretty certain that the tritium found at Kiveri did in fact come from the second injection at Nestani. This would tie up with the fact that when rain falls on the plateau where the sinkholes are situated there is generally an interval of between a fortnight and a month before any impact can be noted at the springs. It would also fit in much better with the general line of flow of the main karstic layer.

The last comment I have to make is one the economics of the project. The considerable number of samples (about 1200) needed for these experiments involved a substantial amount of actual sampling work over a long period (2 yr) and over appreciable distances (20 - 30 km). A considerable amount of laboratory time was also required. The resultant cost of the analyses was therefore fairly high. This might be one point where improvements could be made when this very useful experiment is repeated.

B. R. PAYNE: Thank you for your interesting remarks on the hydrogeology of the area. As to the flow rate of the spring at Kiveri the figure of 20 m$^3$/s is, of course, based on the assumption that all of the 400 c came out of the spring. If the actual figure is of the order of 5 m$^3$/s, it remains to be seen what has happened to the balance of tritium. Periodic sampling is being continued so that the water leaving the various springs may be analysed for tritium.

I would mention that in collaboration with Dr. W. Dansgaard we are also measuring the oxygen-18 and deuterium content of water from different parts of the area to obtain some information on the origin of the various spring waters.

R. HOURS: I understand that well over 1000 water samples were taken and that only a fraction of these were measured - without being concentrated
beforehand - in an automatic TRI-CARB liquid scintillation spectrometer. By using electrolysis to concentrate the samples slightly and then carrying out measurements in a large-volume low-background gas counter, I would have thought that you could have achieved at the very least a 50-fold increase in the sensitivity of your measurements. You could then have decreased the quantity of tritium injected by the same factor. Ten or twenty curies might have been sufficient.

Obviously, if you had used this system individual measurements would have taken much longer. However, since the radioactive wave could reasonably be expected to take a long time to reach the distant sampling points, I wonder whether it would not have been possible to reduce considerably the number of samples measured if not the number actually taken. Once the samples were found to be active, the number of measurements could always have been increased.

B. R. PAYNE: Your suggestion would have led to a saving on the cost of tracer but I think this would have been absorbed by the increased cost of analysis and might indeed have led to a more expensive experiment. I am sure that the analytical work would have taken much longer. However, we have used electrolytic measurement of some of the samples taken during the second appearance of tritium at Binikovi.

D. B. SMITH: What is the annual rainfall in this area?

R. AMBROGGI: In the mountainous area where the sinkholes are situated, the rainfall amounts to 1000 - 1200 mm. At the coast the figure is around 400 mm.

D. B. SMITH: Assuming then an annual rainfall of 100 cm, of which 30% enters the groundwater, and taking the present average tritium activity of the rain to be 150 tritium units, approximately 60 c of tritium will enter the groundwater in an area 20 km square, i.e. similar to the area of the investigation. I think this would invalidate the suggestion made by Mr. Hours that 10 c of tritium could have been used. The natural tritium which occurs would outweigh a small amount of this sort. The use of 400 c is fully justified, I think.

R. HOURS: An initial average concentration of 150 tritium units in the water would correspond to a value of about 0.5 μc/m³. Since the peak activity recorded at Kiveri amounted to 135 μc/m³, a peak of several μc/m³ would still have been visible. This would have meant an initial injection 10 or 20 times less, i.e. 40 or 20 c. I agree that this peak would not have been so obvious and that in an experiment on this scale it is wiser to operate with a comfortable margin and thus a higher degree of sensitivity. There is the further argument that once several hundred curies of tritium have been injected into an aquifer there can be no question of effecting further natural-tritium measurements for some considerable time afterwards. So I suppose that on the whole it is better to inject several hundred curies so as not to miss any waves of activity.

There is, however, one point in favour of using a low initial activity. It does mean that stronger doses can be injected into the same aquifer later on and that one will be distinguishable from the other. But of course this presupposes the use of facilities capable of measuring very low levels of activity.
J. F. CAMERON: I should like to give one or two details of a similar experiment that was carried out near Trieste by the Trieste Geophysics Institute in collaboration with the Yugoslav Electricity Authority and the IAEA. The Yugoslav Electricity Authority was thinking of using a stream with a flow rate of about 2 m³/s as a source of hydroelectric power. This stream disappears into a limestone grotto and was thought to re-appear at a spring at Timavo, about 37 km away from the point of disappearance. As this spring supplies most of the water for the town of Trieste, it was important to make sure that the water supply was not being interfered with. To find out whether there was in fact any connection and also to measure the contribution of the stream to the spring water, 200 mc of tritium were injected. Samples were taken at Timavo and also at some intervening caves and boreholes. From an analysis of the tritium content of the water it was found that it takes about 11 d for the water of the stream to travel the 37 km to Timavo, and that the stream accounts for only about 1% of the Timavo discharge.

In this experiment about 60% of the tritium was recovered. After the stream disappears into the ground it runs alongside some impermeable strata and comes into contact with the karst again about 5 km before the spring, so that there is quite a high probability of some of the water again passing into the karst and thence into the sea. In view of this, a 60% recovery is quite reasonable.

Throughput curves were analysed at each of the sampling points and it was found that in its passage underground the stream sometimes diverged and then converged again; the problem of underground caverns and tunnels is quite complicated in karstic areas.
TWO USES OF BROMINE-82 IN UNDERGROUND HYDRODYNAMICS. The measurement of velocities in aquifers in alluvium is of great importance in determining the most significant parameters in underground hydrodynamics.

The authors describe the use of radioactive tracers in velocity measurements in aquifers previously studied by refined mechanical methods and methods based on the laws of transitory movement, as formulated by Theis and adapted by de Gelis.

These measurements are carried out by injecting Br\(^{82}\) - labelled NH\(_4\)Br into a piezometer and then surveying the arrival of the activity in a neighbouring shaft. The results obtained provide valuable clarification of the distribution of velocities in an alluvial formation and represent a contribution, based on standard means of detection, to the verification of Theis' law under conditions which depart appreciably from the basic assumptions.

Another application of radioactive labelling of water with Br\(^{82}\) which is dealt with in the paper concerns the localization of permeable strata or seams in a geological calcareous marl series, intended to support quay walls in a harbour basin.

The tracer, after being forced into the formation by inactive water, was pumped back and then local and overall measurements of activity permitted estimation of the recovered fraction and precise localization of the zones of recharge.

In both cases, the radioactive tracers represented the only means at present available for resolving the problems raised.
Дается описание применения радиоактивных индикаторов при измерениях скоростей движения воды водоносного пласта, уже изученных с помощью механических методов, основанных на законах переходного движения, определенных Цейсом и принятых Де Желисом.

Измерения были предприняты для того, чтобы изучить, после введения в пьезометр $\text{BrNH}_4$, меченного бромом-82, проявление активности в соседнем колодце. Полученные результаты позволяют сделать интересные уточнения относительно распределения скоростей в наносном образовании и содействуют, при использовании существующих методов обнаружения, проверке действенности закона Цейса в условиях, значительно удаленных от условий основных гипотез.

Другой упомянутый метод применения радиоактивного мечения воды с помощью брома-62 относится к локализации водопроницаемых слоев и прослоев в серии геологических пород горного известняка, используемого для облицовки стен набережной в морском порту. Индикатор, попавший под действием неактивной воды в образование, был откачен обратно; местные и общие измерения активности позволили установить осадок и более точно локализовать зоны восстановления.

В обоих случаях использование радиоактивных индикаторов представляло собой единственный имеющийся в настоящее время в распоряжении путь решения поставленных проблем.

I. MESURE DE LA VITESSE D'UNE NAPPE A PROXIMITÉ D'UN PUITS DE POMPAGE

1. But de la mesure

L'utilisation de traceurs radioactifs a été décidée pour tenter de donner un complément de précision à l'étude des conditions hydrodynamiques d'une nappe. Toutefois, ces conditions d'écoulement, étudiées par des méthodes classiques, ne mettaient que des évaluations théoriques moyennes des vitesses, à des distances variables de l'axe du puits de pompage.

Or, la vitesse réelle est d'un intérêt évident et l'utilisation de traceurs traditionnels pouvait résoudre en partie le problème.

Cependant, dans le cas d'une nappe en mouvement théoriquement uniforme dans une formation alluviale, il pouvait être intéressant de rechercher les valeurs les plus fortes de la vitesse ainsi que l'influence du pompage sur la direction du mouvement général de la nappe. De plus, l'hétérogéné-
ité naturelle des alluvions conduisait à tenter l'établissement de valeurs comparatives entre les diverses méthodes de mesure de la perméabilité.

2. Conditions géographiques, géologiques et hydrodynamiques

La région étudiée est située dans la vallée du Rhône, à l'aval de Montélimar. Le terrain est plat sans aucun relief, avec une pente moyenne de 1/1200 vers le sud. A l'est, le canal de dérivation du Rhône constitue un axe d'alimentation de la nappe, que l'on peut considérer comme linéaire. A l'ouest s'étend la large plaine alluviale où le Rhône, situé à plusieurs kilomètres, est un axe de drainage qui n'intervient, toutefois, pas directement dans notre étude.

Divers petits canaux de drainage et d'irrigation intéressent localement les caractéristiques géologiques, mais dans des conditions bien particulières. Ainsi, des pluies intenses sont suivies assez rapidement par une montée du niveau piézométrique de la nappe qui peut atteindre le niveau du sol. Ces canaux jouent alors un rôle d'alimentation.

Les conditions géologiques sont simples. Au-dessus de marnes sableuses gris-bleu datant de la dernière grande transgression pliocène dont la perméabilité à l'œdomètre est de 10-9 m/s, des alluvions constituées de galets, de graviers et de sable s'étagent sur la majeure partie du site, sur une hauteur de 7 à 10 m. Dans une zone latérale, toutefois à la suite d'études géophysiques et de sondages de contrôle, on a mis en évidence un surcroissement du lit fossile (bed-rock) jusqu'à une profondeur de 16 à 17 m.

Dans la partie centrale et ouest du site, les alluvions sont exemptes d'éléments fins, la taille minimum des éléments les plus fins étant de l'ordre de 0,3 à 0,5 mm, et en faibles quantités, inférieures à 5%. Ces alluvions sont à texture litée et, au seul aspect d'une tranche de quelques mètres de profondeur, il devient évident que la perméabilité doit varier assez sensiblement dans le sens horizontal et dans le sens vertical. Au-dessus des alluvions, un ensemble comprenant de la terre végétale et des limons plus ou moins organiques s'étale sur 0,5 à 3 m.

D'une manière générale, les alluvions proprement dites sont d'autant plus épaisses et d'autant plus exemptes d'éléments fins que l'on s'éloigne vers l'ouest. De même, l'épaisseur des limons superficiels décroît dans cette direction.

Il résulte des sondages et des mesures de mécanique des sols effectuées sur échantillons, que la marnes sableuse sousjacente est altérée par oxydation et plastique sur 0,30 à 0,50 m d'épaisseur, et que les limons organiques superficiels sont un peu plus plastiques et plus imperméables à leur base, au contact avec les alluvions. Dans l'ensemble, le réservoir de la nappe est bien individualisé.

Au point de vue hydrologique, deux nappes d'importance très inégale sont étagées dans les deux formations superficielles. La première, de faible volume, est contenue dans la partie supérieure des limons. L'eau est chargée en matières organiques colloïdales et l'extrait sec atteint 0,800 g. Cette nappe est très vite épuisée lors des périodes de sécheresse moyenne et ne présente pas grand intérêt.
La nappe des alluvions, par contre, est d'une importance particulière. La constance de composition chimique des eaux, dans l'espace et dans le temps, avec un extrait sec de l'ordre de 340 à 420 mg/l, permet de penser à un cheminement régulier disposé dans des formations homogènes en grand.

La perméabilité des alluvions et les caractéristiques hydrodynamiques de la nappe ont fait l'objet de quelques études que nous avons tenu à pousser assez loin, pour tenter de résoudre le problème du rythme d'alimentation saisonnière et annuelle, notamment. Les mesures de perméabilité ont été effectuées de deux manières:

a) Par étude différentielle, à l'aide de micromoulinet, de la perméabilité dans les sondages équipés en piezomètres.

Le micromoulinet mesure l'augmentation de vitesse de l'eau dans le sondage, en fonction des valeurs cumulées des perméabilités le long de l'axe. Une crépine suffisamment ajourée permet à l'eau de circuler librement dans le sens terrain-sondage. Cette mise en vitesse est provoquée soit par pompage, soit par injection. La mise en dépression par pompage paraît préférable.

Ces mesures ont montré l'existence de passages à perméabilité assez forte de 1,9 et 2,7 m²/s/sur des épaisseurs de 0,50 m environ.

Certains sondages, notamment les deux cités ci-dessus, montrent des zones de 1,5 à 2 m sans perméabilité apparente, du moins sous la dépression assez faible provoquée ici.

Enfin, la base des alluvions, sur 1 à 3 m, apparaît d'une manière générale comme assez perméable.

b) Par pompage dans les zones choisies en fonction de leurs caractéristiques géographiques et géologiques, et interprétation suivant les méthodes de Dupuit et de Theis.

La comparaison des deux méthodes a donné de bonnes approximations. Ainsi la transmissivité*, calculée par la méthode THEIS [1] (avec l'approximation satisfaisante de DE GELIS [2]), d'après le rabattement, donne à différentes distances du puits les valeurs suivantes:

- à 3 m du puits $T = 7,04 \cdot 10^{-2} m^2/s$,  
- à 12 m du puits $T = 6,80 \cdot 10^{-2} m^2/s$,  
- à 15 m du puits $T = 7,50 \cdot 10^{-2} m^2/s$,  
- à 235 m du puits $T = 7,72 \cdot 10^{-2} m^2/s$.

On peut en déduire la valeur du coefficient de perméabilité $K$, ainsi que la vitesse apparente (Darcy) et la vitesse réelle effective.

De la valeur moyenne de $T$, calculée soit à partir de la formule du régime transitoire de Theis, soit à partir de la formule classique de Dupuit, on tire

$$K = 8,5 \cdot 10^{-3} m/s.$$  

Toutefois, certaines caractéristiques de la courbe du débit spécifique

* Rappelons que la transmissivité est le produit $K \cdot H$ du coefficient de perméabilité $K$ (de Darcy) par la hauteur de la tranche perméable.
en fonction de log t permettent de conclure à des passages où

\[ K = 2 \cdot 10^{-2} \text{ m/s}. \]

La vitesse \([3]\) apparente devient donc, avec un gradient de 1/1200,

\[ V_a = 1 \cdot 10^{-6} \text{ m/s soit } 8,6 \cdot 10^{-2} \text{ m/j}. \]

La vitesse réelle effective peut donc être évaluée à 0,17 m/j.

Un calcul analogue montre que sur une distance de 3 m depuis 1'axe du puits, la vitesse réelle effective, avec une porosité de 50%, serait de 1'ordre de 15 m/h. Nous verrons que cette valeur est très largement dépassée par les mesures directes.

3. Modalités de l'injection, du pompage et des mesures

Les conditions que devait remplir le traceur radioactif choisi, le \(^{82}\)Br, étaient:

- Faible concentration maximum admissible (CMA): \(3 \cdot 10^{-3} \text{ c/m}^3\).
- Une période courte donnant la possibilité de faire plusieurs injections successives. La période de 36 h convient particulièrement.
- Peu ou pas d'adsorption par le terrain. A ce sujet des recherches antérieures [4, 5, 6] ont montré que le \(^{82}\)Br présentait les adsorptions les plus faibles sur de nombreux terrains. Nos essais ont largement confirmé les résultats antérieurs. Toutefois, un entraîneur a été utilisé à chaque injection du traceur.
- Détection facile et immédiate, ne nécessitant pas de prélèvements. Le rayonnement gamma du \(^{82}\)Br est facile à détecter, notamment avec des scintillateurs portatifs.
- Discrétion d’emploi.

Le brome irradié à la pile EL3 de Saclay sous forme de BrNH\(_4\) a été utilisé avec des traceurs traditionnels colorants et chimiques de l'eau: fluorescéine et bichromate de potassium.

3.1. Première série d'injections

Dans la première série de mesures de vitesses, les injections ont été effectuées dans trois piézomètres situés autour du puits de pompage (fig. 1). Le niveau du puits était rabattu de 0,928 m par un pompage à débit constant de 150 m\(^3\)/h, commencé quarante-huit heures avant le premier traçage. Ainsi le cône de dépression de la nappe autour du puits était à peu près stabilisé pendant la durée des observations (fig. 2).

L'activité était mesurée à 200 m environ du puits de pompage, à la sortie de la conduite de rejet, avec le matériel suivant:

- trois compteurs Geiger-Müller du type SRAT, GMT 14 et Scintillac,
- un scintillateur PRI avec enregistreur (cristal 7\(\text{°}\)X 6\(\text{°}\)).

\* La CMA pour \(^{131}\)I est de \(1\cdot10^{-3} \text{ c/m}^3\) pour le personnel exposé et 30 fois moindre pour les populations non exposées.
Un relevé du bruit de fond et un étalonnage des appareils ont été faits avant de procéder aux quatre injections successives du traceur.

a) Piézomètre Nord, situé à 10 m du puits, première injection (fig. 1 et 3)
   5,4 mc de $^{82}$Br,
   50 g de KBr en solution dans 10 l d'eau,
   300 g de fluorescéine.
   La solution active contenant l'entraîneur a été simplement déversée dans le piézomètre.
   La sortie du traceur a été décelée d'abord par une première et courte bouffée d'activité enregistrée 34 min 30 s après l'injection. Puis la fluorescéine a été vue deux minutes plus tard. Enfin, l'apparition de l'activité principale a commencé 50 min après l'injection.

b) Piézomètre Nord, deuxième injection (fig. 1 et 4)
   100 mc de $^{82}$Br
   200 g de KBr en solution dans 10 l d'eau.
DEUX UTILISATIONS DU BROME-82

La solution active et l'entraîneur ont été homogénéisés sur toute la hauteur du piézomètre.

La détection est très nette et commence 2 h 14 après l'injection.

c) Piézomètre Sud, situé à 8 m du puits (fig. 1 et 5)
18 mc de $^{82}$Br
250 g de KBr en solution dans 10 l d'eau.
Solution active et entraîneur ont été déversés sans homogénéisation.
Le $^{82}$Br s'est manifesté, d'une façon particulièrement nette, 18 min après son injection.

d) Piézomètre Est, situé à 40 m du puits (fig. 1)
18,5 mc de $^{82}$Br
250 g de NaBr en solution dans 10 l d'eau.
Le $^{82}$Br a été décelé, sans grande précision, 6 h environ après son injection.

Résultats

Il est possible, connaissant les heures précises d'injection et de détection du traceur, d'évaluer la vitesse réelle d'écoulement de l'eau à proximité du puits. On obtient ainsi les résultats suivants pour les différents essais:

a) Piézomètre Nord, première injection: 17,4 m/h pour la première bouffée d'activité et la fluorescéine et 11,3 m/h pour la deuxième détection. La vitesse la plus rapide détectée par le bref pic de radioactivité correspond
vraisemblablement à un écoulement superficiel plus rapide précédant l'arrivée de l'onde principale. Le phénomène est en accord avec les formules théoriques. Remarquons que les deux minutes qui séparent la détection de la radioactivité de celle de la fluorescéine peuvent être le fait d'une plus grande précision de détection du brome.

b) Piézomètre Nord, deuxième injection: la vitesse est de 4,53 m/h, correspondant à un marquage homogène sur toute la hauteur du piézomètre.

c) Piézomètre Sud: la vitesse est de 26,60 m/h et correspond au mouvement superficiel des filets liquides.

d) Piézomètre Est: la vitesse est plus basse, de l'ordre de 6,5 m/h, et se rapproche de celle obtenue au piézomètre Nord.

La direction générale NNE-SSO de la nappe explique aisément cette vitesse ainsi que la mauvaise détection de l'arrivée de l'activité qui, à cet essai, a été assez faible. L'hétérogénéité de la nappe alluviale, due à la répartition lenticulaire de la granulométrie, paraît être assez bien mise en évidence.

D'autre part, il est possible à partir de l'étalonnage de l'appareil et des enregistrements d'activité à la sortie de l'eau de pompage, de connaître le pourcentage de récupération du traceur.

Dans deux des cas où l'enregistrement a donné des résultats particulièrement nets, on a pu calculer que le pourcentage de récupération était, lors de la première série d'injections, de 40% à la deuxième injection du piézomètre Nord et de 86,5% au piézomètre Sud de cette même série. Dans le cas du piézomètre Est, une petite partie seulement de la masse d'eau marquée a été intéressée par le pompage.
3.2. Deuxième série d’injections

La deuxième série d’injections (fig. 6) de $\text{Br}^{82}$ a été réalisée sur un site analogue à celui de la première série, mais avec intervention d’un canal dont le rôle devait être précisé dans le bilan du pompage. La perméabilité moyenne est également du même ordre de grandeur ou légèrement inférieure.

Initialement, on pouvait penser que dans le tronçon étudié, le canal d’irrigation alimentait la nappe, compte tenu de rapides différences de niveau du canal, de l’ordre de 0,25 m d’amplitude. La conduite des essais a été menée en fonction de cette hypothèse de travail.

Les modalités de pompage ont été analogues à celles de la première série. Le rabattement, pour un débit de 80 m$^3$/h, a été de 0,57 m après 50 h de stabilisation. Les modalités d’exécution, par contre, ont été quelque peu différentes. En effet, les injections successives ont été réalisées de la manière suivante:

a) Piézomètre 1P, distant de 10 m du puits

Injection de 16 à 17 mc de $\text{Br}^{82}$ en 38,5 ml, avec une solution de 400 g de NaBr dans 5 l d’eau. Dans le même temps, 300 g de fluorescéine ont été ajoutés dans 30 l d’eau.

La radioactivité a été décelée en même temps que la fluorescéine, assez atténuées l’une et l’autre, 7 h 36 après l’injection, soit une vitesse de 1,33 m/h.
b) Piézomètre Nord-Est, distant de 50 m du puits
Onze heures après la première injection, mise en œuvre de 150 mc de $^{82}$Br avec 800 g de NaBr dissous dans 5 l d'eau accompagnés de 1 kg de Cr$_2$O$_7$K$_2$ dissous dans 5 l d'eau et de 50 l d'eau de lavage.
Aucune radioactivité n'a été décelée en onze heures d'enregistrement dans le puits, et les analyses n'ont pas montré de présence de bichromate.

c) Piézomètre Sud, distant de 15 m du puits
Injection de 75 mc de $^{82}$Br avec une solution de 800 g de NaBr dans 5 l d'eau, puis de 50 l d'eau.
Aucune radioactivité décelée. On constate même une légère baisse par rapport à l'activité résultant de la première injection. Les mesures ont duré 30 h environ.

**Résultats**

La recherche de l'activité a été faite dans toutes les zones où l'on pouvait accéder, et notamment en bordure du canal d'irrigation. Dans ce dernier cas, quelques pointes ont bien été constatées, mais leur valeur assez faible n'a pas permis de les différencier à coup sûr des fluctuations extrêmes du bruit de fond.

On se trouve donc en présence de deux injections qui n'ont pas été suivies de réapparition du traceur et d'une seule injection où l'atténuation de l'activité du $^{82}$Br n'a été enregistrée que par le scintillateur à cristal de $^{7}{\gamma\times 6{\gamma}}$, particulièrement sensible. Une confirmation est apparue avec la présence de fluorescéine, décelable seulement en lumière plongeante dans le puits de pompage, soit à une concentration un peu supérieure à 10 γ/l, limite de détection en vision normale.

Toutefois, dans les deux derniers cas, cette absence de traceur dans le puits est significative. On peut en effet envisager un écoulement de la nappe totalement différent de celui envisagé initialement et dirigé à 45° vers le canal. Une telle direction expliquerait les résultats constatés.

Rappelons que lors de la première série, à la suite de l'une des injections (piézomètre Est, figure 1) réalisée dans une zone où la direction de la nappe convenait mal à la restitution, on n'a obtenu également qu'une faible mise en évidence du traceur.

Enfin, le bichromate n'a été retrouvé nulle part.

**II. MÉTHODES DE L'INJECTION DANS UN SONDAGE**

L'étude de la perméabilité et surtout de la localisation des venues d'eau dans un sondage peut être réalisée par injection d'éléments radioactifs, puis par la mesure de l'activité suivant diverses opérations.

1. But de la mesure — Méthodes classiques d'étude de la perméabilité

1.1. But de la mesure

La localisation et les mesures de perméabilité avaient pour but de fixer les conditions dans lesquelles pourraient être effectués des travaux d'agran-
dissémination d'un bassin à flot du port de Boulogne. Les terrassements doivent atteindre des profondeurs importantes, et la proximité de la mer dont les marées dépassent 6 m d'amplitude oblige à étudier particulièrement les circulations dans les séries géologiques intéressées par les travaux.

Par ailleurs, un projet d'écluse conduit les projeteurs à envisager les percolations possibles dans les séries géologiques du bassin et de ses environs.

1.2. Méthodes classiques d'étude de la perméabilité

La perméabilité peut être mesurée par des moyens traditionnels, en utilisant des formules générales du type classique basées sur la loi de Darcy, et consistant en un pompage ou une injection d'eau dans une cavité de forme géométrique connue (essai dit «Lefranc»):

\[ Q = CKh \]

où \( Q \) est le débit pompé ou injecté,
\( C \) est un coefficient dépendant de la forme du volume testé,
\( K \) est le coefficient de perméabilité en m/s (Darcy), et
\( h \) est le gradient correspondant à la hauteur d'eau au point d'essai.

La valeur de \( C \) est en général difficilement déterminable avec précision, mais on arrive la plupart du temps à des résultats utilisables.

D'autres méthodes sont basées sur la mesure de la vitesse de l'eau dans un sondage, vitesse obtenue soit par pompage depuis la tête du sondage, soit par injection d'eau. La mesure de cette vitesse est effectuée par un micromoulinet permettant de compter le nombre de tours d'hélice par unité de temps. La limite inférieure de la vitesse mesurable est de l'ordre de 1 à 1,5 cm/s.

Lorsque le procédé de pompage est choisi, c'est-à-dire de la mise en dépression du sondage, ce qui est le cas général en raison de la commodité d'exécution, la courbe des vitesses cumulées donne l'allure des variations de la perméabilité. Les figures 7a et 7b donnent les résultats obtenus dans les sondages étudiés ici. L'interprétation est basée sur l'hypothèse suivante:

Dans un terrain formé de couches horizontales et homogènes, où les écoulements se font dans un plan horizontal, il est possible d'utiliser une formule dérivée de la formule de Dupuit. En effet, pour chaque unité stratigraphique - ou chaque couche - de ces terrains, la perméabilité de la tranche traversée par le sondage peut être définie par la formule suivante:

\[ K = \frac{Q \ln(R/r)}{2\pi h Z} \]

où \( K \) = la perméabilité en m/s,
\( \ln \) = le logarithme népérien,
\( Q \) = le débit en m³/s,
\( R \) = le rayon d'action,
\( r \) = le rayon efficace du forage,
\( Z \) = le rabattement,
\( h \) = l'épaisseur de la couche essayée.
R et r ne sont pas connus dans les cas courants, mais on sait par ailleurs que le logarithme népérien de $R/r$ varie dans des limites étroites que l'on peut négliger en première approximation et qui conduit à admettre

$$\ln(R/r) = 2\pi$$

d'où la formule

$$K = \frac{Q}{hZ}.$$  

Toutefois, la précision des mesures est perturbée par les tourbillons provoqués par le passage de l'eau à travers les trous de la crépine, ainsi que par le désaxage possible de l'hélice par rapport à la direction du courant. Il en résulte que la détermination du débit provenant d'une couche de terrains perméables fournissant moins de 10% du débit total circulant à ces niveaux dans le forage est difficile.

* Ces précisions sont basées sur de nombreuses expériences réalisées par le Bureau d'études de la Société Solétanche.
Enfin, il aurait été possible, dans certains cas, et bien que la précision soit notablement plus faible, d'utiliser des différences de résistivité, par exemple en injectant dans le sondage des solutions chargées de chlorure de sodium. Ces procédés, qui nous ont donné des résultats intéressants dans d'autres cas, étaient inutilisables ici, car nous avions affaire à de l'eau de mer.
2. Conditions géologiques et hydrodynamiques

La figure 8 donne une allure générale de la zone où ont été réalisées les reconnaissances géologiques, géotechniques, et hydrodynamiques. D'une manière générale, les terrains sont disposés sans accident géologique notable, avec un faible pendage vers l'ouest.

Par ailleurs, un ancien fleuve côtier quaternaire, la Liane, traversait l'emplacement actuel du port de Boulogne, suivant une direction générale est-ouest.

2.1. Conditions géologiques

Les caractéristiques géologiques des terrains intéressés par le projet d'extension du bassin actuel et par les injections de brome sont les suivantes, par ordre chronologique croissant:

a) Sédiments quaternaires correspondant au lit fossile de la Liane, dont le tracé a été retrouvé au cours de différents travaux effectués dans le port
de Boulogne. Ces sédiments peuvent s'étager sur une épaisseur de 33,50 m. Ils sont composés de sable en général homogène, avec quelques galets de silex, quelques plaquettes de grès et des fragments de coquilles. La granulométrie est assez homogène, les dimensions pouvant aller de 0,1 à 0,5 mm, avec des pourcentages de 95% du total entre ces deux mailles. Des bancs de vase sont intercalés dans les sables, mais se trouvent surtout à la base du quaternaire sur 3 m environ d'épaisseur. Ces vases comprennent de 5 à 10% d'éléments plus petits que 5 µ, et 40 à 50% de silt. Par ailleurs, des vestiges de bois fossile ont été rencontrés par endroit.

La figure 10 donne une idée de la succession lithologique de ces sédiments. On verra que, de 0 à 13,5 m, sous une apparente homogénéité, existent en réalité des passages à perméabilité différentielle importante.

b) Série de calcaires et de marnes du Jurassique supérieur en bancs d'épaisseur assez variable qui constitue les terrains situés en dehors du recouvrement des sables et des vases quaternaires de la Liane. L'épaisseur maximum des bancs calcaires intéressés par les sondages injectés ne dépasse pas 3,5 m, et encore faut-il signaler la présence de petits bancs de marne de quelques décimètres, intercalés sous forme de joints.

Les figures 9 et 10 donnent la succession lithologique rencontrée aux deux sondages S4 et S5.

L'étude des calcaires au microscope montre des éléments composants classiques tels que fragments de coquilles, grains de quartz, calcite, qui n'interviennent pas dans notre étude.

La phase argileuse des marnes a été étudiée en diffractométrie aux rayons X. On a pu déterminer la présence de montmorillonite, d'ililite, et de kaolinite. Ainsi un échantillon prélevé dans le sondage S4 à 11 m de profondeur montre un pourcentage de 90% de montmorillonite + illite, et de 10% de kaolinite.

Des vestiges de dissolution et d'altération sont visibles, notamment dans les bancs calcaires, quelque peu gréseux à l'origine. Certaines dissolutions ont même enlevé une partie du ciment de calcite, laissant des bancs altérés formés principalement de sable. Ces altérations jouent un rôle évident dans la répartition de la perméabilité.

2.2. Conditions hydrodynamiques

Les conditions hydrodynamiques sont relativement simples. En effet, le port de Boulogne, notamment à l'emplacement du projet d'extension du bassin, est affecté par des circulations d'eau douce provenant de la côte, par drainage naturel, donnant ainsi en profondeur une circulation que l'on peut rendre responsable des dissolutions de calcite dont nous avons parlé plus haut. Des mesures de résistivité ont mis ces circulations en évidence tout le long des sondages.

Par ailleurs, les marées provoquent un mouvement de va-et-vient des masses d'eau à l'intérieur des séries de grès, de sables et de calcaires, mouvement dont l'intensité est d'autant plus grande que l'on s'approche de la mer. La figure 11 donne une idée des variations de niveaux d'eau dans quelques-uns des sondages intéressés. Il y aura lieu de tenir compte de l'effet de ces mouvements de marées au moment de l'interprétation des résultats.
Modalités de l’injection

1. 1.000 m³ d’eau de mer en 1440 s, soit $Q_1 = 0.694 \text{ l/s}$.
2. 1.770 m³ d’eau de mer +10 mc de $^{85}\text{Br}$ en 1500 s, soit $Q_2 = 1.180 \text{ l/s}$.
3. 0.940 m³ d’eau de mer en 1065 s, soit $Q_3 = 0.882 \text{ l/s}$.
4. Pompage à débit $Q_4 = 2.888 \text{ l/s}$.

Radioactivité mesurée au scintillateur

2. 17 h après la fin de l’injection, 16 h 50 après le début du pompage, 15 h après la fin du pompage à la marée descendante le 18 juillet 1962 à 17 h 30.
3. Méthode d'injection du traceur

Le choix du $^{82}$Br comme traceur a été basé, comme pour l'utilisation précédente, sur les critères classiques suivants que le brome satisfait entièrement:
- période convenable de l'élément,
- faible adsorption par les minéraux argileux,
- rayonnement facilement détectable.

En ce qui concerne l'adsorption, des mesures avaient été faites préalablement dans l'hypothèse d'une mesure de vitesse entre deux sondages voisins, technique différente de celle utilisée ici.

Ces déterminations d'adsorption ont été effectuées sur deux traceurs traditionnels, la rhodamine $B$ et la fluorescéine, par contact à l'agitateur, de 100 g de matériau avec le colorant, passage à la centrifugeuse et mesure de la coloration dans la phase liquide au spectrophotomètre.

La fluorescéine n'a montré aucune adsorption décelable dans les marnes alors que la rhodamine $B$ a montré une sensibilité considérable, 92,5 et 96,06% de ce colorant ayant été fixés par les minéraux argileux.

On notera par ailleurs que, dans le cas de Boulogne, la présence de brome dans l'eau de mer — 0,188% de Br (en pourcentage de l'extrait sec total [7]) — constituait à elle seule un effet analogue à celui d'un entraîneur.

On a donc jugé que le problème de l'adsorption du traceur était déjà plus ou moins résolu dès l'origine, mais malgré cela, un entraîneur a été ajouté au cours de l'injection, à raison de 45 g/m$^3$ [4]. Le traceur a été irradié sous forme de BrNH$_4$ à la pile EL3 de Saclay, avec une activité spécifique de 0,16 mc/mg.

Les opérations ont été conduites sensiblement de la même manière pour les deux sondages. En premier lieu, on a procédé à un relevé du bruit de fond (fig. 9 et 10). Puis l'ensemble des injections a été mené avec des moyens de pompages assez puissants et selon la chronologie suivante:

1° Injection d'eau de mer inactive:
- 1000 l au sondage $S_4$,
- 2145 l au sondage $S_9$.

2° Injection d'eau de mer marquée au $^{82}$Br:
- 1770 l avec 10 mc de $^{82}$Br au sondage $S_4$,
- 2645 l avec 20 mc de $^{82}$Br au sondage $S_9$.

3° Injection d'eau de mer inactive:
- 940 l au sondage $S_4$,
- 320 l au sondage $S_9$.

4° Après remontée des tiges, pompage avec:
- débit moyen de 10,4 m$^3$/h (= 2,89 l/s) au sondage $S_4$,
- débit moyen de 9,5 m$^3$/h (= 2,63 l/s) au sondage $S_9$.

Le pompage a duré 1 h 38 au sondage $S_4$ et 2 h 26 au sondage $S_9$. La différence sensible de durée du pompage est due à la longueur différente des sondages.
PROFONDEUR (m)
DEUX UTILISATIONS DU BROME-82

Figure 10
Diagramme du sondage S9.

Modalités de l'injection
1. 2,145 m³ d'eau de mer en 2940 s, soit $Q_1 = 0.729$ l/s.
2. 2,645 m³ d'eau de mer + 20 mc de $^{82}$Br en 1350 s, soit $Q_2 = 1.959$ l/s.
3. 324 m³ d'eau de mer en 420 s, soit $Q_3 = 0.771$ l/s.
4. Pompe à débit $Q_4 = 2.63$ l/s.

Radioactivité mesurée au scintillateur
1. Bruit de fond avant l'injection, le 18 juillet 1962.
2. 3 h 20 après la fin de l'injection, 3 h 10 après le début du pompage.
3. 11 h 30 après la fin de l'injection, 11 h 20 après le début du pompage, 8 h après la fin du pompage.
4. 14 h après la fin de l'injection, 13 h 50 après le début du pompage, 10 h 30 après la fin du pompage.

Figure 11
Variation des niveaux d'eau dans les sondages S4 et S9 en fonction de la marée.
La mesure de l'activité a été réalisée, pour chaque sondage, avec le matériel suivant:
- deux scintillateurs type SPP3 SRAT (cristal \(1'' \times \frac{1}{2}''\)) dans les sondages,
- une sonde Geiger-Müller type GMT3T SRAT dans les sondages,
- un scintillateur PRI avec enregistrement sur le tuyau de refoulement de la pompe (cristal \(7'' \times 6''\)).

Certains appareils sont restés à la même profondeur pendant une partie de la durée des mesures. C'est le cas notamment du gammarmètre GMT3T qui au sondage S4 est resté 41 min au fond du sondage, à la profondeur de 24 m au début du pompage et pendant 18 min à la fin du pompage.

Les mesures ont été effectuées pendant deux heures consécutives au sondage S4, puis à deux autres reprises, 17 h et 17 h 30 après la fin de l'injection du traceur radioactif.

Au sondage S5, les mesures ont été effectuées sans maintien d'appareil à une profondeur fixe et se sont échelonnées pendant deux heures, avec deux à trois appareils, puis à trois reprises, huit heures et demie, neuf heures et treize heures après l'injection d'eau active et après l'arrêt du pompage.

4. Résultats

Les diagrammes des relevés de l'activité pour les deux sondages injectés sont représentés dans les figures 9 et 10. Les contrastes apparaissent d'une manière particulièrement nette. Remarquons tout d'abord que le bruit de fond, bien différencié, n'a pratiquement aucune influence sur les résultats. Nous examinerons successivement les résultats pour les deux sondages.

a) Sondage S4

La courbe 2 du diagramme constituant la figure 9 correspond à une activité mesurée 17 h après la fin de l'injection d'eau marquée.

*Terrains superficiels et récents, de 0 à 8,50 m*

Ces séries sableuses ne sont pas aussi homogènes que leur aspect en carottes de sondages pouvait le laisser prévoir. L'activité passe du simple au quintuple, de 3 à 8 m. Une tranche plus perméable apparaît nettement vers 7-7,50 m, à la base des sables jaunes et gris.

*Terrains jurassiques gréseux, calcaires et marneux*

Une première pointe apparaît vers 13 m. Elle correspond à un grès formé de coquilles de Lamellibranches, dont l'épaisseur n'est que de 0,30 m. Les marnes sus- et sous-jacentes montrent bien que la petite couche de grès est seule responsable de cette perméabilité.

Une deuxième pointe apparaît vers 20 m. Elle correspond à une série sableuse rendue perméable probablement par dissolution partielle du ciment de calcite.

Une troisième pointe, entre 22 et 23 m, correspond à des petites intercalations de lits sableux dans les marnes. Cette mise en évidence du
rôle prédominant de petites couches sableuses pour le drainage est d'une particulièr e importance.

Une quatrième zone active bien individualisée est située entre 25,5 et 26,5 m et correspond à un banc de grès très diaclasé, avec quelques passages marneux. Cette série a déjà été repérée au cours des mesures de résistivité comme provoquant une arrivée d'eau douce.

Bien que les travaux de terrassements ne descendent pas au-delà d'une douzaine de mètres de profondeur, il a paru intéressant d'effectuer des mesures dans le temps pour déterminer l'allure de la décroissance d'activité due au pompage*. Les mesures ont été localisées entre 14,50 et 24 m et ont été mesurées à l'aide d'un compteur G-M spécialement équipé (GMT3T). Les résultats indiqués dans la figure 12 correspondent à des mesures effectuées pendant et après le pompage. Les courbes mettent en évidence la décroissance de l'activité au droit des séries les plus perméables. Remarquons tout de suite que la mesure de l'activité 18 h environ après la fin de l'injection ne donne que trois valeurs nettement au-dessus de celles du bruit de fond.

On peut, en outre, faire les remarques suivantes:
- La décroissance de l'activité n'est pas directement liée à la valeur maximum initiale.

Ainsi, à 19 m, l'activité baisse de 100 à 48 cps en 43 min, alors qu'à 21 m, on passe dans le même temps de 80 à 60 cps.

A 16 m en 42 min, on passe de 50 à 20 cps, alors qu'à 18 m la variation, dans le même temps, est de 80 à 20 cps.

L'explication de ces types de variations nous paraît devoir tenir compte des principaux paramètres suivants:
- Influence de la perméabilité des couches injectées. Plus une couche est perméable, plus elle absorbe d'eau à l'injection, et à l'inverse, plus elle en restitue au pompage.

Ainsi, à 16 m, mais surtout à 19-19,50, à 21-21,50 et au-dessous de 24 m**, les couches perméables apparaissent nettement.
- Influence des pertes de charge sur la vitesse de restitution**. Ces pertes de charge sont dues aux caractéristiques granulométriques du matériau, ou à l'épaisseur des fissures.

Vers 16 m, les diaclases du calcaire doivent être fines, et par ailleurs, la perméabilité n'est pas très forte.

Par contre, dans la tranche 18,50-19,50, la forte perméabilité se manifeste par une forte absorption et une forte restitution de radioactivité, mais la circulation est facile dans ces grès sableux altérés et l'activité baisse rapidement. Ce caractère est encore plus net vers 18,50 m.

* * * * *

* Pendant un laps de temps court, de 2 à 3 h, nous avons admis que la décroissance de la radioactivité naturelle du $^{82}$Br pouvait être négligée.

** Mesure incomplète, l'appareil n'ayant pu descendre aussi bas que le scintillateur SPP3 du diagramme du $S_4$ (fig. 9).

*** Rappelons que l'injection de l'eau dans le sondage a utilisé une différence de pression nettement supérieure à la dépression créée lors du pompage, par rapport au niveau d'eau en équilibre avec les mouvements de marée.
Enfin, vers 21 m, l'absorption est forte au niveau des petits bancs sableux intercalés dans les marnes, mais la restitution est lente, due à la finesse de ces sables.

L'étude quantitative du phénomène est assez complexe, et fait actuellement l'objet d'études sur modèles.
- L'influence du mouvement des masses d'eau sous l'effet de la marée ne peut, pour l'instant, que faire l'objet de commentaires qualitatifs.

En effet, la résultante de ces mouvements conduit la masse d'eau activée au brome vers la mer, suivant en cela le mouvement dû au drainage naturel. Le va-et-vient doit donc donner une décroissance d'onde amortie (independamment de la décroissance de la radioactivité de l'élément).

La figure 11 montre que le décalage entre les niveaux d'eau dans les sondages et les résultats obtenus au marégraphe est relativement faible, la nappe contenue dans le terrain réagissant rapidement à l'effet de la marée.
Quoiqu'il en soit, ces mouvements alternatifs des masses d'eau ont eu pour résultat d'augmenter la dilution de l'eau marquée. Nous avons tenté d'évaluer la quantité de radioactivité récupérée ou récupérable au cours du pompage.

- La détermination de l'activité récupérée au cours du pompage a été basée sur la mesure de la radioactivité à l'aide d'un scintillateur placé contre la conduite de refoulement.

D'une manière générale on peut estimer que les courbes de récupération ont une forme logarithmique de période $T$, $T$ étant le temps au bout duquel la concentration $C$ de l'eau récupérée est la moitié de la concentration $C_0$ de l'eau récupérée au début du pompage. Cette concentration est de la forme

$$C = C_0 e^{-\frac{0.7t}{T}}.$$  

L'étalonnage de l'appareil fournit les concentrations $C_0$. La quantité récupérée pendant le temps compris entre $t$ et $t+dt$ est

$$dA = CQ dt,$$

$Q$ étant le débit pompé.

Si $t_f$ est le temps total de pompage, $A_{\text{pompe}} = C_0 Q \int_0^{t_f} e^{-\frac{0.7t}{T}} = \frac{C_0 Q T}{0.7} \left[ 1 - e^{-\frac{0.7 t_f}{T}} \right],$

$$A_\infty = \frac{C_0 Q T}{0.7},$$

$A_\infty$ correspondant à un temps de pompage infini.

Dans le cas du sondage 4, le calcul montre qu'à la suite d'une injection de 10 mc dans le sondage et une concentration $C_0 = 0.5$ c/l, $T = 30$ min pendant la 1ère période de pompage, et $T = 40$ min pour la 2ème période,

$$A_\infty$$ étant égal respectivement pour chacune de ces périodes à 3,6 et 3,2 mc.

On voit donc que, pour ces deux sections de courbe, la quantité récupérée pour un pompage de temps infini serait de l'ordre de 1/3 de l'activité injectée. Comme le pompage s'est effectué pendant 1 h 20, le rapport de la fraction récupérée réellement à celle qu'il était possible de récupérer pour un pompage de temps infini est

$$\frac{At_f}{A_\infty} = \left(1 - e^{-\frac{0.7 t_f}{T}}\right) \sim 0,90 \quad \left\{ \begin{array}{l} 0,87 \text{ pour } T = 40 \text{ min}, \\ 0,94 \text{ pour } T = 30 \text{ min}. \end{array} \right.$$  

b) Sondage $S_9$

En ce qui concerne le sondage 9, des calculs analogues nous conduisent à estimer qu'avec une injection de 20 mc, la fraction récupérée réellement
par rapport à ce qu'il était possible de récupérer pour un pompage de temps infini est de l'ordre de 0,97.

On voit donc que, dans l'ensemble, la dilution est relativement faible.

On peut en conclure, en première approximation, que les variations de niveaux dans les sondages ne correspondent pas à un mouvement tangentiel important des masses d'eau contenues dans le terrain. Dans le cas du sondage 9, la présence de matériaux sableux non cohérents est dans la ligne logique de cette constatation.

Le drainage vers la mer et la circulation d'eau douce dans certaines couches sont donc assez localisés et ne doivent pas correspondre à un phénomène généralisé sur toute l'épaisseur des terrains.

III. CONCLUSIONS

Dans de nombreux cas, l'utilisation de traceurs colorés tels que la fluorescéine, ou chimiques tels que le bichromate de potassium, peuvent donner de bons résultats dans une étude d'hydrodynamique souterraine.

Le dosage de l'un et l'autre pose toutefois des problèmes de concentration limite qui ont déjà été traités par ailleurs, notamment par Hours, et imposent des prélèvements qui alourdissent la méthode. D'autre part, il n'est pas toujours aisé de doser avec précision les valeurs intermédiaires entre la concentration moyenne injectée dans la nappe à proximité immédiate du point origine et la limite inférieure du dosage.

Or, dans la généralité des cas, ces valeurs différentielles ont une importance particulière car, compte tenu des inévitables hétérogénéités naturelles des séries géologiques, les variations dans le temps et dans l'espace des concentrations de traceur sont une indication sur la direction locale ou générale de la nappe.

Dans la recherche des passages perméables d'une série géologique, seuls des moyens physiques peuvent convenir, tels que la résistivité, avec, toutefois, une imprécision évidente.

Au cours des mesures exposées ici, l'analyse quantitative des résultats enregistrés permet une différenciation immédiate des vitesses de l'eau, de la direction et de la fraction intéressée par le pompage ainsi que de l'importance des venues d'eau dans une série géologique perméable.

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RÉFÉRENCES

DISCUSSION

R. HOURS (Chairman): One point that struck me about this investigation was the way in which the tracer technique was combined with conventional methods, current-meter measurements, and so on. That is something, I think, which we could all well imitate.

These investigations - and especially the first series of experiments - are a particularly good example of the value of a tracer like bromine that is not retained in the ground through which it passes. When one is trying to measure the amounts of the tracer recovered, this is naturally an essential requirement.

I also have one comment on the Boulogne experiment, which was carried out in sea water. The fact that bromine is naturally present in sea water obviously facilitated the circulation of the radioactive bromine. Experiments of this sort are regularly carried out in oil wells, but the present investigation is remarkable for the degree of precision obtained in the diagrams. This would clearly be difficult to achieve in experiments performed several hundreds of metres below the ground.

R. W. NELSON: What value did you adopt for the porosity with the tracer results in the first draw-down test?

P. C. LÉVÊQUE: The porosities varied as a function of the grain sizes measured in the different types of alluvions. Values from 40% to 60% were found. We took 50% as the average.

R. AMBROGGI: To revert for a moment to this question of microcurrent meter versus tracer techniques, it might have been useful to have prepared a diagram comparing the data obtained with the two methods.

P. C. LÉVÊQUE: It would certainly have been useful if we could have superimposed on the radioactivity graphs the permeability data we obtained on the basis of microcurrent measurements. The only reason we did not do this was that the reproduction of diagrams even more overloaded than the present ones would have raised severe practical difficulties.

R. AMBROGGI: Could you comment on the relative advantages of the two methods?

P. C. LÉVÊQUE: The microcurrent meter was useful as a means of indicating the presence of the more obvious permeable series. The microcurrent technique is a cumulative one, i.e. when a new permeable layer appears the number of turns is increased cumulatively. In other words, the microcurrent meter is useful above a certain lower limit of, say,
1. 5 cm/s and up to an upper limit which does not tell one a great deal. It cannot, however, pinpoint the permeable layers and therefore the inflows of water with the same degree of precision which we have attempted to achieve in our paper. As has already been emphasized, the two techniques complement one another extremely well.

R. AMBROGGI: I should like to say that I find this a particularly interesting paper from the point of view of its potential contribution to the study of the stratification of water levels in an underground reservoir.
FIELD AND LABORATORY TESTS OF CHROMIUM-51-EDTA AND TRITIUM WATER AS A DOUBLE TRACER FOR GROUNDWATER FLOW

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Abstract — Résumé — Аннотация — Resumen

FIELD AND LABORATORY TESTS OF CHROMIUM-51-EDTA AND TRITIUM WATER AS A DOUBLE TRACER FOR GROUNDWATER FLOW. Since 1958 field experiments and laboratory tests have been made in a study of groundwater flow in different geological and mineralogical environments by the use of gamma-emitting tracers and tritium water.

The velocity of groundwater flow in soil is rather low, and tracers with medium or long half-life must be chosen to trace the movement. A stable EDTA-complex of Cr$^{51}$ (half-life 28 d) was developed for this purpose and used together with tritium water. With this double tracer it was possible to follow the groundwater flow by measurement of the gamma radiation from Cr$^{51}$ directly in the field and thereby to reduce the number of water samples for precise laboratory assessment. By comparison of the measured activities of Cr$^{51}$ and tritium it was possible to determine whether there was any retardation or loss of the chromium complex as a result of adsorption.

Six field investigations, each of about two months' duration, have been made in glacifluvial sand and gravel. The results from these show that the chromium complex is transported as rapidly as the tritium water is, even at low concentrations (0.01 ppm) of the complex. 17 field investigations of one to three months' duration with this double tracer have been carried out in various till (moraine) soils for a study of certain hydrological problems.

Laboratory tests with soil and water from the various areas of field investigations have shown that the chromium complex does not hydrolyse at concentrations above 0.01 ppm. Further laboratory tests of the reliability of the chromium complex in different mineralogical environments are in progress.

A number of investigations of groundwater flow through fissures and channels have also been made. When the velocity of flow was assumed to be very high, Br$^{82}$ as bromide ion or Rhodamine-B, a fluorescent organic dye, were used. EDTA-Cr$^{51}$ and tritium water were, however, used when the velocity was considered low or when, as in karst, a great number of channels or large subterranean water magazines were presumed.

EXPÉRIENCES SUR LE TERRAIN ET EN LABORATOIRE À L'AIDE D'EAU TRITIÉE ET DE EDTA-51Cr POUR L'ÉTUDE DU MOUVEMENT DES EAUX SOUTERRAINES. Depuis 1958, on procède à des expériences sur le terrain et en laboratoire pour étudier le mouvement des eaux souterraines à l'aide d'indicateurs émetteurs gamma et d'eau tritiée dans différents milieux minéraux et géologiques.

Le déplacement de l'eau souterraine dans le sol étant assez lent, on doit utiliser des indicateurs à période moyenne ou longue pour le suivre. On a mis au point à cette fin un complexe EDTA-Cr$^{51}$ chimiquement stable (période 28 jours), que l'on utilise avec de l'eau tritiée. Grâce à cet «indicateur double», on peut suivre les déplacements de l'eau souterraine en mesurant le rayonnement gamma du Cr$^{51}$ directement sur le terrain, ce qui réduit le nombre d'échantillons d'eau nécessaires pour des mesures plus précises en laboratoire. En comparant les mesures de l'activité du Cr$^{51}$ et du tritium, on a pu déterminer s'il y avait un retard ou une perte du complexe au chrome par adsorption.

On a procédé à six expériences sur le terrain, chacune d'une durée de deux mois environ, dans des sables et graviers fluvioglaciaires. Les résultats montrent que l'indicateur au chrome est transporté aussi rapidement que l'eau tritiée, même lorsque sa concentration est faible (0,01 ppm). Dix-sept expériences de un à trois mois chacune ont été effectuées sur le terrain avec cet «indicateur double», dans des moraines de différentes catégories, pour l'étude de certains problèmes hydrologiques.
Les essais en laboratoire avec des sols et de l'eau provenant des divers terrains d'expérience ont montré que l'indicateur au chrome ne s'hidrolyse pas si la concentration est supérieure à 0,01 ppm. D'autres expériences se poursuivent en laboratoire sur différents milieux minéraux pour étudier jusqu'à quel point l'indicateur au chrome donne des résultats sûrs.

On a procédé également à plusieurs expériences sur l'écoulement de l'eau à travers des fissures et canaux souterrains. Lorsqu'on supposait la vitesse d'écoulement très élevée, on s'est servi de Cr51, sous forme d'ion bromure, ou de Rhodamine-B (colorant organique fluorescent); mais on a utilisé EDTA-Cr51 et de l'eau tritiée lorsque la vitesse supposée était faible, ou lorsque, comme dans des terrains karstiques, on soupçonnait l'existence d'un grand nombre de canaux ou de grands réservoirs souterrains.

**MEDICIÓN DEL FLUJO DE AGUAS SUBTERRÁNEAS EN LABORATORIO Y SOBRE EL TERRENO CON Cr51-EDTA Y AGUA TRITIADA COMO DOBLE INDICADOR. Desde 1958, los autores vienen estudiando en el laboratorio y en diversas formaciones geológicas y mineralógicas el flujo de aguas subterráneas empleando emisores gamma y agua tritilada.

La velocidad de circulación de las aguas subterráneas en el suelo es reducida, y para seguir su desplazamiento es preciso utilizar indicadores de período medio o largo. Con este objeto los autores prepararon un complejo del ácido etilendiaminotetra-acético (EDTA) y Cr51 (período: 28 d) y lo utilizaron junto con agua tritilada. Con este doble indicador, fue posible estudiar el flujo de las aguas subterráneas midiendo directamente sobre el terreno la radiación gamma del Cr51, lo que permitió reducir el número de muestras de agua que se han de analizar exactamente en el laboratorio. Comparando las actividades del Cr51 y del tritio medias, se pudo determinar si se producía o perdía del complejo de cromo por adsorción.

Los autores han realizado sobre el terreno 6 investigaciones de arenas y gravas de origen glaciofluvial, de unos dos meses de duración cada una. Los resultados muestran que el complejo de cromo no se hidróliza si se emplean concentraciones superiores a 0,01 ppm.
**Cr\textsuperscript{51}-EDTA AND HTO AS A DOUBLE TRACER**

Para establecer si el complejo de cromo da resultados fidedignos, se prosiguen los estudios en el laboratorio con diversas composiciones mineralógicas.

Los autores han realizado también varias investigaciones del flujo de aguas subterráneas por grietas y canales. Para velocidades de flujo muy elevadas, emplearon como indicadores \textsuperscript{82}Br, en forma de ion bromuro, o rodamina-B, un colorante orgánico fluorescente. El \textsuperscript{51}Cr-EDTA y el agua tritada fueron empleados cuando se consideraba que la velocidad de flujo era pequeña o cuando, como sucede en las formaciones kársticas, se presumía la existencia de un elevado número de canales o de grandes depósitos subterráneos de agua.

**PURPOSE OF THE INVESTIGATIONS**

Since 1958 field experiments and laboratory tests have been carried out in order to find — and establish the limitations of — a suitable gamma-emitting tracer for the study of groundwater flow in different geological and mineralogical environments. Tritium water has been used as a reference tracer in these experiments. The methodological and some hydrogeological results will be summarized here. Full reports of the hydrogeological results will be published elsewhere.

**SELECTION AND PREPARATION OF THE TRACER**

Being identical to water, tritium water (HTO) could be considered an ideal radioactive tracer. Its relative harmlessness and its low cost are further advantages. It is possible that the tracer may be retarded or lost by exchange of tritium between the tritiated water and the adsorbed water and the inter-layer water in clay minerals (montmorillonite). KAUFMANN and ORLOB \cite{1} found a retardation of tritium as compared with chloride in sandy loam; some tests in various sandstones \cite{2} showed no tritium retardation. The general opinion seems to be that the retardation of tritium is not significant in soils having a small content of clay and silt. If experiments have to be made in clayey soils, the retardation of tritium can be calculated (see ERIKSSON \cite{3}). Unfortunately HTO cannot be detected in the field at present because of its very soft beta radiation. For this reason it would be more convenient to use a gamma-emitting substance as a secondary tracer in order to reduce the number of samples which have to be measured or to replace tritium with a tracer measurable in the field. The tracer has to follow the moving water and must not be lost or retarded in such a way as to give misleading results. Due to adsorption and ion-exchange phenomena, cations are lost from, and retarded relative to, liquids flowing through porous media, especially when they contain clayey and/or organic colloids with particularly strong surface forces. Anions and neutral complexes are generally less strongly adsorbed. Of simple anions, only the three halogen ions Cl\textsuperscript{-}, I\textsuperscript{-} and Br\textsuperscript{-} can be considered for use as radioactive tracers in groundwater. Chloride ion (Cl\textsuperscript{36}) has proved \cite{1, 4} to be an ideal tracer as it does not participate in any exchange in the ground. Unfortunately, it has only limited practical usefulness because of its very high cost, the absence of gamma radiation and the extremely long half-life (3×10\textsuperscript{5} yr). Iodide ion (I\textsuperscript{131}) has been successfully used by several investigators \cite{4, 5}, especially in pumping tests in oil wells and for the checking of the efficiency.
of water-flooding operations [6, 7, 8]. However, for field studies of undisturbed ground water flow and large-scale investigations, the half-life of $^{131}\text{I}$ (8 d) is too short. $^{131}\text{I}$ has another disadvantage: its maximum permissible concentration in water is very low. In our investigations $^{131}\text{I}$ was chosen for some preparatory field experiments. Bromide ion (Br$^{82}$ with a half-life of only 36 h) has turned out to be a good tracer [5] and was selected in the present investigations when very fast groundwater flow in fissures and channels was expected.

To obtain suitable tracers with medium half-life we must therefore resort to the use of stable complexes. Such compounds can be found among the so-called metallic chelates. These chelates are composed of a metal atom closely bound to an organic molecule. The chelate may be neutral or charged. Many chelating agents are known; and since there are several metals which have radioactive isotopes with half-lives reckoned in weeks or months, the number of possible chelated tracers is very large. The best-known chelating agent is ethylene diamine tetraacetic acid, EDTA. Examples of useful complexes are EDTA-chelates of In$^{145m}$, Cr$^{51}$, Sc$^{46}$ and Sb$^{124}$.

Of the isotopes mentioned, Cr$^{51}$ possesses several favourable properties. Its most attractive characteristics are the absence of beta-radiation, the low energy of the gamma radiation and the consequently high permissible concentration level in drinking water. The soft gamma radiation permits good localization of the tracer. The disadvantages are that only about 8% of the disintegrations give rise to gamma radiation and that the cross-section for the formation of Cr$^{51}$ by irradiation of chromium is relatively small, resulting in low specific activity. With a thermal-neutron flux of somewhat more than $10^{14}$ n/cm$^2$ sec which is at present available in Sweden, the lowest chromium concentration at which the Cr$^{51}$ activity will still be readily detectable is 0.1 part per $10^9$ with our present counting procedure. This means that the use of the chromium complex as a groundwater tracer will be restricted to moderately sized projects even if the stability of the complex could be established at even lower levels than 0.1 part per $10^9$.

The stability constant of the Cr$^{51}$-EDTA-complex is of the order of $10^{24}$. The complex has a characteristic violet colour and is present in acid or neutral solutions as a monovalent ion. At higher pH it is slowly transformed into a divalent form of blue colour. It has been shown that the formation and decomposition of the complex is slow in contrast to those of most other EDTA complexes. The formation has been studied in detail by HAMM [9].

For the experiments reported here, the Cr$^{51}$-EDTA complex was chosen. The work with the Cr$^{51}$-complex was inspired by the successful results reported by LACEY and de LAGUNA [10]. Later investigators have, however, been of the opinion that the chromium complex is inferior to other complexes [2, 11]. This will be discussed later in this text. Cr$^{51}$ has a half-life of 28 d, and this is long enough for many groundwater investigations. The half-life is, on the other hand, short enough to make the Cr$^{51}$ activity decay to non-significant levels in a reasonable time before the tritium assays are carried out.

The complex was made as follows: Analytical-grade chromium metal

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* The terms part per $10^9$ and ppm are used for the concentrations of chromium, 1 part per $10^9$ meaning 1 mg of Cr per m$^3$ of water and 1 ppm (part per million) meaning 1 g of Cr per m$^3$ of water.
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was irradiated to the specific and total activity needed for the investigations. It was then dissolved in an excess of 4-N hydrochloric acid, and the solution was filtered. A clear solution of analytical grade EDTA, dissolved in the presence of sodium hydroxide, was added to this solution in an amount of 10 g EDTA/g of chromium. The reaction took place in two stages, the first giving rise to an intense green colour which changed into characteristic violet. The solution was kept at a pH lower than 3 for at least 15 min after the violet colour had appeared. After the solution had cooled and the pH had been adjusted to that of the groundwater to be studied (generally from 6.5 - 7.5), HTO was added from a concentrated stock solution.

Tritium water was selected as a reference tracer for our investigations. Together with Cr\textsuperscript{51}-EDTA, it constitutes a double tracer. With this it was possible to follow the groundwater flow by measuring the gamma radiation from Cr\textsuperscript{51} directly in the field, thus reducing the number of water samples necessary for laboratory assessment. By comparison of the measured activities of Cr\textsuperscript{51} and tritium it was possible to determine whether there was any retardation or loss of the chromium complex in relation to tritium water. Field experiments were made in glaciofluvial sand and gravel, in various till soils, in organic soils and in karst. Laboratory tests were performed with soils from the areas investigated as well as from some other mineralogical environments.

ACTIVITY MEASUREMENTS AND INSTRUMENTATION

The field measurements of the gamma radiation were carried out by means of scintillation counters. In the first series of experiments in 1958, two mains-operated counters, connected to recorders, were used. The counters comprised Atomic Instrument Co. type 410 counting-rate meters and water-tight scintillation probes with 1 in X 1 in thallium-activated sodium-iodide crystals. Esterline-Angus Co. recorders were used. The detector probes were lowered into the observation wells and the activity measured at various times during the experiment. With this technique the counter measures the activity of the water within the well and of the groundwater near the well, since the half thickness of the Cr\textsuperscript{51} 0.32 MeV gamma radiation is about 6 cm in water and somewhat less (4 - 6 cm) in groundwater. The inner diameter of the observation well tubes was 5 cm and the wall thickness 0.3 cm.

It was experienced, however, that mains-supplied instruments were less suited for field measurements since it was generally difficult to arrange a power supply. This type of instrument is also, as a rule, heavy and bulky and thus not easily portable in the field.

For this reason all field measurements from 1959 on were carried out with portable transistorized scintillation counters. These instruments were manufactured by "Laboratorium Prof. Dr. Berthold" and were of the types LgS/B and LgS/C. The detector probes were equipped with 1 in X 1 in NaI crystals. These instruments provide a 25 \(\mu\)A output for full-scale deflection, which could be registered on spring-driven Esterline-Angus 1 mA recorders with a single transistor preamplifier.
The radiation doses to the personnel handling the concentrated radioactive solutions in the field were checked by means of dose-rate meters, pocket dose meters and film badges. The risk of inhalation of tritium water vapour was practically negligible since all handling outside the preparation laboratory was carried out in the open air.

The laboratory measurements of \( \text{Cr}^{51} \) were made by means of a well-type scintillation counter with a background of 40 counts/sec and a \( \text{Cr}^{51} \) counting efficiency of 45% for 5 ml samples (related to the emitted gamma radiation).

The tritium measurements were made with an Ekco type N 664 liquid scintillation counter with a background of 25 counts/sec and a \( \text{H}^3 \) counting efficiency of 43% for a 0.5 ml water content in the scintillator.

The standard deviations of all activity values measured in the laboratory have been calculated from the counting statistics. In the diagram texts the detection limits for \( \text{Cr}^{51} \) and tritium have been given. The detection limit has been defined in this paper as the standard deviation of the background measurement.

FIELD EXPERIMENTS

Glaciofluvial soils

The first series of experiments was carried out in the Nybro esker, "Nybroäsen", at Vassmolösa, 20 km southwest of Kalmar, in 1958. The grains of the sandy soil (Fig. 1) consist of granite, quartz-porphyry, sandstone and a minor portion of limestone. The concentration of salts in the groundwater is low; its pH value is about 6.5 and its temperature 8-9°C. The hydraulic gradient was measured to be about 1:1000. 20 perforated tubes, hereafter referred to as wells, were driven into the ground by wash-boring so as to form a sector in the general direction of the groundwater flow. A series of preparatory experiments was made with \( \text{I}^{131} \) as iodide ion in order to get an idea of the velocity for the planning of the main experiment. As the preliminary value of the velocity was only 0.1 m/d, \( \text{I}^{131} \) was here found to have a somewhat short half-life. \( \text{Cr}^{51} \)-EDTA and HTO (200 and 100 mc respectively) were injected with a squirt at one point. Unfortunately, the radioactive water was detected only in two wells. Measurements after digging showed that this resulted from the fact that the radioactive water was confined to a very limited band. This agrees with the theoretical argumentation presented by SKIBITZKE [12]. It was possible to plot curves of \( \text{Cr}^{51} \) and tritium activities only for one well. They are identical in shape but differ in dilution [13]. The flow velocity was the same as that determined with \( \text{I}^{131} \).

Another experiment was made in a flat part of the Nybro esker at Gårsdryd, 4 km southeast of Nybro, in 1959. A thin bed of boulder-rich, well-graded soil overlay homogenous glaciofluvial gravel (Fig. 1) composed of granitic and quartz-porphyric grains. The groundwater had pH values of 6.2-6.9 and a temperature of 8-10°C. 36 wells were placed in a sector in the general direction of the groundwater flow. In order to ascertain the interception of the band of radioactive water everywhere a ditch, longer...
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Mechanical composition of representative soils of the field areas

a. Glaciofluvial sand from Vassmolnsa
b. Glaciofluvial gravel from Gårdbyd

c. Fine-sandy till from Kryseboda

than the shortest distance between two wells, was dug upstream of the well area. A solution (365 ml) of Cr\textsuperscript{51} -EDTA (500 mc) and HTO (1 c) was mixed with the water (80 l) in the ditch, which was then cautiously filled up. Because of a severe drought the groundwater flow changed its direction out of the well area soon after the injection. In order to secure experimental results pumping was started in a well on the opposite side of the area. For 40 d the active band flowed through the observed area. All the time measurements were made directly in the field by means of scintillation counters, and water samples were taken three to four times a day. 700 samples were measured for Cr\textsuperscript{51} activity and 300 for tritium activity in the laboratory. For 14 wells complete curves of both Cr\textsuperscript{51} and tritium activities could be plotted. The curves for the 11 wells, which were first passed, are mostly identical in shape as well as in dilution (Fig. 2). After a transport of 11 m and further Cr\textsuperscript{51} -EDTA is retarded several hours and adsorbed to some extent. This occurs at a concentration below 0.01 ppm. The longitudinal dispersion can be followed very well in the curves (Fig. 2). For HTO it is certainly only hydraulic dispersion, but for Cr\textsuperscript{51} -EDTA there must be dispersion resulting from interaction in the ground after a transport of 11 m.

For a study of the undisturbed groundwater flow of the area at Gårdbyd a new investigation was made in 1960. The well area was altered and enlarged with 30 new wells (Fig. 3). The solution of Cr\textsuperscript{51} -EDTA and HTO was injected into three wells by means of a pressure squirt. These wells were situated at a distance of 0.2 m from each other, perpendicular to the flow direction. Thus a band of radioactive water broad enough not to escape the wells was formed. Measurements and sampling took place at intervals of 3 - 4 h at the beginning. Later on the intervals were prolonged. When the radioactive water had flowed as a limited band for 7.5 m through the well area, the direction of groundwater flow changed because of heavy rains, and the active band flowed out of the area through a gap between two wells.
Concentrations of Cr$^{51}$-EDTA and HTO in relation to injected solution in wells at Gardsryd in 1959.

The wells were situated in the central zone of the band of radioactive water at distances of 2, 5 and 11 m, respectively, from the injection point.

The detection limit of Cr$^{51}$-EDTA was $0.3 \times 10^{-4}$ and that of HTO $0.03 \times 10^{-4}$.

- Cr$^{51}$-EDTA
- HTO

Fig. 2

Map of the well area at Gardsryd in 1960

60. = Well No. 60

Fig. 3
Concentrations of Cr\textsuperscript{51}-EDTA and HTO in relation to the injected solution in well No. 9. The fluctuations of the curves result from the great variations in the hydraulic gradient.

The detection limit of Cr\textsuperscript{51}-EDTA was 0.03 \( \times 10^{-4} \) and that of HTO 0.001 \( \times 10^{-4} \).

The results from 19 wells confirm the identical transport of Cr\textsuperscript{51}-EDTA and HTO in general (Fig. 4). Yet at the lowest concentration of the Cr\textsuperscript{51}-EDTA complex — 0.05 - 0.2 ppm — no hydrolysis of the complex with an accompanying loss by sorption could be observed. An exceptional result is that from well No. 20, where the front of Cr\textsuperscript{51}-EDTA has a higher concentration than that of tritium. As this is the only example of retardation of tritium, and as the curve is plotted from only three measurements, it is no evidence that tritium is retarded in comparison with a salt solution. The velocity of the front of the undisturbed groundwater flow along the central transport zone was, on the average, 0.6 m/d with a maximum velocity of 0.8 m/d. The differences resulted mainly from the great variations in the hydraulic gradient (1 : 400 - 1 : 2600) but also from small heterogeneities of the hydraulic conductivity. The band of radioactive water reached its maximum breadth — about three times the breadth of the injection zone — after a flow of 3 m, and then the breadth was constant or sometimes even decreased (cf. Skibitzke [12] and the first of our experiments). These results do not agree with the opinion that a tracer solution forms a cone opening outward from the injection point in the flow direction. But the velocity of the groundwater flow decreased more and more laterally from the central transport zone, which seems to be a result of lateral dispersion. The longitudinal dispersion is somewhat greater than in the earlier experiment, because of differences in injection technique and differences between non-uniform and slow, undisturbed groundwater flow and a more uniform and rapid flow established by pumping.

In the well area at Gärdsryd experiments have also been made for comparison of the suitability of radioactive isotopes with that of organic dyes as tracers. Cr\textsuperscript{51}-EDTA, HTO and Lissamine, and later also Rhodamine B, were injected into the groundwater. The dyes were in general adsorbed after a transport of 2 m but not the radioactive tracers. A further disadvantage of these fluorescent dyes is that they cannot be measured in the field as there is no portable battery-operated spectrophotometer available.

Two experiments of a more practical nature have been carried out in glaciofluvial soils. In the well area at Gärdsryd an attempt was made to
determine if river water and groundwater were isolated from each other. A solution of Cr$^{51}$-EDTA was injected into a well on the north side of the river (Fig. 3). On the opposite side pumping at 35 l/min was started. Measurement and sampling of the river water and of the groundwater in wells both on the sides and in the river were made. The radioactive water was injected partly into gyttja, which surrounded the river, and partly into glaciofluvial gravel. The flow was very slow in the tight organic soil but rapid in the gravel. After one day the tracer was detected in a well in the middle of the river but not in the river water, and one day later the radioactive water had reached the pumping well. It had been transported in the gravel under the tight bottom soil of the river. In another experiment at Lake Rostock, "Rostockasjôn", 2km north of the village of Lindås, the object was to determine if the lake water was infiltrated into the groundwater of an esker, "Lindåsen". A solution of Cr$^{51}$-EDTA, HTO and Lissamine was poured out in a little dam at the shore of the lake. The vegetation of the shore and on the bottom was not disturbed. After fourteen days low activities but no dyes were detected at a distance of 10 m in the groundwater of the esker.

These two experiments illustrate the suitability of Cr$^{51}$-EDTA as a tracer for practical investigations of groundwater supply.

Till soils

Groundwater flow in till (moraine) soils is more complicated than in glaciofluvial soils. Experimental work offers many difficulties. All boring is very troublesome and time-consuming as the soil is often very boulder-rich and hard. The groundwater flow is sometimes confined to lenses of coarser material; and when a series of wells is placed in an area, the flow might escape the wells.

17 experiments with radioactive tracers have hitherto (1960 - 1962) been performed. The most extensive experiment is the only one with a series of wells. In a till pit at Krysseboda, 1 km north of the village of Emmaboda, 30 wells were placed. The soil consisted of hard, fine-sandy till (Fig. 1) with some lenses of coarser material. The composition of the soil was granitic, dioritic and quartz-porphyric. The groundwater had a pH value of 6.2, and its temperature was 8 - 9°C. The gradient varied considerably from point to point but was on an average 1: 60. A tracer solution of Cr$^{51}$-EDTA, HTO and Lissamine was injected into three wells. Between those and well No. 2 a rapid flow (0.8 m/d) took place in the contact zone between a very large boulder and the till and thereafter in a lens of sandy gravel. The average velocity was much lower (0.2 m/d). In this case the flow took place in the normal soil of the area, the fine-sandy till, which was very tight. 40 d after the injection, low activities were measured downstream at a distance of 30 m from the injection wells, which represents an average velocity of 0.6 m/d. This means that a rapid flow had taken place in a lens or in a bed of coarser material. The concentration curves for Cr$^{51}$-EDTA and HTO clearly indicate the two types of flow. Another remarkable thing is that in well No. 3 Cr$^{51}$-EDTA was retarded 1 d and had been adsorbed to some extent (Fig. 5). This phenomenon becomes more significant in wells
Concentrations of \( \text{Cr}^{51} \)-EDTA and HTO in relation to the injected solution in well No. 3 at Krysseboda

The detection limit of \( \text{Cr}^{51} \)-EDTA was \( 0.03 \times 10^{-2} \) and that of HTO \( 0.00008 \times 10^{-2} \).

--- \( \text{Cr}^{51} \)-EDTA
--- HTO

Nos. 6 and 7, but in wells Nos. 8 and 12 it is rather the contrary. The dye was adsorbed after a transport of 2 m.

Two other experiments were performed in the pit at Krysseboda. The subterranean communication between two small pools was determined by means of \( \text{Cr}^{51} \)-EDTA and HTO. In another part of the pit the groundwater flow could be followed by measurements from the surface as the groundwater table lay at a depth of only 0.2 - 0.5 m. The hydraulic gradient was about 1 : 30. A winding band (0.5 - 1.0 m) of radioactive water could be detected to a length of 10 m with an average velocity of 0.45 m/d. When the principal part of the band had passed a point, a small hollow was dug and water samples were taken for control measurements in the laboratory. Four other experiments of similar character have been made with great success. For comparison of the groundwater flow conditions of various morainal topographies, the method of MOSER et al. [14] has been used with \( \text{Cr}^{51} \)-EDTA and HTO as tracers. Seven experiments were carried out last summer in a region of ablation moraine at Knappsmåla, 1.5 km southwest of Lindås and three experiments in a drumlin region at Rörbo, 3 km southwest of Emmaboda. The preliminary results show that the method gives good re-
lative data concerning percolation in different till soils. When the final results are available, a comparison of the capacities obtained from these experiments with those of the pumping tests can be made.

All experiments show that Cr$^{51}$-EDTA is a suitable tracer for the study of the complicated paths of groundwater flow in till and for the measurement of the velocity of groundwater flow over short distances.

Organic soils

Apart from the experiment mentioned above in gyttja, only one experiment has been made in organic soils. A raised peat bog, "Torståmåla fly", 3 km northeast of the village of Algutsboda, is drained at the edge by a brook, which suddenly disappears into a hole in the peat. 200 m away at another part of the edge there is a number of springs. The subterranean communication between the brook and the springs was determined by means of Br$^{82}$ as bromide ion. The transport time was just about 1 h.

Karst

The first series of experiments was performed in 1962 at Björkliden in Lappland, where a karst topography in miniature has been formed in hard limestone and slate. The water of the region had a pH value of 7.1 - 8.7. 18 small experiments in various subterranean brooks (0.5 - 65 l/min) were made with Br$^{82}$ as bromide ion and Rhodamine B. The flow velocities were very high (5 - 10 m/min) as the channels were narrow and relatively straight and the hydraulic gradient large (1 : 10 - 1 : 20). The communication between a lake with a subsurface outflow and four springs (230 l/sec together) was determined with Cr$^{51}$-EDTA, HTO and Rhodamine B as tracers.

The next experiment was made in the river Bjurälven (5 m$^3$/sec) in Jämtland, which has a subterranean course of about 2 km in very hard limestone. The flow was followed with Cr$^{51}$-EDTA, HTO and Rhodamine B. The dilution was very considerable, but Cr$^{51}$-EDTA could be measured both in the field and in the laboratory in significant concentrations. The dye could not be detected. The flow velocity was, on an average, 10 m/min.

In the loose limestone of the island of Gotland several subterranean water courses have been formed. The biggest one is the river of Lummelunda. Upstream of the suburface part the water capacity is very variable: 0 - 6000 l/sec. Downstream of this area the capacity is more even. The river disappears into six large sinkholes and many small ones. After a subterranean course of 1.3 km the river reappears in a cave. The hydraulic gradient is about 1 : 65. The water has pH values of 7.4 - 7.9 and a temperature of 9.0 - 15.0°C. A preparatory experiment was made from an inner cave to the out-flow cave (80 m) with Br$^{82}$ as bromide ion and Rhodamine B. In the main experiment a solution of Cr$^{51}$-EDTA (250 mc), HTO (2.5 c) and Rhodamine B (95 g) was poured into the largest sinkhole in use. After 3 d Cr$^{51}$ activities were measured in the inner cave, but no dye was detected. The laboratory measurements show that the dilution was 1 : 6.5 millions. This, together with the relatively low velocity (16 m/h), means that there are large subterranean water magazines in the loose limestone at Lummelunda.
The experiences from the investigations in karst are that Br82 as bromide ion can be used when the velocity of flow is assumed to be very high, but in other cases Cr51-EDTA and HTO are preferable.

LABORATORY TESTS

The purpose of these tests is to establish the usefulness of gamma-emitting tracers for the study of groundwater flow. A series of careful investigations in the laboratory was planned so as to give the lowest concentration at which Cr51-EDTA is still not retarded or adsorbed in soils from the investigated areas and in materials of various other mineralogical compositions. Other gamma-emitting nuclides in suitable chelated form will also be tested. In addition, it might be of interest to study other complexes in laboratory investigations. Tritium water has been and will also be used in future tests with the gamma-emitting tracers as a reference. During the spring of 1961 laboratory equipment consisting of a column (500 mm × 60 mm diameter), a micropump (maximum capacity: 43 ml/h) and an automatic sampler (Fig. 6) was built. The column was packed with sand; then water was pumped through it for several hours, followed by tracer solution for 24 h, and then water was again pumped through the column for 24 h. Water samples were taken once every hour.

In the first investigations, which were performed with Cr51 of high specific activity (12 c/g) purchased from a commercial producer of radio-isotopes, the complex compound behaved very unsatisfactorily. The concentrations of Cr51 recovered were very low and completely independent of the amount of chromium carrier in the entering solutions. The maximum recovery was for 1 ppm = 25.7%, 0.1 ppm = 15.0%, 0.01 ppm = 18.7% and 0.001 ppm = 20.9%. As there were reasons to doubt these results, the tests were repeated after a very cautious complex preparation with irradiated chromium metal *. The explanation of the result of the first series must then be that chelation had not been completed. This may give an interpretation of the results published by the American team (HEEMSTRA et al. [2]). In their investigation 11 chemical forms of 9 radioisotopes, including the Cr51-EDTA complex, were tested in a laboratory set-up; and while, for instance, the EDTA-complexes of Sb124, Co60 and Ir192 were transported through the column without retardation as compared with HTO, the Cr51 complex was considerably retarded and did not reach more than 61% of the entering concentration. Upon recirculation through the column, considerable amounts of all tested substances except tritium and the Sb124 complex were gradually sorbed. The Cr51 complex, however, showed the surprising property of not being sorbed any further after recirculation despite the great initial loss. The Cr51 complex was rejected as not suitable. If in their tests the chelation had been unsatisfactory, the initial loss and the absence of any further sorption would be explained. We should consider Cr51-EDTA, if this explanation holds true, a good water tracer, judging from their results.

The investigations were repeated with soils and water from the area at Gårdsryd to determine the lowest concentration at which the Cr51 complex

* In these experiments a complete recovery of the tracer was obtained.
is still reliable as a tracer in such soils. At the lowest concentration tested, 0.01 ppm, the plateau of the recovery curve lay at about 97% of the entering solution, which must be considered very satisfactory. Compared with HTO, the complex was not retarded. Consequently, Cr$^{51}$-EDTA is a completely reliable tracer down to 0.01 ppm in glaciofluvial sand and gravel consisting of granitic and porphyric grains; this was also established in the field experiments.

In another series of investigations Cr$^{51}$-EDTA was tested in pure quartz sand (99.7% SiO$_2$) and groundwater with a pH value of about 7.0. At the concentration tested (0.52 ppm and 0.02 ppm) the plateau of the recovery curve lay at 99% of the entering solution (Fig. 7). With a Cr$^{51}$-preparation of very high specific activity (10 c/g) tests at concentrations of 0.1, 1, 10 and 100 parts per 10$^9$ are being carried out. The above-mentioned sand (medium
sand) and groundwater will be used as standard components. The quartz sand will then be mixed with about 15 various fine-ground minerals (among others, orthoclase, plagioclase, muscovite, biotite, hornblende, augite, calcite, kaolinite, montmorillonite and illite) in the first series of runs. These investigations are at present in progress.

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DISCUSSION

R. HOURS (Chairman): Our Swedish colleagues are to be very much congratulated on this extremely interesting paper. They have shown admirable perseverance in studying a problem of considerable practical importance — I think we are all very keenly aware of the need for medium-life gamma emitters — and they have been very fortunate in having selected the right tracer. Work on other EDTA complexes has not been so successful.

The possibility of using these EDTA complexes was first suggested by LACEY and de LAGUNA [10]. However, their experiments were only carried out on a laboratory scale, and their investigations were confined to concentrations of several dozen ppm. The Swedish team has carried out field experiments and laboratory tests at concentrations of a few ppb and investigations are now in progress with concentrations as low as 0.1 ppb. In water the chelates are in a state of equilibrium and their stability can be maintained on the whole only in limited conditions of pH and concentration.

Work has also been done on these complexes by Dr. Watkins' team. But these investigations have been mainly concerned with the highly saline solutions found in oilwells and cannot be automatically extrapolated to fresh water conditions.

The comparison between the behaviour of this complex and tritiated water in field tests is of course particularly valuable. I think it would be extremely interesting to have data on similar experiments performed with chromium on the one hand and cobalt on the other.

Thanks to the work of our Swedish colleagues we now have a fairly clear idea of the potentialities of Cr\(^{51}\) in hydrology, which is certainly more than can be said about many other radiotracers. And since these investigations are continuing, we can look forward to more data in the future.

R. AMBROGGI: Could you give us some indication, on the basis of the first seven experiments you carried out, of the relative merits of Cr\(^{51}\)-EDTA and tritiated water?

G. KNUTSSON: Our general conclusion is that, since the transport of the Cr\(^{51}\)-EDTA complex and the tritiated water is identical in sand and granitic gravel down to a concentration of 0.01 ppm, it is possible to use the chromium complex together with tritiated water as a secondary tracer which can be measured in the field. This reduces the number of tritium samples that need to be measured in the laboratory. In medium-scale investigations the chromium complex can also be used on its own.

D. B. SMITH: In your flow investigation in glaciofluvial soils, your results show that the tracer flow was fairly parallel and did not diverge as is generally found to be the case in this type of investigation. Do you think that this result could indicate that the water flow in this region converges into an underground stream which restricts the lateral dispersion of the tracer?

G. KNUTSSON: After flowing for about three miles the band of radioactive water developed until it was three times as broad as that of the injection zone. After that the breadth remained constant. This is in good agreement with what was postulated by Skibitzke in 1958 and also with our own experience of other investigations in glaciofluvial soils. Furthermore, continuous measurements of groundwater levels over a large area which
included the area of investigation have not revealed any convergence of the streamlines in this area.

R. W. NELSON: In connection with this problem of streamline convergence, I think that Mr. Knutsson and his colleagues are to be complimented on the way they were able to introduce the tracer in this investigation without causing any major distortion of the natural streamline pattern. In my experience, the wide lateral spreading is generally associated with the distortion of the natural flow pattern that takes place when the fluid containing the tracer is introduced. The extent of this disturbance is described by Skibitzke (see reference [12]) and can be very important. The amount of lateral spreading mentioned by Dr. Knutsson is what one would expect from true lateral hydrodynamic dispersion for the rather short travel distances involved.
INVESTIGATION OF THE MOVEMENT OF INFILTRATING ACIDIC HOT-SPRING WATER IN THE GROUND BY MEANS OF RADIOISOTOPES

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Abstract — Résumé — Аннотация — Resumen

INVESTIGATION OF THE MOVEMENT OF INFILTRATING ACIDIC HOT-SPRING WATER IN THE GROUND BY MEANS OF RADIOISOTOPES. Tamagawa hot spring in Akita Prefecture gushes hot water of 1 pH hydrochloric acid at some 140 l/s. This quantity flows into the River Tama with the result that the river has been hindered from any significant hydropower development thus far. For the purpose of mitigating the acidity of the water river, the hot spring water is carried away through channels and infiltrated into the soil of a mountainside for chemical neutralizing through seepage before flowing into the river.

To select the sites where the water can infiltrate and to determine its distribution, the authors studied, with the aid of radioisotope tracers, the capacity and ability for neutralization by mountain soil and the mechanism of the process.

They carried out a total of nine experiments at the site from 1956 to 1960. Such radioisotopes as $^{131}$I, $^{32}$P, $^{86}$Rb, $^{60}$Co and $^3$H as tracers were poured into holes with carriers. The radioactivity of the water appeared in fissures of the river bed located over 200 m away from the input holes; this was measured and the movement of underground water was analysed by using the time variation of radioactivity obtained.

As a result, it was found that the radioactivity increased considerably at sporadic intervals, that several holes had connections to one fissure for water, that the creeping time ranged from 6 to 82 h, that the radioactivity found was less than the input, that behaviours were different among the nuclides applied and that the aging of the underground water course was noticeable.

ÉTUDE, A L'AIDE DE RADIOINDICATEURS, DU MOUVEMENT D'INFILTRATION DANS LE SOL DE L'EAU. La source Tamagawa située dans la préfecture d'Akita débite par seconde 140 l environ d'une solution chaude d'acide chlorhydrique d'un pH égal à 1; cette eau alimente la rivière Tama si bien que, jusqu'à maintenant, celle-ci n'a pu guère être aménagée pour la production d'énergie électrique. Afin d'atténuer l'acidité des eaux de la Tama, on a détourné l'eau de la source chaude au moyen de canaux de telle sorte qu'elle pénètre dans le versant d'une montagne où elle subit une neutralisation chimique en filtrant à travers les terrains, avant de rejoindre le cours d'eau.

Pour choisir les endroits où l'eau devait s'infiltrer et déterminer sa distribution, les auteurs ont étudié, à l'aide de différents radioindicateurs, la capacité de neutralisation des terrains montagneux et le mécanisme du processus.

Ils ont fait au total neuf expériences sur les lieux, de 1956 à 1960. Après avoir injecté dans des puits des radioisotopes tels que $^{131}$I, $^{32}$P, $^{86}$Rb, $^{60}$Co et $^3$H, accompagnés d'entraineurs, ils ont mesuré la radioactivité de l'eau qui jaillissait de fissures du lit de la rivière, situées à plus de 200 m des orifices d'injection et analysé le mouvement des eaux souterraines d'après les variations de la radioactivité dans le temps.

Au cours de ces expériences les auteurs ont constaté que la radioactivité accusait de fortes augmentations sporadiques, que plusieurs orifices étaient en communication avec une seule fissure, que la durée du cheminement de l'eau variait entre 6 et 82 h, que la radioactivité mesurée était moindre que la quantité introduite dans le puits, que les nucléides utilisés avaient des comportements différents et que le vieillissement du cours d'eau souterrain est notable.

ИССЛЕДОВАНИЕ ДВИЖЕНИЯ КИСЛОЙ ВОДЫ ГОРЯЧИХ ИСТОЧНИКОВ ПОСРЕДСТВОМ РАДИОИЗОТОПОВ. Вода горячего фонтанирующего источника Тамагава в префектуре Акита, имеющего дебит приблизительно 140 д/с и содержащего кислоту порядка 1 pH, попадает в реку Тама. В связи с этим на реке нельзя было до сих пор отстроить какие-либо значительные гидроэнергетические сооружения. В целях уменьшения кислотности воды в реке горячая вода отводится в канал и инфильтрируется в почву у подножья горы с

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The Tamagawa hot spring is situated on the mountainside of a volcano, Yakeyama, 765 m above sea level, in the eastern part of Akita Prefecture, Japan. This spring has a yield of 140 l/sec with water of pH 1.0 and water temperature of 98°C. This water flows into the Tama River, and at 70 km below the discharge point its pH value becomes approximately 4.1. Owing to its low pH value, aquatic life cannot exist in it, and the river water is considered toxic. Countermeasures for the improvement of the quality of the water have been a problem for a hundred years. Several decades ago it became clear that the toxicity of this water is caused by the hydrochloric acid flowing into the river from the hot spring. The low pH value of the river water has deterred the economic development of the valley. An artificial filtering method for the neutralization of the low pH value of the hot spring water by reaction with soil or rock has been carried out for some time.

For effective use of this method it is necessary to obtain information on the stability of the neutralization, selection of a suitable site, distribution of filtered water and capacity and chemical mechanism of this method. For these reasons the authors have used nine times from 1956 to 1960 radio-isotopes as a tracer to analyse the behaviour of the infiltrating water.
first, second and third of these nine experiments have been reported previously [1, 2]. Furthermore, a part of this report has already been presented at the third Isotope Conference [3]. In this paper the authors intend to review briefly the previous reports and to describe the results obtained from the latest experiments, Nos. 4-6.

II. THE NEUTRALIZING METHOD USED AT THE TAMAGAWA HOT SPRING

The hot-spring water of the Tamagawa hot spring flows 300 m down from the source and is directed into the dual channel. This channel has a total length of 1230 m, and from this channel 22 distribution pipes are arranged to distribute all of the spring water into the injection wells.

The injection wells, drilled into the mountainside, have a diameter of 2 m and a depth of 3-4 m; they are packed with rocks and are situated about 7-8 m from the channel. A wooden distribution pipe connects the channel with each injection well. As shown in Fig. 1, the first six injection wells are situated on an almost flat area on the mountainside; and because of their suitable locations, these wells form the most convenient well group into which the hot-spring water is injected for filtering. Each of these six wells has a filtering capacity of 10 l/sec, greater than all other wells, and they have been in use for a long time. The No. 1 well of the No. 1 conveyance channel (hereafter referred to as the 1-1 well) and the No. 1 well of the No. 2 conveyance channel (the 2-1 well) have been over-filled and have overflowed. These two wells have been used intermittently since 1935, but they show no signs of deterioration in permeability.

The spring water, with a temperature of more than 70°C, injected into the well percolates 150-200 m down through the ground and reacts with the rock and clay. After 90% of its acidity is neutralized, it spouts out along the left side of the Shibukuro River. The points of spouting usually do not shift except in the case of a flood. Spout points Nos. 1, 2 and 3 are situated on the rock bed, and those of Nos. 4, 5 and 6 are situated
in the fissure of the rock bed. Apart from the above-mentioned spout points, there is also a group of tiny spout points, and in addition a group of spout points is found on the river bed. But no spout points are found along the right side of the Shibukuro River. When wells situated downstream of the conveyance channel are used, a group of spout points is noticeable along the left side, downstream, of the Shibukuro River.

Since the amount of chlorine ion effluent of the Shibukuro River almost equals that of spring water from the Tamagawa hot spring, it is almost impossible to conclude that spring water injected in the underground aquifers does not rise to the surface again. Table I shows the chemical composition of the injected spring water and the effluent water. The linear distance between the injection well and the spout point is nearly 200 m, with a difference of 100 m in height.

Because of the nature of the materials used for conveyance channels, the maintenance of these channels is difficult in the winter; therefore, this neutralizing method at present cannot be operated throughout the year.

III. BRIEF REVIEWS OF PREVIOUS EXPERIMENTS

The three experiments, involving the use of radioisotopes to trace the behaviour of filtering spring water, were carried out in August and October, 1956, and in June, 1957. A discussion of the behaviour of filtering water, based on the results obtained from the radioactivity effluent curves, is presented.

A. Experimental procedures

The 1-1 well and the 2-1 well were chosen for this experiment. Radioisotopes together with a carrier were put into the injection wells. Sampling was made at the spout points at intervals of 20 min, 30 min or 1 h. The radioactivity of the samples was measured at the laboratory. The radioactivity of a part of the samples was measured in the field by means of immersion type G-M counters or by precipitation of AgI or (NH₄)₅PO₄·12MoO₃·2HNO₃·H₂O from the water. In addition, pH, chlorine ion concentration, acidity and water temperature were also measured for confirmation of whether the filtered water was in an equilibrium state and for determination of what factors could interfere with the attainment of equilibrium.

In the first experiment 100 mc of I¹³¹ was put into the 2-1 well; and in the second experiment 60 mc of I¹³¹ was put into the 1-1 well and 50 mc of P³² into the 2-1 well at the same time. Further, in the third experiment 200 mc of P³² and 200 mc of I¹³¹ were put into the 1-1 well and the 2-1 well, respectively. Sampling was made for 100 h.

For the most part the tracing experiment was carried out while the neutralizing reaction was in an equilibrium state. But in some cases, such as during the repair of conveyance channels or flood, the sampling had to be suspended.
### TABLE I

CHEMICAL COMPOSITION OF THE WATERS (mg/l)

| Sampling points | Acidity (methyl-orange) (meq/l) | Acidity (phenol-phtalein) (meq/l) | Ca | Mg | Na | K | Al | Fe | Cl | SO₄ | SiO₂ | pH | P. | I |
|----------------|--------------------------------|----------------------------------|----|----|----|---|----|----|----|----|-----|-----|----|----|---|
| Hot-spring water | 70.8                          | 99.6                             | 146.7 | 72.5 | 96.0 | 37.2 | 463.5 | 118.8 | 3134 | 1449 | 58.0 | -   | 0.70 | 7.7 |
| No. 3           | 7.3                           | 56.2                             | 183.9 | 52.5 | 100.0 | 36.0 | 679.3 | 11.8 | 1986 | 965.4 | 75.2 | 2.2 | 0.09 | 1.0 |
| No. 4           | 4.4                           | 51.6                             | 188.0 | 52.0 | 98.4 | 29.2 | 603.4 | 36.4 | 1915 | 909.4 | 75.0 | 2.2 | 0.21 | 1.5 |
| No. 6           | 10.5                          | 66.6                             | 179.9 | 34.0 | 92.0 | 30.0 | 773.2 | 30.8 | 2220 | 1014 | 80.0 | 2.0 | 0.62 | 2.1 |
| No. 7           | 0.25                          | 14.3                             | 153.9 | 49.2 | 73.5 | 28.4 | 196.3 | 0.4  | 955.6 | 197.9 | 89.5 | 3.4 | -   | -   |
| No. 8           | 0.20                          | 13.1                             | 173.5 | 41.2 | 65.3 | 30.9 | 178.0 | 0.2  | 946.8 | 182.7 | 77.2 | 3.4 | -   | -   |
| No. 9           | 0.13                          | 10.4                             | 193.2 | 27.9 | 56.4 | 24.1 | 163.0 | 0.2  | 831.5 | 152.7 | 110.3 | 3.5 | -   | -   |
| No. 10          | 0.00                          | 5.7                              | 187.2 | 18.7 | 45.3 | 18.8 | 115.0 | 0.1  | 578.0 | 126.3 | 62.5 | 3.8 | -   | -   |
B. Results

The results obtained from the three above experiments are shown in Fig. 2. This figure shows the relation between the concentration of radioactivity in a 50 cm³ sample of water and the time of appearance at the spout point. Here the time of appearance is taken from the difference between the time of radioisotope injection and the time of sampling. In Fig. 3 the change of chlorine ion concentration by time is shown. These results were obtained from the second experiment.

C. Summary of the results

1. The characteristics obtained from the radioisotope effluent curve reveal that the appearance of radioactivity was intermittent, most of it appearing within 20 - 40 h, the radioactivity was weak, and the peaks obtained were not dispersing.

At each spout point, that is, sampling point, radioactivity was detected from both the 1-1 well and the 2-1 well. At spout points Nos. 2, 3 and 4 the
radioactivity from the 1-1 well appeared earlier than did that of the 2-1 well, but at the points Nos. 5 and 6 radioactivity from the 2-1 well appeared earlier than did that of the 1-1 well. This curve clearly discloses the underground condition, where the injected hot-spring water has passed through. The injected water filtered through the divergent underground water channels or the layers of different permeability in different volumes depending on the permeability of the layer and the capacity of the underground channel. The neutralizing effects achieved by the filtering process are considered to vary in results depending on the differences in the rate of filtering, geological conditions and water temperatures or differences in the degree of mixing with underground water.

2. The broad peak of the radioisotope effluent curve indicates the existence of a prevailing channel leading to the particular spout point. The relatively well-shaped peak indicates the existence of a permeable layer with a uniform coefficient of permeability. In addition, the dispersed peak shows where filtering water passed through aquifers of different permeability and suffered adsorption and desorption of radioisotopes, undergoing mixing and disturbance by aquifers of large volume.

Further, the peak appearing in a relatively short time is considered as showing water which passed through a layer with a high coefficient of permeability and the peak appearing later as showing that the water passed through a layer with a low coefficient of permeability or took a detour.

Concerning the effect on neutralization, it can be assumed that the dispersed and delayed peak usually represents a higher degree of neutralization, and the peak appearing in a relatively short time usually represents a lower degree of neutralization.

3. The spring water, filtering from the injection well, which diverges by fingering and then spouts out after finding its way through the crevices, has mixed with groundwater. This phenomenon is especially obvious at the second and third spout points but not so obvious at the fourth, fifth and sixth spout points. In the latter cases the spring water has filtered through a rock bed where there was no prevailing ground water.
4. Spring water from both the 1-1 and the 2-1 wells was detected at all of the six spout points. This means that the two kinds of water mixed before they reached the rock crevices or that intertwined crevices exist.

5. In the third experiment several peaks of high radioactivity were detected within one hour after injection. This peak probably indicates that underground channels of a considerable size from the 1-1 well had been formed during the previous eight months of the filtering process. On the other hand, there is no evidence of the existence of large underground channels from the 2-1 well to the Nos. 1, 2, 3 and 5 spout points. If a flow of hot-spring water of 40 l/sec has been treated during those eight months, then nearly 1000 tons of $\text{Al}_2\text{O}_3$ must have been dissolved by the neutralizing reaction, resulting in an increase of porosity of the underground soil of 0.2%. This increment in porosity takes place locally in underground channels; therefore, it can be assumed that characteristics of the underground channels have been changed. On some occasions the volume of effluent decreased suddenly. This was probably caused by clogging in the channel.

6. In these three experiments, as is evident from analysis of the water quality, equilibrium of neutralization has not been established. With the analysis data obtained the water storage capacity of aquifers can be calculated. For example, assuming that an aquifer with volume $A$ leads to spout point No. 2 and that an influx $Q'(\text{m}^3/\text{h})$ enters this system, then by the use of the relation that the decrease of chlorine ion concentration is a logarithmic function of time, and by the equilibrium chlorine ion concentration value of $4.82\times10^{-2}\text{eq/l}$ obtained in the second experiment, $A = 128 Q'$. By the same procedure $A = 236 Q'$ from the values observed in the third experiment. This means that the water content of this aquifer has increased, and, if $Q'$ is assumed to be 5 l/sec, $A$ is about 4200 m$^3$.

IV. THE FOURTH EXPERIMENT

It is clear that the equilibrium state in neutralization has not been established in the previous experiments. So the fourth experiment was planned for comparison of the data previously obtained with the relevant data obtained in an equilibrium state. In this experiment $^{131}\text{I}$ and $^{32}\text{P}$ were injected at an interval of 5 h into the same injection well. Two radioisotope effluent curves were obtained.

A. Experimental procedures

Hot-spring water had been injected into the injection well for 17 d before the experiment was started. The existence of an equilibrium state of neutralization was confirmed by the measurement of the chlorine ion concentration. Then radioisotopes were put into the 1-1 well on 26 September 1957.

At first 100 mc of $^{131}\text{I}$ and 5 h later 100 mc of $^{32}\text{P}$ with carrier were poured in respectively. At spout points sampling was carried out at intervals of 15 min for 80 h. The radioactivity of the water samples was measured at the laboratory.

5 cm$^3$ of N/100 Na$_4$ solution was added to the sample as a carrier, then 0.5 cm$^3$ of N/10 Na$_2$PdCl$_4$ solution, filtered through a glass filter after being
stirred and heated for 10 min at 80°C, was added. The precipitated PdI$_2$ amounted to nearly 9 mg, and self-absorption of $\beta$-radiation was found negligible. Then 1 cm$^3$ of N/10 (NH$_4$)$_3$PO$_4$ solution, 10 cm$^3$ of 6 N-HNO$_3$ and 7.5 cm$^3$ of 1 N (NH$_4$)$_2$MoO$_4$ solution were added to the filtrate, separated from PdI$_2$ precipitate, after being stirred and heated at 80°C; and a yellow precipitate was obtained. The precipitates settled for 6 h at room temperature. The precipitates obtained amounted to nearly 70 mg, and the effect of $\beta$-radiation self-absorption was negligible. Radioactivity was measured by a G-M counter.

The chlorine ion concentration, acidity, water-temperature and volume of effluent were also recorded.

B. Experimental results

The radioisotope effluent curves are shown in Fig. 4. The abscissa indicates the time elapsed after the radioisotope injection. The detection standard of radioactivity was set at 3.12 $\sigma$. Chlorine ion concentrations and acidities of the samples collected from each spout point virtually did not change throughout this experiment.

C. Discussion of the results

(1) The characteristics of radioisotope effluent curves shown in Fig. 4 are as follows: All radioactivity detected was weak; and although a considerable concentration of radioisotopes was detected at the Nos. 2 and 3 spout points, no radioactivity was found at the Nos. 4 and 6 spout points. At the Nos. 2 and 3 points the appearance time of the peak showed a time lag of nearly 15 - 30 min. Despite the fact that a few peaks were recorded on the I$^{131}$ curve within a short time, there was only one peak in the P$^{32}$ curve,
and it was impossible to compare these peaks. No remarkable peak of strong radioactivity, such as that detected in the third experiment, was recorded in this experiment.

(2) In respect to the latter part of (1) above, the absence of a remarkable peak and of a time difference which should have reflected the time lag of the radioisotope injection suggests the disappearance of the prevailing underground channel which had been formed during the third experiment. The number of different water samples in which $^{32}$P or $^{131}$I was detected indicates that the rate of loss of $^{32}$P and $^{131}$I was not identical because of precipitation or absorption during the infiltration process.

(3) The time difference due to time lag of radioisotope injection was not reflected in the data obtained. This may result from the unstable conditions of underground soil, the ion exchange effect and the time lag between $^{32}$PO$_4^{3-}$ and $^{131}$I$^-\text{in absorption and desorption by soil. These phenomena have been studied by McAULIFFE [4], YUAN [5] and YUAN and ROBERTSON [6] as resulting from the absorption of phosphorus caused by the isotope effect. The same study on this phenomenon has been reported by SEATZ [7] and is considered to be applicable in the case of $^{131}$I too. The neutralization effect of Tamagawa hot-spring water by filtering is mainly caused by the dissolution of Al$_2$O$_3$; hence high aluminium and ferric ion concentrations were detected in the effluents. Therefore, these ions will turn into hydroxides or phosphates and precipitate in water of high pH value produced by neutralization. Consequently, phosphate ions in the spring water or of the radioactive phosphoric tracer precipitate in the ground. This process is especially evident in a part of channel where the pH has increased above 3. The loss of $^{32}$P by absorption occurs in the vicinity of the injection well, and the loss by precipitation occurs at a relatively distant area from the injection well. The loss of $^{32}$P because of ion exchange with the soil also occurs to some extent, as reported by AMER [8], RUSSEL [9] and TALIBUDEEN [10].

(4) The measurement of hydrogen and chlorine ion concentrations shows that an equilibrium state has been established between the effluent and the underground channel. However, since the effluent is mixed with filtered water which underwent neutralization or dilution by underground water, the quality of the spring water which has passed through an individual channel must be profoundly affected by the underground channel characteristics involved. The time lag noticed in the radioisotope effluent is considered as resulting from this unstable and changing condition of underground soil. Sudden collapse, clogging and contraction of channels and the concomitant change in the coefficient of permeability cause the change in channel characteristics. Besides, the characteristics of channels change with the alternation of the contact surfaces between filtering spring water and rock and with the continuous dissolution of soil or the transformation of the other physical and chemical factors involved in underground channels. Furthermore, the characteristics of channels are constantly changing because of the formation of a high-pH channel underground.
The radioisotope tracing experiment on the behaviour of the filtering hot-spring water was made under equilibrium conditions in respect to neutralization. $^{131}$I and $^{32}$P, which were poured into the injection well at time intervals of 5 h, did not appear with the same time difference. The differences in behaviour between $^{131}$I and $^{32}$P are assumed to be due to the unstable chemical and physical factors involved in ground soil, the settling of $^{32}$P in high-pH underground channels and the different isotope effect of each nuclide on soils and rocks. Quantitative studies of these problems are essential in future studies.

Furthermore, degradation in the neutralization capacity of underground channels, which can be attributed to the long neutralizing process, have been noticed in this experiment.

V. THE FIFTH EXPERIMENT

From the data obtained in the previous four experiments an extensive discussion on the behaviours of spring water during filtering is possible. This experiment was prepared for determination of the possibility of radioisotope tracing at a greater distance than that of the previous 200-m tracing experiment.

A. Experimental procedures

The 1-5 well, which is 500 m below the 1-1 well, was chosen for this experiment. The spout points Nos. 7, 8 and 9 of this injection well were found 400 m downstream from the No. 6, and the No. 10 spout point was found 500 m downstream from it.

The linear distance between the 1-5 well and these spout points was nearly 600 m. Hot-spring water was poured into the 1-5 well for 17 d before the radioisotope injection. The sampling was made at intervals of 30 min within the 24-h period after the radioisotope injection and at intervals of 15 min thereafter for a total of 151 h.

The quantity of radioisotopes used was 100 mc of $^{32}$P, and the measurement procedures were the same as in the previous experiment.

B. Experimental results

Only 49 of all the samples showed a value exceeding 3 $\sigma$ in statistical deviation of their radioactivity in cpm. From these 49 samples, precipitation samples were taken and measured for their radioactivity for 20 min. Five out of 49 samples showed more than 3 $\sigma$ in statistical deviation, and these five samples had been collected from spout point No. 7. The results were as follows:
A. ARIIZUMI and O. KONDO

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Time elapsed (h)</th>
<th>Counts/20 min</th>
<th>Deviation in 1σ</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>116⅔</td>
<td>722</td>
<td>3.24</td>
</tr>
<tr>
<td>2</td>
<td>119¼</td>
<td>726</td>
<td>3.40</td>
</tr>
<tr>
<td>3</td>
<td>121⅓</td>
<td>732</td>
<td>3.64</td>
</tr>
<tr>
<td>4</td>
<td>124⅔</td>
<td>725</td>
<td>3.36</td>
</tr>
<tr>
<td>5</td>
<td>131⅓</td>
<td>772</td>
<td>5.22</td>
</tr>
</tbody>
</table>

Furthermore, chlorine ion concentration at the No. 7 point decreased suddenly 30 h after radioisotope injection. At the No. 8 point an obvious change in chlorine ion concentration was found at about 100 h after radioisotope injection. On the other hand, a sudden increment in chlorine ion concentration was noticed at about 100 h at the No. 9 point. No sudden change was observed at the No. 10 point except the gradual tendency toward a decrease in chlorine ion concentration.

The yields of four spout points from Nos. 7 to 10 were respectively 2.5 1/sec, 1.5 1/sec, 1.2 1/sec and 3.0 1/sec with a pH value of 3.4, 3.5, 3.5 and 3.8 and water temperature of 20.3, 19.4, 16.4 and 11.7°C.

C. Discussion of the results

(1) The existence of radioactivity was not clear in those samples which showed a deviation of 3σ in measurement of radioactivity, but the sample with a deviation of 5.22σ can be considered to be radioactive. This sample was collected from the No. 7 spout point 131.5 h after radioisotope injection. Hence, the filtering speed in the ground was calculated as 4.4m/h.

(2) Of the four spout points, P$_{32}$ was detected only at the No. 7 point. As shown in Table I, the effluent from the No. 7 point was diluted nearly three times by underground water. Hence, the acidity of this effluent was lowered considerably, and the concentration ratio of aluminium ions to chlorine ions was found less than those obtained from Nos. 2 to 6 effluents. This fact indicates that precipitation of Al(OH)$_3$, accompanied by neutralizing process, took place. Along with precipitation of Al(OH)$_3$, the PO$_4^{3-}$ contained in the spring water and added as a carrier and tracer settled out as AlPO$_4$. This reaction occurred at a pH larger than 3. Since the pH value of all effluents was found to be more than 3.4, P$_{32}$ added as tracer settled out and was trapped in the underground channels. The fact that the P$_{32}$ was detected only at the No. 7 point should be interpreted to mean that the effluent of the No. 7 point had the lowest pH value and that the effluent filtered through relatively low pH channels without suffering precipitation.

(3) It shall be noted that a high pH environment will form at particular parts of a channel and that this state will cause the local precipitation of Al(OH)$_3$ even if the total effluent does not show a pH value high enough to form Al(OH)$_3$. Accordingly, the tracer filtered through this part will settle
out. In such a case P\textsuperscript{32} should be used in forms other than phosphates or used continuously in large quantities.

(4) Except at the No. 7 point, tracing is impossible as long as \textsuperscript{32}PO\textsubscript{4}\textsuperscript{3-} is used. Thus, \textsuperscript{32}PO\textsubscript{4}\textsuperscript{3-} as a tracer for the tracing experiment at the Tamagawa hot spring has limited application. Furthermore, the loss caused by absorption of phosphate ions and the effect of ion exchange, as cited before, should also receive attention.

(5) Sufficient data for the analysis of sudden changes in chlorine ion concentration have not been obtained. At present it is assumed that the unstable states of underground channels and the unsteadiness of flow through the filtering route have caused these sudden changes.

D. Summary

The application of P\textsuperscript{32} in the form of phosphate ion is restricted in tracing underground water containing high concentrations of Fe\textsuperscript{+++} or Al\textsuperscript{+++}. In this experiment the tracing was impossible if pH was larger than 3.5. In the case of a low pH value a tracing distance of 600 m by \textsuperscript{32}PO\textsubscript{4}\textsuperscript{3-} was found possible.

VI. THE SIXTH EXPERIMENT

From the first five experiments the geological conditions of the ground where the neutralization process took place were outlined. In addition, the time lag because of nuclides used in tracing and the loss of radioisotopes due to absorption, usually noticeable in the laboratory tests, were also confirmed in the field test.

In previous experiments anionic ions, namely \textsuperscript{32}PO\textsubscript{4}\textsuperscript{3-} and \textsuperscript{131}I\textsuperscript{-}, were applied as tracers. As the results have shown already that \textsuperscript{32}PO\textsubscript{4}\textsuperscript{3-} was not suitable for a quantitative experiment, tritium, \textsuperscript{86}Rb\textsuperscript{+} and \textsuperscript{32}PO\textsubscript{4}\textsuperscript{3-} were used in this experiment for comparison of their tracing characteristics.

A. Experimental procedures

The spring water was injected into the well for 3 months to permit carrying out of this experiment under an equilibrium state in respect to neutralization. Since the purpose of this experiment was to determine the differences of tracing characteristics among three kinds of radioisotopes, these three radioisotopes were used as a mixture. The 1-1 well and the No. 6 spout point which had shown effluent of highest radioactivity throughout the previous experiments were chosen for this experiment. Instead of being sampled at intervals, the effluent from the No. 6 spout point was led through a pipe to a collection vessel, and sampling was made at intervals of 5 min.

The radioisotopes used were 200 mc of \textsuperscript{32}PO\textsubscript{4}\textsuperscript{3-}, 200 mc of \textsuperscript{86}RbCl and 1 c of H\textsuperscript{3}. These three radioisotopes were mixed in a 2-1 vessel and poured in at one time. Rb\textsuperscript{86} in the effluent was settled out as Rb\textsubscript{2}PtCl\textsubscript{6} in 80% alcohol,
and P₃² was measured by the same procedures as those in the previous experiments. Tritium was measured by the gas-counting method.

The experiment was carried out in September 1958. Samples were collected for 100 h.

B. Experimental results

Of the samples, four with P₃² and five with Rb⁸⁶ were obtained as shown in Table II. Radioactivity was detected in the consecutive samples Nos. 828 and 829, and P₃² was also detected in these samples.

The pH and chlorine ion concentrations in the samples were measured at intervals of 2 h. The results showed that these values did not change during the 100-h period of this experiment. The number of spout points in-

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Time of appearance after addition of radioisotopes</th>
<th>Nuclide</th>
<th>Radioactivities in cpm on 20 Oct.</th>
<th>Concentration of radioisotopes (mc/l)</th>
<th>Diluted volume (10³ m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>779</td>
<td>64°55'</td>
<td>P₃²</td>
<td>36.8</td>
<td>4.6</td>
<td>43.5</td>
</tr>
<tr>
<td>789</td>
<td>65°45'</td>
<td>Rb⁸⁶</td>
<td>30.1</td>
<td>7.4</td>
<td>27.0</td>
</tr>
<tr>
<td>811</td>
<td>67°35'</td>
<td>P₃²</td>
<td>8.5</td>
<td>1.0</td>
<td>200</td>
</tr>
<tr>
<td>824</td>
<td>68°40'</td>
<td>Rb⁸⁶</td>
<td>15.0</td>
<td>3.6</td>
<td>55.5</td>
</tr>
<tr>
<td>828</td>
<td>69°00'</td>
<td>P₃²</td>
<td>214.1</td>
<td>27.2</td>
<td>7.4</td>
</tr>
<tr>
<td>829</td>
<td>69°15'</td>
<td>P₃²</td>
<td>200.9</td>
<td>25.6</td>
<td>7.8</td>
</tr>
<tr>
<td>951</td>
<td>79°15'</td>
<td>Rb⁸⁶</td>
<td>4.3</td>
<td>1.0</td>
<td>200</td>
</tr>
<tr>
<td>973</td>
<td>81°05'</td>
<td>Rb⁸⁶</td>
<td>96.8</td>
<td>23.9</td>
<td>8.4</td>
</tr>
<tr>
<td>987</td>
<td>82°15'</td>
<td>Rb⁸⁶</td>
<td>17.2</td>
<td>4.2</td>
<td>47.6</td>
</tr>
</tbody>
</table>

The counts of radioisotope standard solutions on 20 Oct. were
P₃²: 157.3 cpm/mc
Rb⁸⁶: 81.5 cpm/mc
creased from five in the fifth experiment to 24 in this experiment. In addition, the water temperature increased. New spout points were found in the crevices of rock.

C. Discussion

(1) No relation in time of appearance between P\textsuperscript{32} and Rb\textsuperscript{86} was noticed. Since these two radioisotopes were used as a mixture, it was assumed that P\textsuperscript{32} and Rb\textsuperscript{86} should appear simultaneously. The assumption was not verified, and this can be interpreted as having been caused by the different chemical properties of the channel in relation to P\textsuperscript{32} and Rb\textsuperscript{86} respectively. That is, a channel which tends to absorb more Rb\textsuperscript{+} will absorb less PO\textsubscript{4}\textsuperscript{2-}, and vice versa.

(2) The measured radioactivities were converted to the initial concentrations (meq/l), and the results are shown in Table II. The dilution ratio of the radioisotopes, based on the assumption that radioisotope loss is negligible during infiltration, was calculated and is shown in Table II.

From this table the lowest dilution ratio was found to be in samples Nos. 828 and 829, and their dilution volume was 7000 m\textsuperscript{3}. Since this volume was related to the total quantity of radioisotope added, if it is assumed that the underground channel represented by this peak will pass about one-tenth of the total water injected, the dilution in this channel will be with 700 m\textsuperscript{3} of water. In other words, radioisotopes entering this channel will be diluted in 700 m\textsuperscript{3} of water and spout out within 10 min from the No. 6 spout point.

This assumption leads to an effluent speed of 1 m\textsuperscript{3}/sec and is found to be unacceptable in comparison with the observation value. On the other hand, assuming an effluent of 10 l/sec at the spout point, then the radioisotopes remaining in the effluent reduce to 1% of the initial quantity used. From the measured effluent speed and previous tracing experiments radioisotopes reaching the spout point can be assumed to be less than 0.2% of the total radioisotopes used.

Since this value is based on the channel where the radioisotope loss is the lowest of all, high dilution ratios such as those of samples Nos. 811 and 951 suggest a high percentage of loss of radioisotopes.

(3) A high percentage of loss of radioisotopes resulting from absorption indicates a slow process of filtering or considerable contact of spring water with rock or soil. It took at least 64 h 55 min for the injected spring water to appear at the spout point. This long filtering time suggests a high percentage of loss of radioisotopes.

Further, the underground channels which lead to the No. 6 spout point are estimated to be not less than eight from which radioactivity was detected.

D. Summary

In this experiment, new spout points were noticed and the equilibrium in respect to neutralization was established.

At the No. 6 point three peaks of P\textsuperscript{32} and five peaks of Rb\textsuperscript{86} were observed. But no relationships were established between these peaks. These results
suggest that the chemical properties of the underground channel in relation to PO$_4^{3-}$ and to Rb$^+$ are not the same.

The loss of radioisotope ions resulting from absorption was found to be extremely high, almost more than 99%. This high percentage of loss influences the radioisotope effluence. If the loss is less than 100%, radioisotope peaks will appear, and if it is 100% they will not appear. The earliest peak obtained in this experiment came 64 h 55 min after injection of radioisotopes. In comparison with those obtained from the previous experiments it was the latest, which suggests a high percentage of loss of radioisotopes.

VII. EPILOGUE

The method for neutralization of the Tamagawa hot-spring water by the filtering process has been studied by means of the application of radioisotopes. Tracing with the aid of radioisotopes was found to be an effective method for this study.

Structures of underground channels were outlined from the radioisotope effluent curves. It was found that the spring water filters along divergent and independent underground channels in the rock crevices and finally reaches the spout points. The neutralization reaction in these channels is assumed to be in a transient state rather than in a steady state.

The behaviours of radioisotopes in the channel are different, depending on the nuclides, the isotope effects in absorption and desorption, the ion exchange effect and the precipitation or scavenging accompanying the change of pH value. The loss of radioisotopes during filtering was found to be more than 99%. Estimation of the water storage capacity of aquifers, based on the data obtained, has been made. Aging of the underground channels, resulting from springwater treatment of long duration was noticed.

The data obtained from the analysis of the spring-water quality will be a useful reference for the interpretation of the results of radioisotope tracing.

ACKNOWLEDGEMENTS

The authors are grateful to Y. Mori, M. Oba and M. Makita of the Institute for their co-operation in this study and also to the authorities of Akita Prefecture for their assistances in arranging field experiments.

REFERENCES

DIFFUSION

R. HOURS (Chairman): This is of course a particularly interesting investigation not only because of the novelty of the problem but also because of the variety of tracers and measurements employed. For one thing it is a good example of how various hydrogeological water-analysis methods can be applied to a single problem; and, by involving the simultaneous use of different tracers on one terrain, it is also valuable from the point of view of the information it provides on their relative behaviour.

P. ZUTSHI: Was any attempt made to identify the phosphorus-32 activity in the samples by way of absorption or half-life measurements?

O. KONDO: We did check for phosphorus-32 using the beta-ray absorption method with different thicknesses of aluminium foil but we did not check the half-life of the nuclide.
THE USE OF THE CARBON ISOTOPES IN GROUNDWATER STUDIES

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Abstract — Résumé — Аннотация — Resumen

THE USE OF THE CARBON ISOTOPES IN GROUNDWATER STUDIES. The bicarbonate in groundwater is dissolved in the upper layers of the soil by the action of biogenic carbon dioxide on limestone. As a result of this solution mechanism the bicarbonate in shallow groundwater appears to have a quite characteristic $^{13}$C and $^{14}$C content so that these isotopes can be used to study isotopic exchange with and storage times in the aquifer. These circumstances also make it possible to date fresh water carbonates precipitated from such water.

Measurements on water from deeper levels indicate that low $^{14}$C concentrations are mainly a result of radioactive decay and thus give true residence times. Values of 10,000 yr and more are not uncommon. Several samples from the Netherlands suggest a stagnant or slowly moving substratum overlain by a thin layer of young water.

Oxygen-18 and deuterium measurements on dated groundwater give information on the isotope composition of the original precipitation and it may be possible to correlate this with climatic changes when more data are available.
EMPLEO DE LOS ISÓTOPOS DEL CARBONO EN EL ESTUDIO DE AGUAS SUBTERRÁNEAS. El bicarbonato de las aguas subterráneas procede de las capas superiores del suelo, formándose por la acción que ejerce sobre las calizas el anhídrido carbónico biogénico. Como consecuencia de este mecanismo de disolución, el bicarbonato de las aguas subterráneas poco profundas parece tener un contenido muy característico en $^{13}$C y $^{14}$C, de modo que estos isótopos pueden servir para estudiar el intercambio isotópico con la capa acuífera y el tiempo de retención en la misma. Dado a estas circunstancias, permiten también determinar la edad de los carbonatos precipitados desde aguas dulces.

Las mediciones realizadas con aguas tomadas a profundidades mayores revelan que las bajas concentraciones de $^{14}$C se deben principalmente a la desintegración radiactiva e indican, por tanto, el tiempo de retención efectivo. No es raro encontrar valores de 10 000 años y aún superiores. Varias muestras tomadas en los Países Bajos sugieren la existencia de un substrato estancado o animado de un movimiento muy lento, recubierto por una delgada capa de agua más reciente.

Las mediciones del oxígeno-18 y del deuterio en aguas subterráneas cuya edad fue previamente determinada informan sobre la composición isotópica de la precipitación original y, cuando se disponga de más datos, quizá sea posible realiación con los cambios climáticos.

INTRODUCTION

The movement of groundwater is often so slow, and storage times so long that the method of spiking the system is found to produce negative results and it seems desirable to look for natural tracers which would cover a longer time span. In this respect carbon-14 with a half-life of 5700 yr appears to offer promising possibilities. The isotope is present in groundwater in the form of dissolved bicarbonate and, subject to certain conditions, could be used to determine residence times ranging up to some 40 000 yr.

If true ages are to be obtained two basic requirements must be fulfilled:
(1) The initial $C^{14}$ content must be known, and
(2) The $C^{14}$ content must not change in the course of time by any other means than radioactive decay.

The extent to which this is the case is discussed in the following paragraphs.

1. ORIGINAL AND INITIAL CONCENTRATION OF $C^{14}$ IN GROUNDWATER

$C^{14}$ produced in the atmosphere by cosmic radiation is present in atmospheric carbon dioxide and living plants at an established constant level. The relative concentration $C^{14} / C^{12}$ in recent material is approximately $10^{-12}$ and will be denoted by 100%.

The initial $C^{14}$ content of the bicarbonate in groundwater depends in the first instance on the solution mechanism involved. By far the most common origin of the bicarbonate is the solution of limestone by the action of carbon dioxide according to the equation

$$CaCO_3 + CO_2 + H_2O \rightleftharpoons Ca^{++} + 2HCO_3^{-}.$$  \hspace{1cm} (1)

Secondly there is the weathering of silicate rocks which can be represented by the equation

$$CaAl_2(SiO_4)_2 + 2CO_2 + H_2O = Ca^{++} + 2HCO_3^- + Al_2O_3 + 2SiO_2.$$  \hspace{1cm} (2)
The extreme softness of groundwater in granitic regions where limestone is absent demonstrates that silicate weathering is a slow process. In such regions, however, it may provide a notable portion of the dissolved bicarbonate.

Finally humic acids will react with calcium carbonate to produce carbon dioxide and bicarbonate in solution:

\[
\begin{align*}
\text{CaCO}_3 + 2 \text{H(Hum)} & \rightleftharpoons \text{Ca(Hum)}_2 + \text{CO}_2 + \text{H}_2\text{O} \\
2 \text{CaCO}_3 + 2 \text{H(Hum)} & \rightleftharpoons \text{Ca(Hum)}_2 + 2\text{Ca}^{++} + 2\text{HCO}_3^-
\end{align*}
\]

(3)

The concentration of humic acid in the soil is usually too small to be of any significance except under the special conditions encountered in peat bogs where the acidity can become remarkably high. An instance where solution obviously is effected in this way is discussed below.

Returning to the normal case represented by Eq. (1) we must next consider the source of the carbon dioxide. Contrary to popular belief this source is not the atmosphere, but the biologically active layers of the soil where considerable amounts of CO$_2$ are continuously being generated by root respiration and the decay of humus. A quantitative consideration of the equilibrium reaction as presented in Fig. 1 will make this clear.

---

**Fig. 1**

Equilibrium concentrations of bicarbonate and carbon dioxide in water in contact with limestone as a function of the CO$_2$ partial pressure in the gas phase. The arrow denotes the normal atmospheric CO$_2$ concentration.
Water in contact with the free atmosphere ([CO₂] = 0.03 vol. %) can only dissolve 1.2 mM HCO₃⁻/l while values of 5 to 8 mM/l are frequently encountered in groundwater, especially under humid climatic conditions and in calcareous soils. A bicarbonate concentration of 8 mM/l approximately corresponds to the maximum partial pressure of carbon dioxide (5\%) occurring in the soil – higher concentrations act as inhibitors and the biological oxidation stops.

The dissolution process takes place in the upper humus-bearing layers of the soil. Below a depth of a few meters there is virtually no more production of carbon dioxide and consequently no more calcium carbonate can be dissolved.

Experiments on lysimeters have actually demonstrated that the bicarbonate hardness of seepage water depends on the composition and biological activity of the soil and that within two or three meters below the surface the water has already essentially attained its ultimate bicarbonate concentration.

It is important to note that the solution of limestone consumes an equivalent amount of carbon dioxide (Eq. (1)) and that the carbon dioxide is of biogenic origin. In addition to the dissolved bicarbonate, the water also contains a certain – usually much smaller – amount of dissolved CO₂, which is required to stabilize or hold the bicarbonate in solution. The actual amount for any specific case can be deduced from Fig.1.

The limestone in the soil, generally having been deposited millions of years ago, is practically C¹⁴-free, while the biogenic carbon dioxide has a C¹⁴ content close to that of the atmosphere (100\%). According to Eq. (1), the bicarbonate would thus be expected to have a C¹⁴ content half as large as that in the plant material from which the soil C is derived. Experience shows that the C¹⁴ content is actually larger (generally around 85\%), owing to the fact that isotopic exchange takes place between the gaseous carbon dioxide and the bicarbonate solution. Depending on local conditions, the C¹⁴ concentration can thus theoretically attain values between 50 and 100\%.

In Central Europe, where the geological and hydrological aspects of the problem are well understood, and where the C¹⁴ content of recent waters has been studied in detail, the error in the initial C¹⁴ content can be as small as 5\%. In arid regions where we have only poor information about the dissolution mechanism it can be a factor of two. This uncertainty affects the accuracy of the age determination. An initial C¹⁴ content of 50\% instead of 100\% causes the water to appear one half-life (5700 yr) older.

2. SUBSEQUENT ALTERATIONS IN THE C¹⁴ CONTENT OF GROUNDWATER

It was pointed out in the previous paragraph that the solution of limestone requires the presence of carbon dioxide. After the water has penetrated to depths where there is no more local production of carbon dioxide, the solution ceases, and the water remains essentially unchanged until it emerges in a spring or is pumped from the ground. Here the stabilizing, dissolved carbon dioxide soon escapes to the atmosphere and CaCO₃ is precipitated as lime sinter, tuffa, etc.
While the water is underground there are two ways in which the C\textsuperscript{14} content can be diminished other than by radioactive decay: first by admixture of magmatic CO\textsubscript{2} and secondly by isotopic exchange with limestone in the water-bearing rocks.

(a) Magmatic CO\textsubscript{2}

In regions of volcanic activity large amounts of CO\textsubscript{2} may be generated. This CO\textsubscript{2} is derived from limestone sediments and contains no C\textsuperscript{14}. Admixture of such CO\textsubscript{2} to the groundwater will reduce the C\textsuperscript{14} content both by solution of additional limestone and by isotopic exchange. These areas are therefore to be avoided.

(b) Isotopic exchange with the mother rock

This important source of error must be considered carefully. Isotopic exchange would tend to distribute the C\textsuperscript{14} equally between the dissolved bicarbonate and the limestone. If it takes place to any large extent, the C\textsuperscript{14}/C\textsuperscript{12} ratio in the water will decrease seriously. Fortunately, the dissipation of C\textsuperscript{14} must be extremely slow, because it involves solid-state diffusion. Of course, a very thin surface layer of the limestone in contact with the water will be in isotopic equilibrium with the bicarbonate, but this layer forms a protective coating against further loss of C\textsuperscript{14}. These considerations, however, are only theoretical and must still be proved.

3. THE ISOTOPIC RATIO C\textsuperscript{13}/C\textsuperscript{12} AS A CRITERION FOR THE ORIGIN AND HISTORY OF CARBON IN GROUNDWATER

In the previous two paragraphs it was shown that under certain circumstances the initial C\textsuperscript{14} content of the carbon in groundwater is known and will not be subsequently altered while in the aquifer. Before the C\textsuperscript{14} content can be used to deduce actual residence times it is necessary to check whether a given sample represents a "normal" case or not. Fortunately the stable carbon isotope, C\textsuperscript{13}, provides much information on this question.

The ratio of the stable isotopes, C\textsuperscript{13}/C\textsuperscript{12}, is approximately 1 to 90. Small variations in this ratio occur owing to isotopic fractionation, the total range being about 4%.

In nature two important fractionation processes respectively cause limestone to be enriched and plants to be depleted in C\textsuperscript{13}. The relative magnitudes of these two effects are shown in Fig. 2. For practical reasons the isotopic ratio of average marine limestone has been chosen as a standard and all other ratios are given in per mille deviation from this value, C\textsuperscript{13}(‰). Marine limestones are enriched by about 8‰ and terrestrial plants are depleted by about 17‰ with respect to atmospheric carbon dioxide.

The depletion of C\textsuperscript{13} during the assimilation of CO\textsubscript{2} by plants is a kinetic effect resulting from the larger mobility and reactivity of the lighter isotopic molecule. The fractionation in the system atmospheric CO\textsubscript{2} - bicarbonate - solid carbonate in the ocean, on the other hand, is an equilibrium process; for instance in the reaction
$\text{H}_2\text{O} + \text{CO}_2 \rightleftharpoons \text{HCO}_3^- + \text{H}^+$

$\text{C}^{13}$ is slightly preferred in the bicarbonate, the difference in the isotopic ratios of the two molecular species being about 8\%.

Equilibrium fractionation effects are generally smaller than kinetic effects. In the equilibrium system $\text{CO}_2 - \text{HCO}_3^- - \text{CaCO}_3$ (solid), the only large fractionation is the one mentioned above. Fractionation in the other reactions involved,

$\text{CO}_2(\text{gas}) \rightleftharpoons \text{CO}_2(\text{dissolved})$ and $\text{HCO}_3^- \rightleftharpoons \text{CaCO}_3(\text{solid})$,

is considerably smaller [1].

The relatively large (kinetic) fractionation observed in plants enables us to trace carbon derived from organic matter in groundwater. The relationship between the carbon isotope ratios in different systems is shown in Fig. 3. On the left the system atmospheric $\text{CO}_2$ - ocean bicarbonate - marine limestone is illustrated. These conditions are not confined to the ocean; any lake in isotopic equilibrium with the atmosphere will show the same behaviour.

The central part of Fig. 3 illustrates the situation for groundwater in humid climates. Biogenic carbon dioxide is mixed in equal portions with limestone carbonate during solution, resulting in bicarbonate with $\delta\text{C}^{13} = -12\%$. Subsequent isotopic exchange with the gas phase causes the bicarbonate to
become still more depleted in C\textsuperscript{13}, eventually reaching -17‰. Complete equilibrium must allow for 8‰ difference between the CO\textsubscript{2} at -24‰ and the bicarbonate. The change from -12 to -17‰ corresponds to a change in the C\textsuperscript{14} content from 50 to 100‰.

In the soil isotopic exchange with the free atmosphere, resulting in a shift in the opposite direction, can normally not take place. The isotopic composition of fresh-water carbonates, which can be dated as well, is also shown [2]. On the right-hand side of the diagram the situation is outlined for thermal waters (magmatic CO\textsubscript{2}). The same situation would arise if humic acids are responsible for the dissolution of limestone. Both cases would result in a low C\textsuperscript{14} content, but can easily be distinguished from normal groundwater by measuring the C\textsuperscript{13}-C\textsuperscript{12} ratio. A δC\textsuperscript{13} = -16‰ would indicate an initial C\textsuperscript{14} content of 80 to 90‰. It is probably not possible, however, to quantitatively calculate the degree of exchange from these considerations, because of the uncertainty in the isotopic ratio of the initial limestone and plant material; but a useful estimate can be made.

C\textsuperscript{13} also provides information on the magnitude of isotopic exchange with C\textsuperscript{14}-free limestone in the aquifer. If this exchange were extensive the C\textsuperscript{13} content of the dissolved bicarbonate would become equal to that of the limestone (0‰). Even in deep groundwater containing very little C\textsuperscript{14} this does not occur. If, on the other hand, only a thin surface layer of the mother rock is affected, the C\textsuperscript{14} content in the bicarbonate would not change measurably. In the intermediate range a loss of C\textsuperscript{14} from the water could occur without producing a noticeable change in C\textsuperscript{13}.

ERIKSSON has recently criticized the method on these grounds [3] but we refer to the paper by MÜNNICH and ROETHER that discusses the matter in detail [4].
4. SAMPLING AND MEASURING TECHNIQUE

About 2.5 g (200 mM) of carbon are necessary for a normal C\textsuperscript{14} analysis. This corresponds to about 20 to 200 l of groundwater, depending on the carbonate hardness.

Samples are collected in 60-l polyethylene bottles and the bicarbonate extracted after acidification in the apparatus as shown in Fig. 4. The water is stirred heavily and the evolved CO\textsubscript{2} carried by thermal convection over an absorbing solution of 4N NaOH. After 24 h the CO\textsubscript{2} is quantitatively transferred to the alkali solution from which it is subsequently evolved again and prepared for the counter.

The apparatus is sufficiently simple to be used near the sampling site and it is not necessary to ship tons of water to the laboratory.

5. RESULTS

Two examples will be given of investigations carried out so far.

(a) Egypt

In collaboration with the Desert Institute in Cairo a series of samples were collected from oases and artesian wells from the Libyan Desert. Two supplementary samples were taken from shallow water galleries on the Mediterranean Coast for comparison. The collection sites are shown on the map (Fig. 5).
The results are given in Figs 6 and 7. Ages have been calculated on the basis of an initial C\textsuperscript{14} content of 72.5%. This value has been chosen to make the age uncertainty, owing to the uncertainty in the initial C\textsuperscript{14} content (50 - 100%), symmetrical (± 3000 yr). The maximum error from this source is denoted by the two arrows on the time scale. The probability distribution of the measured age is, as far as the counting error is concerned, represented by the height of the hatched area.

On the whole the desert water appears to give a C\textsuperscript{14} age between 20 000 and 30 000 yr with a slight tendency of increasing age with decreasing distance from the Mediterranean Coast. These ages would be compatible with the conception that the water was accumulated during the last pluvial. Whether the supply represents local precipitation or whether long distances have been traversed underground for instance from the Tibesti Mountains or the Abyssinian Plateau cannot be distinguished, although the extreme softness of most samples seems to favour the first possibility. For a more detailed discussion of the results see [5].

The deuterium and O\textsuperscript{18} contents of the samples (δD = -8.5%; δO\textsuperscript{18} = -11%, with respect to "Standard Mean Ocean Water") are lower than groundwater in Central Europe today and considerably lower than samples from South Africa. This might be an indication of considerably lower average temperatures at the time of precipitation.
Chemical composition of samples representing the different water types
For comparison, groundwater from Heidelberg and ocean water are shown below.
In collaboration with the Zuiderzee Werken in the Hague the area where a new polder (Southern Flevoland) is being constructed (see Fig. 8) has been sampled in an attempt to determine the rate of underground flow from the adjoining higher regions of the Veluwe. The situation as shown in Figs. 9 and 10 is roughly a large reservoir of sweet water underneath a marine clay deposit of Eemian age, with extremely brackish water on top of the clay. All the deep water seems to have the same age of about 9000 - 10000 yr which suggests practically no subterranean movement or recharge. The ages in this case have been calculated on the basis of an initial $\text{C}^{14}$ content of 85%. The uncertainty is probably not larger than 10% or ± 1000 yr. The figure obtained is quite acceptable if one assumes that, at the close of the last Glacial, after the surface had completely thawed, but the sea level was still low, precipitation could readily recharge the aquifer. With rising sea level the outlet could have been cut off and the deep circulation brought to a standstill.

The shallow brackish water contains far too much bicarbonate for solution in the soil by CO$_2$. Partial pressures of carbon dioxide of approximately 1 atm would be needed. Furthermore the $\text{C}^{13}$ content (about +9%) shows that the carbon is not of biogenic origin. Here the obvious explanation is
that humic acids from the several submerged peat layers in the glacial and
holocene sands have caused the large scale solution of calcium carbonate.
Thus the varying, but generally low $^{14}C$ concentrations do not give any indi-
cation of the age of the water except that it must be younger. The results
clearly show that caution is necessary where peat beds are found.

With these two examples, which cannot be discussed in detail here, we
have tried to demonstrate the use of isotope analyses of carbon in ground-
water studies.
REFERENCES


DISCUSSION

See the discussion following the paper "A comparison of carbon-14 and tritium ages of groundwater", these Proceedings.
A COMPARISON OF CARBON-14 AND TRITIUM AGES OF GROUNDWATER

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Abstract — Résumé — Аннотация — Resumen

A COMPARISON OF CARBON-14 AND TRITIUM AGES OF GROUNDWATER. Simultaneous measurements of C\textsuperscript{14} and tritium in groundwater samples from different horizons have been undertaken. The aim is to find out whether there might be a decrease in C\textsuperscript{14} which exceeds the radioactive decay and could be attributed to exchange with the aquifer. As suggested by ERIKSSON [8] this in principle might be the case despite the fact that the initial C\textsuperscript{13} content is not altered appreciably. A relatively thick but exchangeable surface layer of the individual grains in the aquifer could possibly reach isotopic equilibrium in a time long compared to the C\textsuperscript{14} half-life, so that the deeper portions of the exchangeable layer would be low in C\textsuperscript{14}. Although tritium can only cover a relatively short time span it might provide a valuable check, since any type of exchange must follow the laws of diffusion and be proportional to the square root of time, independent of the numerical value of the diffusion constant.

A few measurements made so far of the same wells in different years show essentially no increase from bomb C\textsuperscript{14}. This might be attributed to a stratification of the decaying organic matter in the soil, so that the very latest material does not contribute appreciably to the solution of lime by the percolating water.

COMPARAISON DES ÂGES DES EAUX SOUTERRAINES DÉTERMINÉS À L'AIDE DU CARBONE 14 ET DU TRITIUM. On a entrepris de mesurer simultanément la teneur en C\textsuperscript{14} et en tritium d'échantillons d'eau souterraine prélevés à des niveaux différents. Ces mesures ont pour objet de déterminer s'il peut se produire une diminution de C\textsuperscript{14} qui excéderait la désintégration radioactive et pourrait résulter d'échanges avec la nappe aquifère. Ainsi que l'a suggéré E. Eriksson, tel pourrait être le cas en principe, bien que la teneur initiale en C\textsuperscript{13} ne diminue pas de façon notable. Il est possible qu'un couche superficielle relativement épaisse, mais susceptible d'échanges, de chacun des grains qui constituent la nappe aquifère puisse atteindre l'équilibre isotopique en un temps plus long que la période de C\textsuperscript{14}, de telle sorte que les parties de la couche échangeable auraient une faible teneur en C\textsuperscript{14}. Bien que le tritium ne puisse couvrir qu'une durée relativement brève, il peut constituer un bon instrument de vérification étant donné que tous les types d'échanges doivent suivre les lois de la diffusion et être proportionnels à la racine carrée du temps, quelle que soit la valeur numérique de la constante de diffusion.

Les quelques mesures faites jusqu'à présent sur les mêmes puits, au cours d'années différentes, n'accusent aucune augmentation de la teneur en C\textsuperscript{14}, consécutive aux essais d'armes nucléaires. Ceci est peut-être attribuable à une stratification des matières organiques en décomposition dans le sol, qui fait que les matières toutes récentes ne contribuent pas de manière appreciable à la dissolution du calcaire par les eaux d'infiltration.

СРАВНЕНИЕ УГЛЕРОДНОГО И ТЮТИЕВОГО ВОЗРАСТОВ ГРУНТОВЫХ ВОД. Предпринимались одновременные измерения радиоактивного углерода С\textsuperscript{14} и трития в образцах грунтовых вод из разных горизонтов с целью выяснить уменьшение содержания радиоактивного углерода С\textsuperscript{14}, которое превышало бы радиоактивный распад и могло бы быть приписано обмену с водоносным горизонтом. По предположению Е. Эриксона такое явление в принципе может быть, несмотря на то, что основное содержание С\textsuperscript{13} заметно не изменяется. Сравниваем слой воды, но способен участвовать в обмене поверхностный слой отдельных зерен в водоносном горизонте мог бы достигнуть изотопного равновесия за период времени, более продолжительный по сравнению с периодом полураспада радиоактивного углерода С\textsuperscript{14} так, что нижние части слоя содержали бы небольшое количество углерода С\textsuperscript{14}. Несмотря на то, что тритий охватывает только сравнительно короткий период, он мог бы служить ценным средством контроля, так как какой обмен должен подчиняться законам диффузии и быть пропорциональным квадратному корню из времени независимо от численного значения постоянной диффузии.
Essentially there are two naturally occurring isotopes which have been applied to groundwater-dating so far, namely tritium and C\textsuperscript{14}. In both cases, however, specific difficulties arise. Therefore, it seems promising to undertake simultaneous measurements of both isotopes on the same sample. This is true despite the fact that the intrinsic time scales of both methods do not in practice overlap in such a way that true "concordant ages" can be obtained.

A study of tritium and C\textsuperscript{14} in various groundwater samples was started in Heidelberg recently. The stable isotopes deuterium, carbon-13 and oxygen-18 are also being measured. We are specifically looking for a slow-flowing groundwater stream sealed off from above, where we can take a series of groundwater samples, identical in type and origin, but different in age. The tritium measurements then make it possible to determine flow rate and age over a time span of about 50 yr. Under favourable conditions it should be possible to extend the total age range by extrapolation downstream and thus arrive at a check for the carbon-14 age. The difficulty is that the groundwater stream in question has to be very well sealed off from above to exclude infiltration of younger water from the overlaying strata, since even slight infiltration could make the tritium ages far too young. This is especially true at the far end of the age range covered by tritium, where one deals with the natural tritium level, while the bomb-tritium level in the water layers above is one or even two orders of magnitude higher. As far as this difficulty of bomb-tritium contamination is concerned, a hydrologically homogeneous vertical groundwater body might be preferable for an age profile, but in this case other disadvantages have to be taken into account.

In Fig. 1 the general trend of the recent variations of bomb-tritium in rain is shown. The diagram is based on measurements by BROWN [1] and taken in part from a tritium list drawn up by the International Atomic Energy
A COMPARISON OF C¹⁴ AND TRITIUM AGES

Fig. 1

Yearly averages of tritium in precipitation [1]
Left hand scale, solid line: Ottawa.
Right hand scale, solid line: presumable tritium level in Central Europe (see text).
Right hand scale, broken line: direct measurements (Heidelberg).

Agency [2]. This data for Ottawa can be used as a basis for a preliminary estimate of the Central European situation where the tritium record for the years prior to 1959 is more fragmentary. Since then, however, simultaneous measurements [3, 4] have shown that it is probably justifiable to divide the Ottawa values of the average tritium concentration in rain by a factor of 1.5 to 1.8 to obtain a rough guess of the corresponding trend in Central Europe.

As is to be seen in Fig. 1 the picture is fairly complicated. This is true even if one takes yearly averages as we did here to avoid the considerable seasonal variations [3, 4]. To arrive at a single and simple age figure for the groundwater the following procedure has been adopted. One asks how large the maximum contribution of an individual year's precipitation to the (measured) tritium content of the groundwater sample in question can be, if one assumes that the rest of the groundwater sample is old water which contains none or a negligible amount of tritium. In doing this one obtains a new histogram, which (if the measured tritium content is 20 tritium units (T.U.)* for example) has low figures for the very recent years (3% for a possible 1962 contribution, 20% for 1961, and so on), then rises with increasing age and finally reaches 100% if the contribution of that year is equal

* A tritium unit (1 T.U.) corresponds to a concentration ratio T/H = 10⁻¹⁸.
to or lower in activity (corrected for radioactive decay) than the ground-water sample measured. This histogram can then be well approximated by a smooth curve which passes the 50% level at a certain age. Taking this age we say that at least half of the water is older than this age, and therefore the figure gives the minimum age of the main part of the water. This age is given in the last column of Table I for a set of groundwater samples thought to be fairly recent. The last two columns contain the measured tritium content and the C\textsuperscript{14} content respectively, and are discussed later.

Perhaps a few words beforehand about the justification of smoothing the tritium rain data over at least a year to get the input function for the groundwater contribution would be appropriate. A fairly large number of shallow wells and lysimeters have been sampled regularly for deuterium analysis by BRINKMANN et al. [5]. He found that the variations, if any, are very small, despite the fact that deuterium in rain varies considerably with the season. This was astonishing at first, at least for most of the lysimeters (only one of the lysimeters studied so far, containing pure sand, showed a definite seasonal variation), since the rainwater only has to pass a few meters of soil, while considerable smoothing can be expected under natural conditions because of the differences in length of the paths from the infiltration site to the mouth of the well. However, it is quite plausible that the mean residence time for the water in the upper few meters of the soil is of the order of years, at least for soils with a small grain size since the amount of water stored there by adsorption and capillarity is by no means small compared with the yearly seepage water contribution to the groundwater.

A similar picture is obtained for the tritium content in the waters from lysimeters. The water given off by a lysimeter with a top layer of loss did not show any response to the large 1962 bomb peak until July, i.e. for at least half a year. Its tritium content has remained fairly constant (between 100 and 130 T.U.) since mid-1959. In view of these results it would appear that the content of tritium in river water at present would provide useful information on the contribution of direct run-off to the river if the average activity level of the groundwater were assayed separately, at least roughly, since the tritium content of this contribution could be an order of magnitude higher than that of the groundwater.

As an initial step in our programme we re-measured the C\textsuperscript{14} content in new samples from the same wells used in the first study of the C\textsuperscript{14} content in groundwater in 1956 [6] and at the same time measured the tritium content of these new samples. The results are given in Table I. In all cases the new results are within the experimental error identical with the earlier ones*. It can be seen that the C\textsuperscript{14} content depends slightly on the surface geology of the area from which the samples come. It is lowest in the shell-lime area, higher in the Jura, and highest in the Rhine valley. This has been stated earlier, but now it can be excluded as a possible age effect, since exactly those wells with the highest C\textsuperscript{14} content are the ones with the lowest tritium content. This is plausible, however, if one assumes that the slow passage of water through the uppermost soil layers containing humus

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* The earlier C\textsuperscript{14} measurements [6] were given relative to wood about 10 yr old. Plant material of that time is 3% lower than the standard used here, as a result of the industrial combustion of fossil fuels. Thus 3% was to be subtracted from the values published in 1957.
# Table 1

<table>
<thead>
<tr>
<th>Location of well</th>
<th>Sampling date</th>
<th>C(^{14}) content (%)*</th>
<th>Tritium content (T. U., ± 5%)</th>
<th>Minimum T age (yr)</th>
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</thead>
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<tr>
<td>Shell-lime area, 50 km E of Heidelberg</td>
<td>March 1956</td>
<td>70.5 (b)**</td>
<td>-</td>
<td>-</td>
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<tr>
<td>Osterburken Pumpwerk</td>
<td>Nov. 1962</td>
<td>70.0 (a)</td>
<td>13.8</td>
<td>&gt; 8</td>
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<tr>
<td>Unterschefflitz Kreuzwiesenquelle</td>
<td>March 1956</td>
<td>75.0 (c)</td>
<td>-</td>
<td>-</td>
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<tr>
<td></td>
<td>Nov. 1962</td>
<td>74.7 (a)</td>
<td>23.7</td>
<td>&gt; 6</td>
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<tr>
<td>Neckar valley, 30 km E of Heidelberg</td>
<td>March 1956</td>
<td>65.8 (c)</td>
<td>-</td>
<td>-</td>
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<tr>
<td>Eberbach Aubrunnen</td>
<td>Nov. 1962</td>
<td>69.0 (a)</td>
<td>34.4</td>
<td>&gt; 4</td>
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<tr>
<td>Rhine valley (loess area) near Heidelberg</td>
<td>Jan. 1956</td>
<td>81.9 (b)</td>
<td>-</td>
<td>-</td>
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<tr>
<td>Heidelberg Großmarkthalle</td>
<td>Dec. 1962</td>
<td>83.3 (a)</td>
<td>3.0**</td>
<td>&gt; 20</td>
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<tr>
<td>Mannheim - Rheinau</td>
<td>March 1956</td>
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<td>-</td>
<td>-</td>
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<tr>
<td></td>
<td>Oct. 1962</td>
<td>82.2 (b)</td>
<td>3.9</td>
<td>&gt; 20</td>
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<td>May 1958</td>
<td>88.3 (b)</td>
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<td>-</td>
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<td></td>
<td>Oct. 1962</td>
<td>89.8 (a)</td>
<td>5.5</td>
<td>&gt; 10</td>
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<td>Location of well</td>
<td>Sampling date</td>
<td>C$^{14}$ content (%)*</td>
<td>Tritium content (T. U., ±5%)</td>
<td>Minimum T. age (yr)</td>
</tr>
<tr>
<td>------------------</td>
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<td>---------------------</td>
</tr>
<tr>
<td>Schwabischer Jura (karst area), 60 km SE of Stuttgart</td>
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<tr>
<td>Blautopf (shallow karst)</td>
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<td>79.2 (b)</td>
<td>-</td>
<td>&gt; 4</td>
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<td>Jan. 1962</td>
<td>83.8 (c)</td>
<td>40.0</td>
<td>&gt; 4</td>
</tr>
<tr>
<td></td>
<td>June 1958</td>
<td>81.0 (b)</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Jan. 1963</td>
<td>83.0 (b)</td>
<td>29.6</td>
<td>&gt; 5</td>
</tr>
</tbody>
</table>

* Given in per cent of natural level of C$^{14}$ in plant material [7]

** rms error
(a) = ±0.5%  
(b) = ±0.8%  
(c) = ±1.0% to 2.0% 

and rootlets and therewith sources of plant CO₂ raises the C₁⁴ content of the water towards the level of the plant CO₂.

The measurements listed in Table I are the first step in the programme outlined above and of course they cannot serve as a new argument for the validity of the C₁⁴ dating method for groundwater. ERIKSSON [8] recently questioned this validity. He was able to show that loss of C₁⁴ in groundwater from an exchange with an aquifer containing carbonates might be possible without a simultaneous change in the C₁³ content.

If the solid-state diffusion in the carbonate were sufficiently fast and the contact time between water and solid carbonate sufficiently long, the C₁⁴ indeed could be lost by exchange with deeper parts of the carbonate surface layer, already in equilibrium with the groundwater, but old enough so that the C₁⁴ brought in by preceding exchange would already be lost by radioactive decay. This, however, would not be true for the stable C₁³ and therefore the C₁⁴ content would be altered, but not the C₁³.

The exchange in any case must be of the diffusion type. This is true for the actual solid-state diffusion as well as for any process of precipitation and re-dissolution of carbonate. Consequently the loss of C₁⁴ should be rapid in the beginning and much slower later on, since the spread by diffusion is proportional to the square root of time or shows similar behaviour. This fact makes a double tracer (artificial C₁⁴ and tritium) experiment as proposed by ERIKSSON [8] very sensitive; the retention of C₁⁴, if any, would show up clearly even if the C₁⁴ loss in a steadily flowing groundwater stream were tolerable. The tracer experiment is a non-equilibrium situation and C₁⁴ can exchange with the top surface, while in a steady-state the surface is probably in equilibrium in any case and an effective exchange at least involves deeper layers of the carbonate grains.

Double tracer experiments of the type mentioned have been carried out in recent years during nuclear weapons testing. Figure 2 shows the recent increase of C₁⁴ activity in plant material [7] caused by atom bombs (broken line). The solid line drawn through the measured points gives the C₁⁴ increase in soil CO₂. The CO₂ was collected by covering a patch of soil in a wood with the open side of a large barrel and putting an alkali absorber underneath for a few days. CO₂ given off from the soil follows the C₁⁴ content of plants with a delay of only a few years. Sooner or later, therefore, a similar increase should show up in groundwater. It can be seen from the data in the Table that in view of the considerable tritium ages of these "recent" groundwaters one at present can expect only a very slight increase in the C₁⁴ content from the bombs. In fact the 1962 measurements of C₁⁴ in the wells listed in general are slightly higher than the earlier ones. This increase, however, is not outside the statistical error, at least not if one only looks at individual samples separately. If the C₁⁴ content of the wells studied does increase within the next few years this then would mean that the exchange between water and solid carbonate and the corresponding C₁⁴ loss certainly is not serious. Otherwise a considerable delay of the C₁⁴ increase must occur as outlined above. If on the other hand, a delay is found, this might also result from the fact that the CO₂ dissolving the lime in the soil must not necessarily have the same age distribution as the CO₂ which leaves the soil surface; it might be a bit older because of the age stratification of the soil organic matter.
Fig. 2

Bomb C¹⁴ increase in percent above the natural level Central Europe
Broken line: in atmospheric CO₂.
Solid line: in CO₂ given off from the soil. Open dots: sandstone area.
Full dots: loess-loam area.

In any case it can be hoped that investigations of the kind reported here will provide strong arguments regarding the usefulness of the C¹⁴ method. It should be pointed out that the method is probably useful only if the exchange with solid carbonate is indeed negligible. Otherwise the "apparent" age will depend to too large an extent on local conditions, such as carbonate content, grain size of the aquifer, bicarbonate concentration in the water, etc.

REFERENCES


DISCUSSION

D.B. SMITH: I should like to congratulate Dr. Vogel and Dr. Münnich on developing this very interesting C¹⁴ method of dating groundwater and
giving us an additional technique which will be useful in the determination of groundwater parameters that have not been available hitherto.

However, the problem of C\textsuperscript{14} loss in groundwater is still an important one and I should like to comment on the suggestion for a double tracer investigation with tritium which was originally put forward by Dr. Eriksson and which is discussed by Dr. Münich.

First, the problem is not essentially one of solid-state diffusion because allowance can be made for any losses arising from this effect using C\textsuperscript{13}. Any loss of C\textsuperscript{14} at natural concentrations will not be rapid initially in an aquifer where equilibrium between C\textsuperscript{14} and the rock surface is already established. The different behaviour of C\textsuperscript{13} and C\textsuperscript{14} in the system is essentially a result of the decay of the C\textsuperscript{14}.

One method of investigating the losses of C\textsuperscript{14} which may occur in groundwater is to consider the total C\textsuperscript{14} content of the rock owing to C\textsuperscript{14} exchange and diffusion in a system where equilibrium is established. Let the total C\textsuperscript{14} activity in the rock be \( A_{\text{total}} \). Then the loss of this C\textsuperscript{14} owing to radioactive decay in time \( T \) will be

\[
A_{\text{total}} (1 - e^{-\lambda T}).
\]

(1)

In order that \( A_{\text{total}} \) should remain constant (equilibrium conditions) the loss expressed by (1) must be made good by C\textsuperscript{14} from the water in the system and this expression determines the loss of C\textsuperscript{14} which will occur.

Considering a slowly flowing system, the total amount of C\textsuperscript{14} carried into the system during this time \( T \) is \( QT S \),

where \( Q \) is the water flow of the system; \( T \) is the same time as in (1); and

\( S \) is the specific activity of the water (C\textsuperscript{14}).

C\textsuperscript{14} from this source will be lost into the rock to replace the C\textsuperscript{24} which is decaying according to expression (1).

The relative value of (1) and (2) is the important quantity which determines errors in dating arising from carbon exchange with the rock.

If (1) \( \ll \) (2) no correction will be necessary for C\textsuperscript{14} loss in the rock.

If (1) < (2) but of comparable value, error will be introduced and too high an apparent age will be obtained.

Unfortunately, it is impossible to determine \( A_{\text{total}} \) by straightforward measurement. Age measurement in a suitable aquifer where an age value can be obtained with tritium and also with C\textsuperscript{14} may give an estimate of \( A_{\text{total}} \) although since such an experiment cannot measure beyond about 50 years it will not be highly sensitive.

The double artificial tracer investigation using a pulse injection suggested by Eriksson, could yield information on the surface exchange between the rock and water and possibly on diffusion of C\textsuperscript{14} into the rock. From this, some information regarding \( A_{\text{total}} \) might be obtained. In order that future C\textsuperscript{14} work in the same aquifer is not excluded, such an experiment could alternatively be carried out with C\textsuperscript{13} and tritium.

K. O. MÜNNICH: I think that the double tracer experiment would be very sensitive, since the C\textsuperscript{14} loss for the extra C\textsuperscript{14} would be higher than in the case of the steady state, because of the diffusive character of the exchange. The technique gives you a limiting value: if both isotopes arrive
at once, you can safely conclude that there is no considerable loss to an additional reservoir. On the other hand, if you find apparent retention, you cannot make any hard and fast conclusion.

J. C. VOGEL: The loss of C$^{14}$ is serious only if it takes place faster than the radioactive decay, an occurrence which can be checked by tritium measurements.

A. NIR: If the delay of the C$^{14}$ front is really only of a few years there is still an open problem. Exchange processes may have, however, delay effects expressed as fractional velocities. These may be small, i.e. a factor of four or ten, compared to the groundwater velocity. How do you account for these effects?

K. O. MÜNNICH: To a first approximation, the CO$_2$ given off by the soil can be considered to be the same as that which forms the bicarbonate in the groundwater. If this is true, the C$^{14}$ content in the groundwater bicarbonate should have increased. If this is the case, and you have a well whose water is known (by tritium dating) not to be very old, it could be expected, since C$^{14}$ is a conservative component of the water, that the excess C$^{14}$ would appear within the next few years. Observation of an increase in the C$^{14}$ of the groundwater in the near future would be proof of the argument.

J. C. VOGEL: I would not expect the C$^{14}$ content in groundwater to rise in the near future, because the CO$_2$ responsible for the dissolution of limestone presumably does not come from the uppermost organic matter.

A. NIR: I also have a question in connection with the paper presented by Dr. Vogel. The error given for the coastal data in Fig. 6 is very small. Is this only the statistical error?

J. C. VOGEL: Yes. The uncertainty in the initial concentration is not included. It amounts to a maximum of ±3000 yr.
Abstract — Résumé — Аннотация — Resumen

A SURVEY OF THE NATURAL ISOTOPES OF WATER IN SOUTH AFRICA. A preliminary survey of the isotopes in fresh water from South Africa has been made. The semi-arid climatic conditions cause the isotopic composition of rain, river and groundwater to differ considerably from that in Europe and the northern United States. Results show that evaporation markedly changes the isotope ratio of the precipitation. Discontinuous recharge of the groundwater supplies is probably responsible for the observed deviations in this reservoir.

An understanding of these different factors has made it seem justifiable to tackle the more specific hydrological problem of subterranean flow-rates with natural tritium. Samples from a series of wells at increasing distances from the catchment area all give ages > 30 yr.

ÉTUDE DES ISOTOPES NATURELS DE L'EAU EN AFRIQUE DU SUD. Les auteurs se sont livrés à une enquête préliminaire sur les isotopes contenus dans de l'eau douce provenant d'Afrique du Sud. En raison des conditions climatiques (climat semi-aride), la composition isotopique des eaux de pluie, des eaux fluviales et des eaux souterraines est nettement différente de celle que l'on trouve en Europe et dans la partie nord des États-Unis. L'évaporation modifie notablement la composition isotopique des précipitations. La discontinuité du renouvellement des eaux souterraines est probablement la cause des différences observées.

Pour comprendre le rôle de ces différents facteurs, on a jugé utile de s'attaquer plus particulièrement au problème hydrologique des vitesses d'écoulement des eaux souterraines, à l'aide du tritium naturel. Des échantillons prélevés dans une série de puits de plus en plus éloignés de la zone de captage ont donné des âges supérieurs â 30 ans.

ИССЛЕДОВАНИЕ ЕСТЕСТВЕННЫХ ИЗОТОПОВ В ВОДАХ ЮЖНОЙ АФРИКИ. Было проведено предварительное исследование изотопов в пресных водах в Южной Африке. Из-за подусловного климата изотопный состав дождевых, речных и грунтовых вод значительно отличается от того, который встречается в Европе и на севере США. Результаты показывают, что испарение заметно изменяет изотопный коэффициент в осадках. Непостоянный сток вероятно является причиной наблюденных отклонений в этих бассейнах.

Учет всех этих факторов делает обоснованной необходимость решения более специфических проблем исследований грунтовых вод путем измерения содержания трития. Образцы из ряда колодцев, находящихся на различных расстояниях от места отвода, показали возраст воды 30 лет.

ESTUDIO DE LOS ISOTOPOS NATURALES DEL AGUA EN SUDÁFRICA. Los autores han realizado un estudio preliminar de los isótopos del hidrógeno presentes en diversas aguas dulces sudafриcanas. Las condiciones climáticas semiáridas hacen que las composiciones isotópicas de la lluvia, así como de las aguas fluviales subterráneas difiera en grado apreciable de las que se observan en Europa y en el norte de los Estados Unidos. Los resultados muestran que la composición isotópica que existía en el momento de la precipitación es alterada considerablemente por evaporación. Las diferencias observadas en este depósito se deben probablemente a la recarga intermitente de las reservas de aguas subterráneas.

De acuerdo con los autores, la comprensión de estos diversos factores justifica que se aborde el estudio del problema hidrológico más concreto de los caudales de las corrientes subterráneas mediante el uso del tritio natural. Las muestras extraídas de una serie de pozos situados a distancias crecientes de la cuenca colectora indican sin excepción edades superiores a los 30 años.
INTRODUCTION

To gain a better understanding of the factors governing the isotope content of fresh water in South Africa, samples have been collected from widely separated areas for isotopic analysis. The purpose of this paper is to report some of the findings so far made. In conclusion the tritium ages of some groundwater samples will be discussed briefly.

The isotopic composition of terrestrial water is strongly dependent on climate, so that the conclusions for Europe and the northern United States, for which the most data is available, do not necessarily apply to South Africa. The greater part of the country has a warm dry climate with considerable water deficiency. The only regions where precipitation exceeds evaporation (e.g. where there is an annual water surplus) are the mountains forming the Eastern Escarpment (Drakensberg) including most of Natal, and some isolated areas along the south coast [1]. The principal rivers consequently all have their origin in these mountains and the adjoining highlands.

Rainfall shows a strong dependence on season and, except for the Western Cape Province, reaches its maximum in summer. The average annual precipitation on the inland plateau ranges from less than 10 cm on the west coast to some 70 cm in the east. The rains occurring in the inland are mainly of the convective type; true frontal rains falling relatively seldom.

As for the groundwater, no real water table exists and movement takes place chiefly along bedding-planes and fissures. The supplies, however, must be quite large judging from the fact that the yields are often practically independent of the seasonal variation in precipitation.

THE SAMPLES AND RESULTS

During the summer of 1959-60 the daily precipitation was collected at six different meteorological stations: Maun, Pretoria, Bloemfontein, Escourt, Durban and East London. The localities are shown on the map (Fig. 1). Samples for analysis were selected from five rain periods. Rivers have been sampled at different places and at different times of the year. Almost the entire course of the Orange and Vaal rivers as well as their tributaries have been covered. Further samples come from the Olifants river which drains a large portion of the Transvaal and several rivers along the east coast. The localities from which groundwater samples have been obtained are denoted by the small numbers on the map. They represent three regions:
(i) The semi-arid to arid regions of South West Africa and the North Eastern Cape Province;
(ii) The catchment area of the Orange; and
(iii) The coastal belt.

The results of the deuterium analysis given collectively in Fig. 2. Rainwater shows the largest scatter, with a considerable portion of the samples being heavier than ocean water. This is somewhat unexpected, especially for the inland stations like Maun and Pretoria. In a previous paper [2] it has been shown that the high deuterium contents are a result of evaporation
Fig. 1

Map of South Africa

(i) Stations where rainfall was collected
(ii) Principal rivers
(iii) Localities from which groundwater samples were obtained (small numbers)

Fig. 2

Deuterium content of water samples from South Africa
Expressed in percent deviation from standard mean ocean water (SMOW)
of the raindrops during their fall to the ground. As the heavy isotopic molecules are less volatile, they become enriched in the remaining liquid. The largest isotopic enrichment was found to occur in light showers and thunderstorms which fell on hot dry days, while enduring rain with high total precipitation tended to be "lighter" and presumably more representative of the moisture in the clouds.

The two coastal stations Durban and East London do not show the large variations of the other stations, the $\delta$-values clustering about 0%. Here apparently, evaporation only has a small influence on the isotopic ratio. This is probably due to the high relative humidity of the air and the low average altitude of the clouds. The fact that the deuterium content is similar to that of ocean water means that the depletion caused during evaporation of the moisture from the sea is just compensated for by the enrichment during condensation in the cloud.

In Fig. 3 the correlation between deuterium and $\text{O}^{18}$ in individual samples is shown. For comparison the results for precipitation at Sindorf, Federal Republic of Germany, are also plotted. The Sindorf samples lie on a straight line with a gradient of 8, showing that the fractionation processes involved take place under equilibrium conditions [3, 4]. The higher values for deuterium and $\text{O}^{18}$ and the more gradual gradient of about 5 for the South African rain indicates fast non-equilibrium evaporation.

The weighted average for the deuterium concentration in all the rainwater samples is markedly lower than the ordinary mean, because, as stated above, the low concentrations are associated with heavy precipitation. To
be able to compare the meteoric water with rivers and groundwater it is necessary to know the mean annual deuterium content. This can be deduced for Pretoria rain from total monthly samples published by ÖSTLUND [5]. We obtain a value of $\delta D = -1.6\%$. The average of all the river-water samples is $-1.3\%$, and does thus not differ significantly from the precipitation. This conclusion is admittedly only tentative, because it assumes the Pretoria rain to be representative of a large area. It thus appears that, although considerable evaporation takes place after the rain reaches the ground, it is not subject to marked fractionation, probably because the evaporation in the soil is largely quantitative. Lysimeter samples from Sindorf have led to the same conclusion; the drainage water has practically the same isotope ratio as the weighted average of the precipitation (about $-6\%$)[6].

The individual rivers show no large variations along their course, and with time. There are, however, slight differences from river to river, but on the whole these are small.

The groundwater throughout contains considerably less deuterium than the rain or river waters. The average is $-3.8\%$. There is also not much dependence on locality, although the coastal samples are definitely somewhat "heavier". The important conclusion which can be drawn from these results is that the groundwater supply is not replenished continuously from the precipitation, but mainly represents water from occasional heavy downpours.

UNDERGROUND FLOW RATES

Following this brief survey of the distribution of the heavy stable isotopes in the natural waters of South Africa, mention shall be made of an attempt to determine underground flow rates with the aid of natural tritium. This investigation is being carried out in collaboration with the Hydrological Research Division of the Department of Water Affairs, Pretoria. The region selected is north-west of Kuruman beyond an extensive range, which presumably forms the catchment area. To the north-west of this range the

<table>
<thead>
<tr>
<th>No.</th>
<th>Farm</th>
<th>Date</th>
<th>D (%)</th>
<th>$O^{18}$ (%)</th>
<th>T (T.U.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>W 9a</td>
<td>Begin</td>
<td>27.11.1961</td>
<td>-3.6</td>
<td>-5.7</td>
<td>&lt; 0.5</td>
</tr>
<tr>
<td>W 9b</td>
<td>Namysin</td>
<td>28.9.1960</td>
<td>-3.4</td>
<td>-3.4</td>
<td>0.85 ±0.25</td>
</tr>
<tr>
<td>W 9c</td>
<td>Sonnestraal</td>
<td>26.9.1960</td>
<td>-4.5</td>
<td>-6.8</td>
<td></td>
</tr>
<tr>
<td>W 9d</td>
<td>Geluksoord</td>
<td>24.11.1961</td>
<td>-4.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>W 9e</td>
<td>Uithou</td>
<td>28.11.1961</td>
<td>-4.4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>W 9f</td>
<td>van Zyl's Rust</td>
<td>27.9.1960</td>
<td>-4.5</td>
<td></td>
<td>0.50 ±0.25</td>
</tr>
</tbody>
</table>
Kalahari desert extends for hundreds of miles. The absence of perennial rivers and the low rainfall presents a relatively simple hydrological situation, at least as far as the origin of the water is concerned. Samples were collected from wells at increasing distances from the catchment area over a distance of some 100 km. The results hitherto obtained are presented in Table I. None of the three samples which have been analysed contain significant amounts of tritium and the water must thus be considered older than 40 yr.

One of the samples (Namysin) definitely contains tritium which either means that the reservoir has an average age of about 30 yr or that a small amount of recent precipitation (about 1%) containing tritium from nuclear tests has been admixed to the supply. From the results it can, however, definitely be concluded that the re-charge and flow rate are slow compared to the half-life of tritium.

REFERENCES


DISCUSSION

E. TONGIORGI: Could you tell us how the rain samples were collected?
J.C. VOGEL: You are probably wondering about evaporation of the rain samples after they had fallen into the collector. The samples were all collected at meteorological stations where it was known that evaporation was not extensive. Of course, there can be slight evaporation of the precipitation in the rain gauge, and this is something that meteorologists are interested in as well.

E. TONGIORGI: But do the figures represent all the precipitation or only part of it?
J.C. VOGEL: They represent all the precipitation in a 24-h period. That is to say, the samples were collected daily at 8 a.m. and, where one period of rain continued over two days, for example, there were two samples for it. In the case of short showers occurring several times in one day, there was a single collective sample representing them all.

C.V. THEIS: What were the depths of the wells from which the groundwater samples analysed from tritium were taken?
J.C. VOGEL: The depths ranged from 60 to 100 m. The well from which the Namysin sample was taken and which was the only one definitely containing tritium, was among the deepest.
E. TONGIORGI: I think that it might be appropriate at this point to comment on some measurements of stable isotopes that we have performed. There is need to emphasize the possibility of using stable isotopes as natural tracers in groundwater research. This technique is applicable in the case of bodies of water having differences in isotopic composition. These differences may result from the following:

(1) Differences in geographic origin. The lighter isotopes prevail in precipitation which falls in a higher geographic latitude as well as at a higher altitude. The latter factor is of great importance, since it is responsible for the presence of isotopically different water even within a region of small size. In Tuscany, for instance, the altitude factor causes the water of the Serchio river, which has a mountainous watershed, to be lighter (averaged over a whole year, \( \delta^{18}O = -7.6 \) per mille lower in \( O^{18} \) than standard mean sea water: \( \delta^{18}O = -7.6 \)) than the water of the Era river, which flows in hilly country \( (\delta^{18}O = -6.4) \). Consequently, the waters flowing in the alluvial sediments of the rivers can be recognized when they supply the groundwater in the coastal plain, since they differ in their isotopic composition.

(2) Differences in evaporation which the waters may have undergone. Some lakes in central Italy have water with a high \( O^{18} \) content: e.g. Lake Trasimene: \( \delta^{18}O = +2.0 \); Lake Bracciano: \( \delta^{18}O = +0.7 \). When the question arose whether certain wells in the vicinity of Lake Bracciano were fed by that lake, it was easily shown that this was not the case, since the figure for the water from the wells was \( \delta^{18}O = -6.0 \), which is that for the average composition of rain-water in that area.

(3) Different contributions of the rains falling at various periods during the year (seasonal effect). In central and southern Italy the summer is a dry period. The autumn rains are largely absorbed by the soil and do not cause much of an increase in river flow. Conversely, winter and spring precipitation, which falls on saturated soils, causes a sudden and large increase in the discharge of the rivers. The light isotopic composition of the river water then obviously reflects the composition of the precipitation on the watershed during the preceding period. However, during relatively dry periods in these seasons, the isotopic composition of the river water tends to reach a value which is the same as that found in autumn rains, but very light water may be added from melting snow. Variations of more than 5% in the content of \( O^{18} \) are observed in the water of the rivers of Tuscany and these may be even larger under other climatic conditions. It is this state of affairs which enables us to get information on the various contributions to the flow of a river.

The application of isotopic methods may be more difficult in the case of groundwater, owing to the mixing of the water in the soils. However, this is not everywhere the case. Professor Cotecchia and I are now studying a large karst area, the Salento peninsula. Our information about the aquifer in this area gives a picture of a nearly ideal and simple groundwater pattern. Cretaceous limestone, going down to remarkable depths below sea level, is the main rock in the region. The limestone is strongly fissured and allows easy infiltration of rain water almost everywhere on the soil surface, so that rivers are completely absent. The limestone retains its karstic character down to a level of more than 100 m below the present sea level. In the deep fissured limestone, sea water is present as an underground link be-
between the Adriatic and Ionian Seas. Throughout the peninsula, which has an average width of about 40 km, the sea water is found at depths which depend on the height of the fresh-water table with respect to sea level.

Sampling was carried out during the period 12-17 November 1962. This consisted of taking water samples from springs and from wells both in and out of use (stations for the measurement of water-table levels). In the latter, samples were — as far as possible — drawn from different depths and for as long as such sampling could be considered significant with regard to the stratification of the water in the aquifer.

The salt content in the water samples ranged from very high levels in those taken from the diffusion zone to very low, but nevertheless significant, ones in those from the upper section of the aquifer. In a diagram in which the d\textsubscript{018} values for the samples are plotted against their Cl" content, almost all the points fall within a narrow triangular area, having as its base (Cl" = 0) the interval between d\textsubscript{018} = -5.6 and d\textsubscript{018} = -6.5, and as its vertex the point with co-ordinates Cl" = 22 g/l (average Cl" content of the Adriatic and Ionian Seas) and d\textsubscript{018} = +1.3 (measured in a sea water sample taken 4 km off Bari at a depth of 70 m and representing the O\textsuperscript{18} enrichment from the high evaporation in the water of this part of the Mediterranean). A figure (d\textsubscript{018} = -6.0) sufficiently close to the mean value of the above-mentioned interval, can be assumed to be the average value for the meteoric waters which supply the aquifer.

Regardless of the time of year at which the precipitation occurred, we shall consider as "summer" waters all those whose d value is higher than the upper limit of the above-mentioned interval (d\textsubscript{018} > -5.6), and "winter" waters all those with d\textsubscript{018} = < -6.5. Actually, in part of the area we have been considering, the uppermost section of the groundwater was made up of "summer" water (d\textsubscript{018} values sometimes as high as -3.6 to -4.0), indicating a certain delay in groundwater recharge. A very different picture was found in the same day's sampling in zone about 100 km north (Tavoliere di Foggia). Here the wells reach a superficial aquifer having Pliocene clays as its basis. The recharge of the aquifer is only possible in the same area at 100-150 m above sea level and must be very rapid. The samples taken at water-table level were made up of "winter" water, sometimes as light as d\textsubscript{018} = -8.1. With increasing depth there was always a trend towards d\textsubscript{018} = -6.0.

The variations in d with depth, observed in wells where the upper layers were seasonally traced, indicates in all cases a mixing with increasing amounts of water of average isotopic composition. This is in agreement with the recharge of the aquifer from the surface, and indicates that the zone below the surface zone is made up of water in which an average d value has been obtained as a result of continuous superimposition of strata of water seasonally traced. In time, these lose their individuality, thus forming water of uniform isotopic composition. Evidence of this was found in a well near Copertino, which had been drilled for research purposes by the Geomining Division of the National Nuclear Energy Committee. Samples were taken here in the course of the drilling. The upper levels were found to contain water of "winter" composition; a layer of water influenced by summer rainfall was still present a few meters below; and, with increasing depth, the
d values became constant down to the level where the diffusion zone was reached.

Under the conditions outlined above, a single set of samples, taken during the transition season, enabled us to identify various hydrological situations. It is clear that the evolution of groundwater stratification as a function of time can be followed and useful information thus obtained for the solution of problems which it has not been possible to tackle thus far, such as the quantitative study of recharge and flow of groundwater.

J.C. VOGEL: At first glance, the isotopic variations in the karst groundwaters found by Professor Tongiorgi seem to be at variance with the results presented by Dr. Münnich. However, this is obviously owing to different geological and climatic conditions. In the Federal Republic of Germany, our lysimeters indicated no seasonal variations, even at depths of 2-3 m, whereas in southern Italy variations are found at depths of as much as 20 m. But in the latter case, we are dealing with a karst region where the water probably sinks below the water-table level within a very short time. Considering the differences in substrata, the measurements are not contradictory.

C.V. THEIS: Professor Tongiorgi is to be congratulated on the thorough study of the hydrological factors involved in his measurements. I would like to add a few words about a phenomenon which could affect tracer studies of groundwater in coastal areas and which has been hinted at by a number of preceding speakers. It is found that whenever, owing to repeated fluctuations from tides or seasonal recharge, the interface between fresh water and sea water becomes diffuse, the salt water mixes to varying degrees with the fresh water, circulates, and is discharged into highly saline springs. In certain limestone aquifers of Florida, this circulation of sea water has been shown to amount to 10% of the fresh-water flow. In coastal areas, therefore, it is quite possible for some of the tracer which has been introduced into a fresh-water stream to discharge into quite saline water.

A. NIR: In closing this session, I would like to congratulate the authors of the papers presented for making a significant and fundamental contribution to dating and tracer methods.
THE RUNOFF OF FALL-OUT RADIOSTRONTIUM FROM A RIVER AND ITS APPLICATION IN HYDROLOGY

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Abstract — Résumé — Аннотация — Resumen

THE RUNOFF OF FALL-OUT RADIOSTRONTIUM FROM A RIVER AND ITS APPLICATIONS IN HYDROLOGY.

Measurements were made of the concentration of Sr$^{85}$ and Sr$^{90}$ in river waters in May 1962. The difference of the radionuclides in their half-lives (51 d for Sr$^{85}$ and 28 yr for Sr$^{90}$) but identical chemical properties and the mechanisms by which they are supplied to land surface were considered to give some information on the pattern of water balance between precipitation and the runoff from the river.

The ratio of Sr$^{89}$ to Sr$^{90}$ in the precipitation is a function of time if the series of nuclear bomb testings by the USSR is assumed a single shot in the autumn of 1961. The measured value of the ratio was approximately 15 in the precipitation at the time of river water sampling, whereas the ratios in the river water showed smaller values, at three stations downstream along the river "Tonegawa", respectively 11, 8.1 abd 7.9.

The ratio of Sr$^{89}$ to Sr$^{90}$ in the cumulated ground deposits which were originated from older debris is approximately zero, because Sr$^{89}$ decayed out of its short half-life. Therefore, the lower values in the ratios in river water are explained by the mixing of the surface runoff of radiostrontium from the fresh debris and that from the older debris which flows into the river by leaching with groundwater.

MESURE DU RADIOSTRONTIUM DES RETOMBÉES DANS LES COURS D'EAU ET SON APPLICATION EN HYDROLOGIE. Les auteurs ont mesuré la concentration de Sr$^{85}$ et Sr$^{90}$ dans les eaux fluviales et la neige en mai 1962. Le fait que ces radionucléides ont des périodes différentes (51 jours pour Sr$^{85}$ et 28 jours pour Sr$^{90}$) tout en possédant les mêmes propriétés chimiques, et les processus par lesquels ils sont fournis au terrain devraient permettre d'obtenir des renseignements sur le bilan hydraulique notamment sur l'origine des eaux, précipitations et cours d'eau.

Le rapport Sr$^{89}$/Sr$^{90}$ dans les précipitations est fonction du temps si la série des essais nucléaires exécutés par l'URSS est considéré comme une seule explosion ayant eu lieu à l'automne de 1961. A l'époque du prélevement des échantillons d'eau fluviale, ce rapport était égal à 15 environ dans les précipitations, alors que dans les échantillons d'eau de la «Tonegawa», prélevés en trois points successifs dans la direction aval, sa valeur n'était que de 11, 8,1 et 7,9 respectivement.

Dans le dépôt accumulé dans le sol à la suite de retombées anciennes, le rapport Sr$^{85}$/Sr$^{90}$ était sensiblement égal à 0, le Sr$^{89}$ s'étant désintégré en raison de sa courte période. Ainsi, les valeurs plus faibles du rapport pour l'eau fluviale s'expliquent par le fait que le radiostrontium des retombées récentes contenu dans les eaux de ruissellement était mélangé au radiostrontium des retombées anciennes amené au cours d'eau par les eaux souterraines après l'lixivation.

ПЕРЕНОС ВЫПАДЕНИЙ РАДИОАКТИВНОГО СТРОНЦИЯ РЕКАМИ И ЕГО ПРИМЕНЕНИЕ В ГИДРОЛОГИИ. В мае 1962 года измерялась концентрация Sr$^{85}$ и Sr$^{90}$ в речной воде. Различные периоды полуразпада этих изотопов (51 день для Sr$^{85}$ и 28 лет для Sr$^{90}$) но идентичные химические свойства и механизм попадания на поверхность земли могут дать некоторую информацию относительно равновесия между водой, выпадающей в виде осадков, и водой, уносящейся реками.

Соотношение Sr$^{85}$ и Sr$^{90}$ в осадках выражается в функции времени, причем серия испытаний ядерных бомб, проведённая Советским Союзом, рассматривается как единое испытание осенью 1961 года. Измеренная величина этого соотношения в осадках равна приблизительно 15 (измерение относится ко времени взятия проб речной воды), тогда как соотношение в речной воде на трех станциях в нижнем течении реки Тонэгава показало соответственно: 11, 8, 1 и 7, 9.
ARRASTRE POR LOS RÍOS DEL RADIOESTRONCIO CONTENIDO EN LAS PRECIPITACIONES RADIOACTIVAS Y SU APLICACIÓN EN HIDROLOGÍA. Los autores han medido la concentración de 88Sr y de 89Sr en aguas fluviales y en nieve, en mayo de 1962. Para obtener información acerca de las características del equilibrio hídrico entre el agua de lluvia y el desagüe del río se han tenido en cuenta la diferencia entre los períodos de semidesintegración de estos radioisótopos (31 días y 18 años, respectivamente), sus propiedades químicas idénticas y los mecanismos por los que llegan a la tierra.

La razón 89Sr/88Sr en las aguas pluviales es una función del tiempo, siempre que la serie de ensayos nucleares de la Unión Soviética se asimile a una sola explosión efectuada en el otoño de 1961. El valor de la razón que se midió en las precipitaciones, en el momento de recoger las muestras de agua fluvial, fue de 15, aproximadamente, mientras que las razones medidas en el agua del río Tonegawa, en tres estaciones dispuestas en el sentido de la corriente, fueron, respectivamente, de 11, 8,1 y 7,9.

La razón 88Sr/86Sr en el manto freático acumulado, originado por aportaciones anteriores, tiende a cero por haberse desintegrado el 83Sr en virtud de su corto período. Por tanto, los valores inferiores de la razón, hallados en el agua fluvial, se explican admitiendo la mezcla del radioestroncio arrastrado por las aportaciones recientes con el de las aportaciones más antiguas, que se infiltra en el río con las aguas subterráneas.

1. INTRODUCTION

Radioisotopes are a useful means to solve problems in hydrology, and tracer experiments have been made on a rather small scale by many investigators. For the solution of problems in a larger area, such as the water balance between rainfall and runoff, however, a great quantity of radioisotopes would be required. For this purpose the world-wide release of radioisotopes by nuclear bomb testings may prove useful. However, any isotope other than tritium may to some extent interact in solution with suspended materials, soils, etc. in a water system, which introduces a difficulty. Consequently, the results obtained for the balance of an isotope cannot be extended immediately to the water balance itself. For example, the runoff of Cs137 by river is 1-2% of the yearly deposition in contrast to 5-10% in Sr90, even when both radionuclides are present in the same water system [1]. The difficulty encountered could be overcome by the use of two isotopes of the same element, because the ratio of the isotopes should not be changed by any physical, chemical or biological reactions which may take place in a water system. This fact encouraged us to apply the measurements of radiostrontium in river water to the estimation of the mixing ratio of surface runoff and ground discharge.

2. PRINCIPLE

Both Sr89 and Sr90 are produced by a nuclear explosion and supplied to the land surface by the same mechanism, but they are quite different in their radioactive decay coefficient. The half-life of Sr89 is short (51 d) compared with that of Sr90, (28 yr), and the ratio of Sr89 and Sr90 infission products is a function of time if they were formed in a single explosion.
Fig. 1 shows the observed ratios of Sr$^{89}$ to Sr$^{90}$ in precipitation plotted in logarithmic scale against time in linear scale. The small circle indicates the ratio in the monthly precipitation measured at the Meteorological Research Institute in Tokyo. Despite our uncertainty about the frequency and scale of the series of nuclear explosions conducted by the Union of Soviet Socialist Republics in the last quarter of 1961, the approximate linearity observed in the ratio of Sr$^{89}$ to Sr$^{90}$ in the precipitation would indicate that the assumption of a single explosion can justifiably be made. On the basis of this assumption, a quite attractive principle emerges, as follows: only time determines the ratio of Sr$^{89}$ to Sr$^{90}$ in the surface waters, whether it be fresh precipitation or accumulated snow.

In Fig. 2 is shown a schematic diagram of this principle. The ratio of Sr$^{89}$ to Sr$^{90}$ in precipitation at the time of river-water sampling was found
to be 15, and this value was also considered valid for accumulated snow; nevertheless we obtained the ratio of 11 in river water at Station A and of 8.1 and 7.9 downstream at Stations B and C, respectively (Fig. 3). The lower values in the river water could not be explained by simple dilution with groundwater.

As shown in Table I, as much as 28.6 mc of Sr$^{90}$ had accumulated on the ground because of its long half-life of 28 yr, a greater fraction originating from older debris since 1954. On the other hand, the Sr$^{89}$ from older debris had decayed out because of its short half-life of 51 d. Therefore, in surface soil a much greater quantity of Sr$^{90}$ could be found than in current precipitation. The ratio of Sr$^{89}$ to Sr$^{90}$ in surface soil was calculated to be 0.73 at the time of river-water sampling (beginning of May 1962) in contrast to the ratio of 15 in current precipitation.

The second assumption was made in regard to the exchange reaction between water and soil, as follows: if surface water has penetrated into and passed through surface soil and an equilibrium in radiostrontium has been attained between water and soil, the original ratio of Sr$^{89}$ to Sr$^{90}$ in the surface water, namely 15, would be changed to 0.73, which is equivalent to the ratio in the surface soil. This change is likely to occur in a relatively short period of time, because the presence of Sr$^{90}$ in a relatively thin layer of the
RUNOFF OF FALL-OUT STRONTIUM

TABLE I
FALL-OUT RATES AND CUMULATIVE GROUND DEPOSITS OF Sr$^{89}$ AND Sr$^{90}$
(End of April 1962*)

<table>
<thead>
<tr>
<th></th>
<th>Sr$^{89}$</th>
<th>Sr$^{90}$</th>
<th>Ratio Sr$^{89}$/Sr$^{90}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fall-out rate</td>
<td>11.2</td>
<td>0.76</td>
<td>15</td>
</tr>
<tr>
<td>(mc/km$^2$month)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cumulative ground deposit</td>
<td>21</td>
<td>28.6</td>
<td>0.73</td>
</tr>
<tr>
<td>(mc/km$^2$)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Measurements by the Meteorological Research Institute, Tokyo.

Ground should be affected by a relatively rapid exchange reaction between water and soil. A laboratory experiment on the exchange reaction performed by MIYAKE and TSUBOTA [2] would also support this view. Then, the lower values of the ratio in river waters would be explained by the mixing of surface runoff in which the ratio is 15 and groundwater in which the ratio is 0.73.

On the assumption of equal concentration of Sr$^{90}$ in surface water and groundwater, the surface runoff $Q_s$ and the ground discharge $Q_d$ are given by the following equations:

$$Q_s + Q_d = Q$$

$$(15Q_s + 0.73Q_d)/(Q_s + Q_d) = R,$$

where $R$ is the ratio of Sr$^{89}$ to Sr$^{90}$ in the river water and $Q$ is the total discharge. The values of $Q_s$ and $Q_d$ at each sampling station can be calculated on the basis of observed values for $Q$ and $R$ shown in Table II.

3. RESULTS ON APPLICATION

The collection of river waters was made at the beginning of May 1962 at three stations along the River Tonegawa, which has the largest drainage area in Japan (15,760 km$^2$). Fig. 3 shows the map of the area under investigation. The River Tonegawa drains a mountainous region in which the highest mountains rise about 2000 m above sea level. At the beginning of May the mountainous region was still covered with snow.

River-water sampling was made at three guaging stations where the observation of discharge is continuously made by the Regional Hydrologic Station. A, B and C indicate the positions of sampling. Twenty liters of water was collected at each station on one day and analysed for Sr$^{89}$ and
TABLE II
RUNOFF OF Sr\textsuperscript{89} AND Sr\textsuperscript{90} FROM A RIVER
(River Tonegawa, Japan, May 1962)

<table>
<thead>
<tr>
<th>Sampling Station</th>
<th>A Komatsu</th>
<th>B Iwamoto</th>
<th>C Menuma</th>
</tr>
</thead>
<tbody>
<tr>
<td>Area of water shed (km\textsuperscript{2})</td>
<td>430</td>
<td>1650</td>
<td>5370</td>
</tr>
<tr>
<td>Area of snow cover (km\textsuperscript{2})</td>
<td>356</td>
<td>733</td>
<td>937</td>
</tr>
<tr>
<td>Discharge (Q) (m\textsuperscript{3}/s)</td>
<td>68</td>
<td>168</td>
<td>218</td>
</tr>
<tr>
<td>Sr\textsuperscript{89} in river water (pc/l)</td>
<td>10</td>
<td>5.3</td>
<td>4.0</td>
</tr>
<tr>
<td>Sr\textsuperscript{90} in river water (pc/l)</td>
<td>0.91</td>
<td>0.65</td>
<td>0.51</td>
</tr>
<tr>
<td>Ratio of Sr\textsuperscript{89}/Sr\textsuperscript{90} (R)</td>
<td>11</td>
<td>8.1</td>
<td>7.9</td>
</tr>
</tbody>
</table>

Sr\textsuperscript{90} by a conventional, well-established radiochemical method. Usually, 10-20 pc of Sr\textsuperscript{90} and from 100 to 200 pc of Sr\textsuperscript{89} were measured by low-background beta counting equipment. The results of analysis and pertinent data are given in Table II.

Table III shows the estimates of surface runoff and ground discharge at three stations based on the assumption of the ratio of Sr\textsuperscript{90} concentrations in the surface water and the groundwater being (1) 1.0 and (2) 10.

As we had little rain at that season, the surface runoff was probably caused almost entirely by melting snow. This would imply the large contribution of surface runoff to the total discharge at Station A, as compared with other stations downstream. On the basis of the assumption that the surface runoff was entirely from snow melt, it would be possible to calculate the average rate of snow melt for the mountainous area above Station A. Let the area of snow cover be 356 km\textsuperscript{2} and the surface runoff be 49 m\textsuperscript{3}/s; then the rate of snow melt is calculated as 1.2 g/cm\textsuperscript{2}d. This estimate, however, cannot be taken as more than a minimum value, because it does not include the water infiltrated into the soil.

4. CONCLUSION

In the application of this method to estimate the mixing proportion of surface runoff and ground discharge, the following conditions in the fall-out radioactivities must be fulfilled: first, the accumulation of long-lived Sr\textsuperscript{90}
TABLE III

ESTIMATES OF SURFACE RUNOFF AND GROUND DISCHARGE
based on the assumption of the ratio of Sr$^{90}$ concentration in surface water and groundwater being (1) 1.0 and (2) 10 (River Tonegawa, May 1962)

<table>
<thead>
<tr>
<th>Sampling Station</th>
<th>Assumption</th>
<th>A</th>
<th>B</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>(1)</td>
<td>(2)</td>
<td>(1)</td>
</tr>
<tr>
<td>Surface runoff ($Q_s$) ($m^3/s$)</td>
<td>49</td>
<td>14</td>
<td>87</td>
<td>16</td>
</tr>
<tr>
<td>Ground discharge ($Q_d$) ($m^3/s$)</td>
<td>19</td>
<td>54</td>
<td>81</td>
<td>152</td>
</tr>
<tr>
<td>Percentage contribution of surface runoff to the total discharge ($Q_s/Q\times 100%$)</td>
<td>72</td>
<td>21</td>
<td>52</td>
<td>9.5</td>
</tr>
</tbody>
</table>

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in soil and, secondly, a single shot of nuclear explosion which may supply fresh debris containing a sufficient quantity of Sr$^{89}$. Continuous and irregular shots of fresh debris would occasionally obscure the estimation. One of the disadvantages involved in this method is the impossibility of controlling nuclear explosions to make them of use for the present purpose.

The equal concentration of Sr$^{90}$ in surface water and the groundwater is not likely to occur. In general, the concentration of Sr$^{90}$ in groundwater is lower than that in surface water by a factor of ten or more. As shown in Table III, since the ratio of concentration of Sr$^{90}$ in surface water and groundwater greatly affects the estimation of the mixing ratio, it would be necessary to know the ratio of concentration as exactly as possible.

REFERENCES


DISCUSSION

See the discussion following the paper "Estimation of the direct contribution of meteoric water to river waters by means of fall-out radiocaesium and radiostrontium", these Proceedings.
ESTIMATION OF THE DIRECT CONTRIBUTION OF METEORIC WATER TO RIVER WATERS BY MEANS OF FALL-OUT RADIOCAESIUM AND RADIOSTRONTIUM

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Abstract — Résumé — Аннотация — Resumen

ESTIMATION OF THE DIRECT CONTRIBUTION OF METEORIC WATER TO RIVER WATERS BY MEANS OF FALL-OUT RADIOCAESIUM AND RADIOSTRONTIUM. It may be of interest to determine the ratio of direct contribution of meteoric water to river water. We used as radioactive tracers Cs\textsuperscript{137} and Sr\textsuperscript{89} in river water originating from fall-out to estimate the above ratio.

Ten representative rivers in Japan were chosen: Ishikari (Hokkaido), Mogami, Kitakami, Shinano, Tone, Kiso, Yodo, Asahi (Honshu), Yoshino (Shikoku) and Chikugo (Kyushu).

Sampling of water was undertaken during the period from the end of July to early August 1961. The sampling site of each river was selected to avoid industrial contamination and inflowing sea water. Each 100-l sample was collected at three stations traversing a river. After adding a carrier of 100 mg each of stable caesium and strontium, water was passed through a cation-exchange column (Dowex 50W\textsuperscript{X8}, 50-100 mesh, Na\textsuperscript{+}-form, 500 ml) to fix caesium and strontium in the resin. Caesium and strontium were eluted with a 57\% solution of oxalic acid followed by 3 N hydrochloric acid, and they were determined in the latter fraction.

Sr\textsuperscript{89} was separated by a fuming nitric acid method. Beta counting was done on its daughter, yttrium-90, after milking. Radiocaesium was adsorbed on ammonium molybdophosphate followed by purification by means of passage through a type of phenol-sulphonic acid cation-exchange resin. Beta activity was measured in the form of perchlorate.

The average value of Cs\textsuperscript{137} and Sr\textsuperscript{89} in the water of ten rivers was 0.035 pc/l and 0.2 pc/l respectively, while those in meteoric water were 2 and 0.7 pc/l, respectively, at that time.

Since Cs\textsuperscript{137} which is one brought into contact with soil is difficult to remove, it may be said that most of the Cs\textsuperscript{137} in river water results from the direct pouring of rain water into rivers. Under this assumption, one finds that the direct contribution of meteoric water to river waters was 1.7%.

Using this value, the rate of leaching of Sr\textsuperscript{89} accumulated in the soil is on the order of 1.5%/yr.

ÉVALUATION DE LA CONTRIBUTION DIRECTE DES PRÉCIPITATIONS AU DÉBIT DES EAUX FLUVIALES, À L'AIDE DU RADIOCÉSIUM ET DU RADIOSTRONTIUM PROVENANT DES RETOMBEES. Il peut être intéressant d'évaluer en pourcentage la contribution directe des précipitations au débit des cours d'eau. A cet effet, les auteurs ont utilisé comme indicateurs le \textsuperscript{137}Cs et le \textsuperscript{89}Sr provenant des retombées radioactives.

Ils ont choisi 10 cours d'eau caractéristiques du Japon, savoir : Ishikari (Hokkaido), Mogami, Kitakami, Shinano, Tone, Kiso, Yodo, Asahi (Honshu), Yoshino, (Shikoku) et Chikugo (Kyushu).

Les échantillons d'eau ont été prélevés fin juillet et début août 1961. Sur chaque rivière, on avait choisi les lieux d'échantillonnage de manière à éviter toute contamination d'origine industrielle et l'afflux éventuel d'eau de mer. Pour obtenir chaque échantillon de 100 litres, on a prélevé de l'eau en trois endroits différents pris dans toute la largeur du cours d'eau. Après addition d'un entraîneur - 100 mg de césium et 100 mg de strontium stable par échantillon - on a fait passer l'eau sur une colonne cationique (Dowex 50\textsuperscript{X8}, maille 50-100, forme sodique, 500 ml) pour fixer le césium et le strontium sur la résine. On a élué le césium et le strontium à l'aide d'une solution d'acide oxalique à 5%, puis d'une solution d'acide chlorhydrique 3 N, et on les a mesurés dans cette dernière fraction. On a séparé le \textsuperscript{89}Sr par la méthode de l'acide nitrique fumant, puis extrait son produit de filtration, l'yttrium-90, sur lequel on a procédé au comptage bêta. Le radiocésium a
Étad adsorbé par du phosphomolybdiate d'ammonium, et ensuite purifié par filtration au moyen d'une résine cationique à base d'acide phénolsulfonique. On a mesuré l'activité bêta sous forme de perchlorate.

Pour les 10 cours d'eau considérés, la teneur moyenne des eaux en $^{137}$Cs et $^{90}$Sr s'élevait respectivement à 0,035 et 0,2 picocuries/1, alors que les valeurs correspondantes pour les précipitations étaient à la même époque de 2 et 0,7 picocuries/1.

Comme le $^{137}$Cs, une fois entré en contact avec le sol, s'en sépare difficilement, on peut considérer que la majeure partie du $^{137}$Cs contenu dans les eaux fluviales provient de la chute directe de pluie dans le cours d'eau. En partant de cette hypothèse, on a trouvé que la contribution directe des précipitations au débit des rivières était au maximum de 1,7%.

En utilisant cette valeur, on a admis que le taux de lixiviation du $^{90}$Sr accumulé dans le sol était de l'ordre de 1,5% par an.

**OPRÉDELENNIY NEPPOSREDSTVENNOGO POPADANII DOZHDEVOY VODY V RENCHU NA OCHOSJE VYAPADENIA RADIOAKTIVNYH CZEZIA I STRONCII.** Para éprenta nesopredstvennykh popadaniy dozhdevoi vody c renchku, biyami ispol'zovanyi v kachestve indikatorov $^{239}$Ca i $^{90}$Sr, попавши в renchu v rezultate yvitya radioaktivnyh osadkov.

C etoj cezly byli izobrazeny 10 harakteristicheskikh rek Yaponii, a imey: Ishikari (o–v Hakkaido), Mogami, Kitakami, Shinano, Tone, Kiso, Yodo, Aioi, Isawa (o–v Hokkaido), Isshigaki (o–v Shikoku) i Tikuho (o–v Kyusyu).

Obraztsy vody brylyi v period — kometska ylye nachalo avgusta 1961 goda. Otbor prob vody v jekhoy rench proizvoditsya tak, chto kolichestvo popadaniy priyazhennykh zagryazneniy i morskoi vody.

Kadye 100 l, opravleny v kachestve obraztsa, zabiralsya na trseh stacionakh, peremeshavshiyo po rench. Po leto того, kak v vody dobyvalsya nozhitels, soedinty po 100 mg stabiliizirovshih chevki i strochny, voda propuskaetsya cez kationnoobmennyi koloniyu(Dowex 50W-X8, 50 – 100 moll, 500 mli)
i s tem, chtoby vydyelit' ceszii i stroncii s pomoshhy 5-procentnogo rastwora tsvevednoy kislote s pos-

следующей обработкой в 3N соляной кислоте. Определение производилось в последней фракции. Sr$^{90}$ был отделен методом дымчатой азотной кислоты. Счет бета-частиц был проведен на его дочернем изо-
topeno ittriy-90 po leto vydeleniya poslednego iz materyarnogo veshchestva. Radiooedel'ce byl adsorbirovan

na molibdenovokislotnom svedee vymenya s posleduyshchey ochistkoy proпускonom cez ionnoobmennoy smolu

типа фенолсульфокислоты. Бета-активность измерялась на образце в виде соли хлорной кислоты.

Средние концентрации $^{239}$Ca i $^{90}$Sr v voda deceatiy rek okazaslys ravnymi sootvetstvenno 0,035 i 0,2 mikkokury/л, toqka kak eti vechnosti v dozhdevoi vode soostavili 2 i 0,7 mikkokury/л v te je

samo vreme.

Поскольку $^{239}$Ca, однажды попавший в почву, извёстность оттуда трудно, to moeno eshalo, celo $^{137}$Cs, nachaloegosha v renchke vode, jekhoy kolichestvo nesopredstvennogo popadaniy dozhdevoi vody v rench. Isehoda iz etogo predpostavleniya, moeno utverdilo, celo $^{137}$Cs, nachaloegosha v renchke vode, jekhoy

попавшой v renchke vode, jekhoy kolichestvo nesopredstvennogo popadaniy dozhdevoi vody, popavshoy v renchke vode, jekhoy kolichestvo nesopredstvennogo popadaniy dozhdevoi vody, popavshoy v renchke vode, jekhoy

равно 1,7%. Ispol'zuyu etu velychnyu,

avtorry vyvodit normu yvyelachivania Sr$^{90}$, akkumulyirovannogo v poche, ranu romero 1,5% v yd.

**CALCULO DE LA APORTACION DIRECTA DEL AGUA METEORICA A LAS AQUAS FLUVIALES BASADO EN EL RADIOCESIO Y RADIOESTRONCIO CONTENIDOS EN LAS PRECIPITACIONES.** Los autores estima interesante determinar la raóon aportación directa de agua meteorica: caudal de un río. Para calcularla, han utilizado como indicadores el $^{137}$Cs y el $^{90}$Sr presentes en las aguas fluviales y provenientes de las precipitaciones radiactivas.

Han seleccionado los 10 ríos siguientes, características del Japón: Ishikari (isla de Hokkaido), Mogami, Kitakami, Shinano, Tone, Kiso, Yodo, Asahi (Honshu), Yoshino (isla de Shikoku) y Chikugo (isla de Kyusyu).

Recogieron las muestras de agua entre finales de julio y principios de agosto de 1961. Para efectuar el muestreo, seleccionaron lugares exentos de contaminación por residuos industriales y aguas de mar. Las muestras, de 1001 cada una, fueron recogidas en tres puntos situados en sentido perpendicular a la corriente del río. Después de adicionar, como portadores, 100 mg de cesio y 100 mg de estroncio estables, el agua se hizo pasar a través de una columna de intercambio catiónico (500 ml de Dowex 50W-X8, de 50 a 100 mallas, forma sólida) para fijar el cesio y el estroncio en la resina. Estos dos elementos se clayeron con una solución de ácido oxálico al 5% y, a continuación, con ácido clorhídrico 3 N y se determinaron en esta última fracción. El $^{90}$Sr se separó por el método del ácido nítrico fumante y el recuento beta se efectuó con su producto de fijación, el tritio-90, después de separarlo periódicamente. El radiocesio se adsorbió en fosfomolibdato amónico
CONTRIBUTION OF METEORIC TO RIVER WATER

To determine the ratio of the direct contribution of meteoric water to river waters, we used as radioactive tracers Cs\(^{137}\) and Sr\(^{90}\) in river water originating from fall-out. Ten representative rivers in Japan were chosen for this purpose, as follows: Ishikari (Hokkaido Island), Mogami, Kitakami, Shinano, Toné, Kiso, Yodo, Asahi (Honshu), Yoshino (Shikoku Island) and

![Fig. 1](image)

**Fig. 1**

Japanese rivers from which water samples were collected

An open circle represents a sampling station in each river.

<table>
<thead>
<tr>
<th>No.</th>
<th>Name of river</th>
<th>No.</th>
<th>Name of river</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Ishikari</td>
<td>6</td>
<td>Kiso</td>
</tr>
<tr>
<td>2</td>
<td>Mogami</td>
<td>7</td>
<td>Yodo</td>
</tr>
<tr>
<td>3</td>
<td>Kitakami</td>
<td>8</td>
<td>Yoshino</td>
</tr>
<tr>
<td>4</td>
<td>Shinano</td>
<td>9</td>
<td>Asahi</td>
</tr>
<tr>
<td>5</td>
<td>Toné</td>
<td>10</td>
<td>Chikugo</td>
</tr>
</tbody>
</table>
Chikugo (Kyushu Island). The locations of these rivers and sampling sites for water samples are given in Fig. 1.

The sampling of water was undertaken during the period from the end of July to early August 1961 before nuclear tests were resumed after the three year moratorium. The sampling site on each river was selected to avoid industrial and urban pollution as well as inflowing sea water.

Each 100-l sample was collected at three stations traversing a river. After a carrier of 100 mg each of stable caesium and strontium was added, water was passed immediately after sampling through a 20-cm-long cation-exchange column (Dowex 50W-X8, 50 - 100 mesh, Na-form, 500 ml) in order to trap caesium and strontium in the resin. Caesium and strontium were eluted with a 5% solution of oxalic acid followed by a 3 N hydrochloric acid solution, and they were determined in the latter fraction. Sr\(^{90}\) was separated by co-precipitation with calcium carbonate, from which it was isolated by means of the fuming nitric acid method [1]. Beta counting was repeated on its daughter, Y\(^{90}\), after milking. Radiocaesium was first adsorbed on solid ammonium molybdophosphate and then purified by being passed through a phenol-sulphonic acid type cation-exchange resin [2]. Finally, the $\beta$-activity of Cs\(^{137}\) was measured in the compound form of the perchlorate. Purification of both radionuclides was repeatedly carried out until their specific activities reached constant values.

### Table I

**THE RESULTS OF ANALYSIS OF Sr\(^{90}\) AND Cs\(^{137}\) IN JAPANESE RIVERS**

(July - August 1961)

<table>
<thead>
<tr>
<th>Name of river</th>
<th>Total amount of water flow at sampling site ((x 10^3 m^3/yr))</th>
<th>Surface area of drainage ((10^3 km^2))</th>
<th>Sr(^{90}) ((pc/l))</th>
<th>Cs(^{137}) ((pc/l))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ishikari</td>
<td>14.5</td>
<td>12.7</td>
<td>0.21</td>
<td>0.036</td>
</tr>
<tr>
<td>Mogami</td>
<td>11.2</td>
<td>6.1</td>
<td>0.21</td>
<td>0.045</td>
</tr>
<tr>
<td>Kitakami</td>
<td>10.3</td>
<td>7.9</td>
<td>0.20</td>
<td>0.035</td>
</tr>
<tr>
<td>Shinano</td>
<td>16.2</td>
<td>10.1</td>
<td>0.18</td>
<td>0.031</td>
</tr>
<tr>
<td>Toné</td>
<td>9.1</td>
<td>8.6</td>
<td>0.21</td>
<td>0.033</td>
</tr>
<tr>
<td>Kiso</td>
<td>11.7</td>
<td>4.9</td>
<td>0.11</td>
<td>0.026</td>
</tr>
<tr>
<td>Yodo</td>
<td>10.1</td>
<td>7.2</td>
<td>0.31</td>
<td>0.031</td>
</tr>
<tr>
<td>Yoshino</td>
<td>5.5</td>
<td>2.8</td>
<td>0.09</td>
<td>0.020</td>
</tr>
<tr>
<td>Asahi</td>
<td>1.6</td>
<td>1.6</td>
<td>0.13</td>
<td>0.036</td>
</tr>
<tr>
<td>Chikugo</td>
<td>2.8</td>
<td>1.7</td>
<td>0.11</td>
<td>0.034</td>
</tr>
</tbody>
</table>

**weighted mean** 0.19 ± 0.06 0.034 ± 0.007
The results of analyses of Sr$^{90}$ and Cs$^{137}$ together with other relevant data are given in Table I. Averaged values of Cs$^{137}$ and Sr$^{90}$ in waters of ten rivers were respectively $0.034 \pm 0.007 \text{pc/l}$ and $0.19 \pm 0.06 \text{pc/l}$ while those in meteoric water were respectively $2 \text{pc/l}$ and $0.7 \text{pc/l}$ at that time, according to observations by MIYAKE et al. [3].

Since it is known that Cs$^{137}$ which is once brought into contact with soil is difficult to remove [4, 5], it may be said that the major part of Cs$^{137}$ in river water is derived from direct pouring of rain water into rivers. Under this assumption, with the ratio of $0.035 \text{pc/l}$ of river water and $2 \text{pc/l}$ of rain water, the maximum value of 1.7% was obtained as the direct contribution of meteoric water to river water.

If the same ratio of the direct contribution of rain water to Sr$^{90}$ content is applied, the maximum value of direct pouring of fall-out Sr$^{90}$ into rivers is estimated to be only $0.01 \text{pc/l}$ which is about 5% of the total concentration of Sr$^{90}$ in river waters.

The following experiments were made in order to confirm the above assumption that both Sr$^{90}$ and Cs$^{137}$ in rain water were trapped in the soil at the same rate. $20\text{g}$ of soil samples collected near the sampling sites of river waters was shaken with $300\text{ml}$ of tap water containing radiotracers of Cs$^{134}$ and Sr$^{85}$. Each 5-ml solution was taken out intermittently at regular time intervals and, after centrifugation, 2 ml of supernatant was pipetted out; in the supernatant activities of Cs$^{134}$ and Sr$^{85}$ were determined with a scintillation counter to obtain the adsorption rate of each nuclide on the soil. The result showed that the adsorption rate of each nuclide, which is usually regarded as of the first-order reaction [6], was rapid and almost the same at the earlier stage of adsorption. The adsorption rate constant, $K$ in Eq.(1) below, of Sr$^{90}$ and Cs$^{137}$ within a few hours ranges respectively from 0.51 to 1.6 and from 0.57 to 1.9:

$$K = (1/t) \left(1/2.3\right) \log \left(C_0/C\right), \quad (1)$$

where $t$ is time in h, $C_0$ is the initial activity of each solution and $C$ is the activity at the time $t$.

The result of experiments indicates that the rate of removal of Sr$^{90}$ and Cs$^{137}$ from water into the soil is almost the same and rapid once rain water is brought into contact with soil. Therefore, it may be right to consider that most of Sr$^{90}$ in river water originates from the release of the nuclide from the soil in which fall-out Sr$^{90}$ has been accumulated.

For an estimate of the rate of leaching of Sr$^{90}$ from the soil, it is reasonable to compare the total amount of effluent of Sr$^{90}$ through rivers with the cumulative amount of the same nuclide in the soil. To do so, we determined the cumulative amount of Sr$^{90}$ by analysing soil samples collected near the water sampling sites. The analytical values of soil bearing Sr$^{90}$ are given in Table II. As shown in Table II, it was found that the weighted mean of the rate of leaching of Sr$^{90}$ from the soil was 1.5% annually, and the total amount of effluent of Sr$^{90}$ from the Japanese Islands was about 100 c at the time of observation.

We should refer in this connection to the Second Report of the United Nations Scientific Committee on the Effect of Atomic Radiation (1962) [7], which gives the value of 2% per annum as an adequately cautious average
Y. MIYAKE and H. TSUBOTA

## TABLE II

CUMULATIVE AMOUNT OF Sr\(^{90}\) IN THE SOIL NEAR THE RIVERS CONCERNED AND ANNUAL RATE OF NET LOSS OF Sr\(^{90}\) FROM THE LAND (JAPAN), 1961

<table>
<thead>
<tr>
<th>Name of river</th>
<th>Cumulative amount of Sr(^{90}) in the soil near the river (mc/km(^2))</th>
<th>Total amount of Sr(^{90}) in drainage area (c)</th>
<th>Annual rate of effluent of Sr(^{90}) (c)</th>
<th>Annual rate of loss of Sr(^{90}) from the land (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ishikari</td>
<td>23</td>
<td>290</td>
<td>3.1</td>
<td>1.1</td>
</tr>
<tr>
<td>Mogami</td>
<td>39</td>
<td>240</td>
<td>2.4</td>
<td>1.0</td>
</tr>
<tr>
<td>Kitakami</td>
<td>17</td>
<td>130</td>
<td>2.1</td>
<td>1.6</td>
</tr>
<tr>
<td>Shinano</td>
<td>29</td>
<td>290</td>
<td>2.9</td>
<td>1.0</td>
</tr>
<tr>
<td>Tone</td>
<td>19</td>
<td>160</td>
<td>2.0</td>
<td>1.3</td>
</tr>
<tr>
<td>Kiso</td>
<td>8.2</td>
<td>40</td>
<td>1.3</td>
<td>3.3</td>
</tr>
<tr>
<td>Yodo</td>
<td>19</td>
<td>140</td>
<td>3.1</td>
<td>2.2</td>
</tr>
<tr>
<td>Yoshino</td>
<td>24</td>
<td>67</td>
<td>0.50</td>
<td>0.75</td>
</tr>
<tr>
<td>Asahi</td>
<td>19</td>
<td>30</td>
<td>0.21</td>
<td>0.7</td>
</tr>
<tr>
<td>Chikugo</td>
<td>19</td>
<td>32</td>
<td>0.31</td>
<td>1.0</td>
</tr>
</tbody>
</table>

**weighted mean** 1.5%

of the net loss of Sr\(^{90}\) from the soil, this will result in underestimation of the general level of Sr\(^{90}\) in the soil in the future. It is to be noted that the value of 1.5% obtained in Japan is even smaller than that given by UNSCEAR.

**ACKNOWLEDGEMENTS**

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The radiochemical analysis of Sr\(^{90}\) and Cs\(^{137}\) were made with the assistance of Miss A. Horiuchi, to whom the authors are indebted.

**REFERENCES**

CONTRIBUTION OF METEORIC TO RIVER WATER


DISCUSSION

G. H. LEAN: Mr. Yamagata, I wonder whether you have any information on how the ratio of surface runoff to ground discharge varied with the duration and intensity of the rainfall. This is a point which would be of interest to engineers.

N. YAMAGATA: Our samples were taken at the beginning of May and at that time the surface runoff was almost entirely from snow melt. We could not have carried out the investigation at any other time of the year because the fallout rate for Sr$^{89}$ and Sr$^{90}$ would not have been suitable.

J.C. VOGEL (Chairman): That would explain the difference in the runoff contributions given in Mr. Yamagata's and Mr. Miyake's paper. It is probably from the fact that the samples were collected at different times of the year.

P. K. ZUTSHI: Mr. Miyake, your experiments on the adsorption of Cs and Sr seem to have been performed with a fair amount of stable carrier. I wonder whether this might not explain why no difference was observed in the behaviour of the two isotopes. According to a 1961 National Aeronautics and Space Administration report on fallout from bomb debris, there is some geo-chemical effect operative in the troposphere, as a result of which various nuclides are liable to be affected by fractionation. This would be likely to affect the ratio of Cs to Sr in fallout.

Y. MIYAKE: In fission products, the Cs$^{137}$ - Sr$^{90}$ ratio is between 1.7 and 2, whereas in rain-water the figure is sometimes as high as three or four in Japan. On the other hand, the ratio is much lower in river water, which means that it is difficult for the caesium to be removed from the soil.
MEASUREMENT OF RIVER DILUTION AND FLOW
BY MEANS OF RADIOACTIVE TRACERS
AND ACTIVATION ANALYSIS

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Abstract — Résumé — Аннотация — Resumen

MEASUREMENT OF RIVER DILUTION AND FLOW BY MEANS OF RADIOACTIVE TRACERS AND ACTIVATION ANALYSIS. The contamination of rivers is one of the most serious problems of the Tokyo Metropolitan Government from the sanitary point of view.

The use of radioactive tracers has overcome the difficulties encountered by other conventional methods for determination of the contribution of the effluent waste water from a particular plant to the contamination of rivers.

This paper describes the method adopted by us to measure the velocity and the dilution rate of radioactive NaCl solution injected in a river as a point source and of non-active NaBr or NH₄Br solution injected continuously.

In our experiment radioactive tracers acted as indicators of the arrival of non-active tracer chemicals at the observation stations downstream.

One liter of NaCl solution containing 3 g of NaCl, whose total activity was 13.7 mc, was poured into the river first as a point source; this was followed by a continuous injection for 30 min of 70 l of NaBr solution, whose concentration was 13 per cent. In the second tracer experiment we used 100 kg of NH₄Br as a non-active tracer to compare the effectiveness of Br⁸¹ and Br⁸₂.

Immediately after the completion of injection, one liter of radioactive NaCl solution was again added. By means of a wafer-tight scintillation counter immersed in the stream, the change of the activity downstream was recorded continuously. The dilution rates up to 10⁶ and the mean flow velocity at each interval were obtained from the calibration of the measured values.

The specific activities determined by radiometric analysis of the water samples showed good accordance with those obtained by the method mentioned above.

A small portion (0.1-1 cm³) of the water samples was irradiated for (3 min-6 h) in a neutron flux of about 4 x 10¹¹ n cm⁻² s⁻¹.

From the activities of Na²⁴, Br⁸¹ or Br⁸₂ we have also determined the dilution rates, which showed about one hundredth values of those in the case of the point-source method.

We have thus established a method for tracing the constant injection of effluent water using as small an amount of radioactivity as possible.

EMPLOI DE RADIOINDICATEURS ET DE L'ANALYSE PAR ACTIVATION POUR LA MESURE DU DÉBIT DES COURS D'EAU ET DE LA DILUTION DANS LEURS EAUX. La contamination des eaux fluviales est l'un des plus graves problèmes d'assainissement que la Municipalité de Tokyo ait à résoudre.

L'emploi de radioindicateurs a permis de surmonter les difficultés que l'on éprouve à déterminer, par les méthodes classiques, dans quelle mesure les effluents d'une installation donnée contribuent à contaminer un cours d'eau.

Les auteurs du mémoire décrivent la méthode qu'ils ont adoptée pour mesurer la vitesse de déplacement et le taux de dilution d'une solution de NaCl radioactif, injectée dans un cours d'eau comme source ponctuelle, ainsi que d'une solution de NaBr ou NH₄Br stable injectée de manière continue.

Lors de cette expérience, le radioindicateur a été utilisé comme « marqueur » pour signaler l'arrivée des indicateurs chimiques stables aux points d'observation situés en aval.

Le déversement dans le cours d'eau, en tant que source ponctuelle, d'un litre de solution de NaCl contenant 3 g de NaCl, dont l'activité totale s'élevait à 13.7 mc, a été suivi de l'injection continue, pendant 30 min, de 70 l d'une solution de NaBr à une concentration de 13%. Pour la seconde expérience, les auteurs ont utilisé 100 kg de NH₄Br comme indicateur stable pour comparer l'activité de Br⁸¹ et de Br⁸₂.
Aussitôt cette opération terminée, on a versé de nouveau un litre de solution de NaCl radioactive. A l'aide d'un compteur à scintillations étanche, immergé dans le cours d'eau, on a enregistré de manière continue les variations de l'activité en aval. D'après les valeurs mesurées, on a obtenu des taux de dilution allant jusqu'à 10^8 et la vitesse moyenne du courant entre les différentes stations d'observation.

On a constaté que les activités spécifiques, déterminées par l'analyse radiométrique des échantillons d'eau, concordaient de manière satisfaisante avec celles qui avaient été obtenues par la méthode susmentionnée.

Une petite fraction (de 0,1 à 1 cm^3) des échantillons à été irradiée pendant divers laps de temps (de 3 min à 6 h) dans un flux de neutrons de 4 \times 10^{11} n/cm^2 s environ.

D'après les activités de ^{24}Na, ^{60}Br ou ^{82}Br, les auteurs ont aussi déterminé les taux de dilution et on trouvé des valeurs de l'ordre du centième environ de celles qui avaient été obtenues avec la méthode de la source ponctuelle.

Les auteurs ont ainsi mis au point une méthode permettant de suivre, à l'aide d'une quantité de radioactivité aussi faible que possible, l'injection continue d'effluents liquides dans les cours d'eau.

ИЗМЕРЕНИЕ РАЗБАВЛЕНИЯ И ФОРМИРОВАНИЯ ПОТОКОВ В РЕКАХ ПОСРЕДСТВОМ РАДИОАКТИВНЫХ ИНДИКАТОРОВ И АКТИВАЦИОННОГО АНАЛИЗА. С точки зрения санитарии загрязнение рек является одной из наиболее серьезных проблем для муниципальных властей Токио.

Использование радиоактивных индикаторов позволило преодолеть трудности, с которыми сталкивались при использовании обычных методов определения степени загрязнения рек сточными водами отдельных заводов.

Дается описание метода, примененного нами для измерения скорости и степени разбавления радиоактивного раствора NaCl, введенного в реку в качестве точечного граничного источника, а также неактивного раствора NaBr или NH₄Br, вводимого постоянно.

Радиоактивные индикаторы выступали в опыте в качестве указателей поступления неактивных химических индикаторов в район расположения наблюдательных станций, находящихся в нижнем течении реки.

Сначала в качестве сигнального источника в реку выливался 1 л раствора NaCl, содержащий 3 г NaCl, общая активность которого составляла 13,7 милликюри. После этого в течение 30 мин непрерывно выливались 70 л раствора NaCl, концентрация которого составляла 13%. При проведении второго опыта с применением индикаторов мы использовали 100 кг NH₄Br в качестве неактивного индикатора для сравнения эффективности Be⁶⁰ и Be⁸². Сразу же после этого сковородился 1 л радиоактивного раствора NaCl. В нижнем течении реки с помощью водонепроницаемого сцинтилляционного счетчика, опущенного в воду, проводилась непрерывная запись изменения активности. Степень разбавления достигала 10^6, в значение средней скорости потока в каждом интервале было внесено путем калибровки измеренных величин.

Удельная активность, определенная с помощью радиометрического анализа образцов воды, показала точное соответствие с результатами, полученными с помощью вышеупомянутого метода.

Исходя из активности Na²⁴, Be⁶⁰ или Br⁸², мы определили также степень разбавления, которая оказалась равной почти одной сотой от значений, определенных методом сигнального источника.

Таким образом, мы разработали метод измерения непрерывных сбросов сточных вод в реку с применением минимальных активностей.

MEDIDA DE LA DILUCIÓN Y DEL CAUDAL EN LOS RÍOS POR MEDIO DE INDICADORES RADIACTIVOS Y DE ANÁLISIS POR ACTIVACIÓN. Desde el punto de vista sanitario, uno de los problemas más graves planteados a las autoridades municipales de Tokio es la contaminación de los ríos.

Gracias al empleo de indicadores radiactivos, se han superado las dificultades inherentes a la aplicación de otros métodos tradicionales para evaluar la aportación de las aguas residuales provenientes de una instalación industrial determinada a la contaminación de los ríos.

Los autores describen el método que han utilizado para medir la velocidad de desplazamiento y el índice de dilución de una solución de NaCl radiactiva inyectada en un río a manera de fuente puntiforme, así como de una solución inactiva de NaBr y de NH₄Br, inyectada en forma continua.

En el experimento, el indicador radiactivo se empleó para señalar la llegada de las sustancias indicadores inactivas a los puntos de observación dispuestos en el sentido de la corriente.
Inicialmente se descargó en el río, como fuente puntiforme, 1 l de solución de NaCl que contenía 3 g de NaCl, cuya actividad total era de 13,7 mc. Seguidamente, se inyectaron durante 30 min en forma continua 70 l de solución de NaBr, al 13%. En el segundo experimento se utilizaron 100 kg de NH₄Br como indicador inactivo, para comparar la eficacia del ⁴⁰Br y del ⁸₂Br.

Inmediatamente después de la inyección, se añadió otro litro de solución de NaCl radiactivo. La variación de la actividad se registró de manera continua aguas abajo con un contador de centelleo estanco, inmerso en la corriente. Sobre la base de los valores medidos, se han determinado índices de dilución de hasta 10⁴, así como la velocidad media de la corriente en cada intervalo.

Las actividades específicas determinadas por análisis radiométrico de las muestras de agua concordaron satisfactoriamente con los resultados obtenidos por el método antes mencionado.

<table>
<thead>
<tr>
<th>24Na</th>
<th>⁴⁰Br</th>
<th>⁸₂Br</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 min y 6 h en un flujo neutrónico de 4·10¹¹ n/cm²·s, aproximadamente.</td>
<td></td>
<td></td>
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<tr>
<td>Basándose en las actividades del ²⁴Na, ⁴⁰Br y ⁸₂Br, los autores han determinado también los índices de dilución, cuyos valores resultaron ser 100 veces inferiores, aproximadamente, a los obtenidos con el método de la fuente puntiforme.</td>
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</table>

Con ello, se ha elaborado un método para observar la inyección continua de efluientes utilizando una cantidad mínima de sustancia radiactiva.

I. INTRODUCTION

In the field of civil engineering many experiments with radioactive tracers have been carried out. As for flow study, reports of field experiments on a large-scale as well as those of rather theoretical works which concern the solution of the diffusion equation [1, 2] have been published by many workers [3, 6].

In most tracer experiments of flow study, the "spot" injection of radioactive solution has been done conventionally because of the simplicity of the mathematical analysis of the results. In our recent work we tried to ascertain the contribution of industrial effluent water to the pollution of rivers running through a large city. It was necessary for us to use a new technique because radioactivity in the city must be avoided in view of the ensuing radiation hazards which would endanger the public.

This paper describes experimental work on the measurement of the dilution patterns of "non-active" tracer solution injected continuously into the river at a constant rate: The work was done by means of activation analysis of water fractions taken up at each observation station. For this work we determined first to adopt the following methods so as to establish the best conditions for practical experiments in the near future:

1. For obtaining conditions similar to the injection of the effluent water the "continuous" injection method mentioned above must be adopted.
2. A sufficient amount of radioactivity must be used as the tracer of the "non-active" tracer to indicate the time of the arrival of the non-active tracer solution at each observation station. This activity must be reduced as much as possible in the future; moreover, it must be eliminated completely if possible.
3. By means of "spot" injection of the radioactive solution we can obtain information about "absolute" dilution rates.
4. A radioactive nuclide having as short a half-live as possible must be chosen.
5. The solute of non-active tracer solution must be a compound whose
components give one radionuclide with a large activation cross-section but no other "disturbing" radionuclides.

Elements which are present in considerable amounts in river water must be carefully avoided, because they lower the sensitivity of the activation analysis of the tracer element.

II. EXPERIMENTAL METHOD

Our experiments were done on the Kanda, which runs through the city of Tokyo from north to south.

In Fig. 1 we can see the injection points 0A and 0B as well as the observation stations located downstream (1A, 2A, ..., 1B, 2B, ...).

As non-active tracers we adopted two sorts of chemical compounds, sodium bromide and ammonium bromide. Sodium bromide (NaBr) was tried first for comparison of the practical utility of sodium with that of bromine. In this case we prepared an aqueous solution of sodium bromide which contained about 10 kg of NaBr in 701 of water.

Two capsules, each containing 3 g of sodium chloride (NaCl) were irradiated in a thermal-neutron flux of $4 \times 10^{11} \text{n/cm}^2 \text{sec}$ for 4 h; each of them gave 13.7 mc of sodium-24. Two lots of sodium chloride solution were then prepared, each of which contained 13.7 mc of sodium-24 per liter.

![Map of Kanda river showing injection and observation stations](image-url)

Fig. 1

Map showing the injection and observation stations along the river Kanda
The successive injections were carried out at the injection point (point 0A), the time curve of which can be seen in Fig. 2.

The continuous injection of sodium bromide solution was limited to a 30 min period. Fig. 2 also shows that the radioactive tracer solutions were poured into the water as "pointed" sources at the beginning and the end of the continuous injection.

At the observation stations situated at 0.88, 4.73, 11.32, 15.79 km respectively from the injection point, the change of the activity was measured directly during the passage of the radioactivity by a watertight scintillation detector.

At regular intervals water fractions were taken up until the detector immersed in the water showed that the activity had completely disappeared. These water fractions were transported immediately to the laboratory for measurement of the specific activity of sodium-24.

At the laboratory thin, solid samples were prepared by the evaporation of 100 cm$^3$ of each fraction; the radioactivities were measured by a low-background, gas-flow counter. The usual counting-rate of the background of our counter is about 0.7 cpm. The dilution configuration at each observation station was obtained from these results.

Two sorts of capsules were prepared for neutron activation, one containing 0.1 cm$^3$ water fraction of "lower dilution" and the other containing 1 cm$^3$ water for "higher dilution". These irradiation samples were sealed in polyethylene tubes for prevention of unfavourable contamination of radioactivity.

The activation analysis was performed at the Atomic Energy Research Institute of Tokai by the use of the nuclear reactor JRR-1 (water-boiler type). After 2 h of irradiation in a neutron flux of 4 \times 10^{11} n/cm$^2$ sec, the gamma-ray spectrums were measured with a 512-channel spectrometer (Nuclear Data ND 130).

In the second tracer experiment we used a solution of ammonium bromide which contained 100 kg of NH$_4$Br in 201L.

After a "spot" injection of radioactive NaCl solution (11 containing 24.6 mc of sodium-24), we continued the injection of NH$_4$Br solution for 20 min at a constant rate. The time schedule of the injection in this case is shown in Fig. 3. The operation at each station and the measurement of
the specific activity of water fractions were the same as those in the first experiment.

The activation analysis was carried out at the Nuclear Research Institute of Saint Paul's University (Rikkyo University), where the reactor TRIGA was used for the neutron irradiation.

After 3 ~ 5 min of irradiation in a thermal-neutron flux of $3 \times 10^{11}$ n/cm$^2$sec, each sample's decay curve was measured; this showed the presence of several radionuclides, such as bromine-80 (half-life, 18 min), bromine-80m (half-life, 4.5 h), bromine-82 (half-life, 35.9 h), sodium-24 (half-life, 15.0 h), chlorine-38 (half-life, 37.3 min) and manganese-56 (half-life, 2.58 h).

The graphical analysis of these decay curves being somewhat difficult, we determined the contents of bromine by the application of the measured values of $Z$ to the following equation:

$$A_0 e^{-0.693t/T_a} + B_0 e^{-0.693t/T_b} + C_0 e^{-0.693t/T_c} + \ldots = Z$$

where $A_0$: initial radioactivity of nuclide A (cpm),

$B_0$: initial radioactivity of nuclide B (cpm),

$C_0$: initial radioactivity of nuclide C (cpm),

$T_a, T_b, T_c, \ldots$: half-lives of nuclides A, B, C, ...

To establish a more simple method of activation analysis by using the activity of bromine-82, we performed the 6-h irradiation in the same density of neutron flux.

About 40 h of "cooling" time after the irradiation made the measurement of bromine-82 possible. In this case a $\gamma$-ray spectrometer (ND 130) was used not only for identification of a particular radionuclide but also for determination of its concentration.

III. RESULTS OBTAINED

In Figs. 4 and 5 we can see the curves which show the change of the rate-meter indication obtained at each observation station. It would seem
to be possible to obtain not only any dilution rate at a particular station but also the change of the dilution-rate by time after an appropriate calibration of results. This was not true in our case, however, because the depth of the river where the detector was immersed was not constant; in other words, the effective volume, which affects the scintillation crystal, varied according to the place.

To determine the mean flow velocity we must use the velocity of the centre of gravity of the tracer distribution curve, but we found some difficulties in doing so. We then defined the time of arrival as the moment when the maximum value was given by the recorder of the rate-meter.

In Table I we can see the distance of each observation station from the
Rate-meter recordings of radioactivity measured directly at observation stations downstream, showing the time of arrival of the radioactivity in the second tracer experiment injection point, the time required by the flow from one station to the next, and the mean flow velocity at each station.

The use of a low-background counter enabled us to measure the specific activity of water samples up to 30 pc/1; in other words, we could determine the dilution rate of $5 \times 10^8$ as maximum in the case of a "spot" injection of sodium-24 solution.

In Fig. 6 we can see an example of the $\gamma$-ray spectrum of a water sample irradiated for 2 h. In this case the activity of bromine-82 ($E_\gamma 0.535 - 1.45$ MeV) was almost entirely hidden in the spectrum of the other activities.

First we measured the activity at the photoelectric peak of 1.38 MeV to determine the concentration of sodium. After 120 h of cooling the activity of bromine was determined (Fig. 6 (b)).

Because of the rather high concentration of sodium ions in the river water the use of sodium compounds as a tracer seems to be unfavourable; however, it affords a fair amount of information. We must choose a shorter irradiation time for the activation analysis of bromine in this case.
TABLE I

DISTANCES OF OBSERVATION STATIONS AND MEAN FLOW VELOCITIES AT EACH INTERVAL

<table>
<thead>
<tr>
<th>First tracer experiment</th>
<th>Designation of Station</th>
<th>Distance from the Injection point (km)</th>
<th>Distance between neighbouring stations (km)</th>
<th>Elapsed time of flow at each interval (min)</th>
<th>Mean flow velocity (cm/sec)</th>
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<tr>
<td>0a</td>
<td>0.9</td>
<td></td>
<td>0.9</td>
<td>135</td>
<td>11</td>
</tr>
<tr>
<td>1a</td>
<td>0.9</td>
<td></td>
<td>3.9</td>
<td>315</td>
<td>21</td>
</tr>
<tr>
<td>2a</td>
<td>4.8</td>
<td></td>
<td>6.5</td>
<td>400</td>
<td>27</td>
</tr>
<tr>
<td>3a</td>
<td>11.3</td>
<td></td>
<td>4.5</td>
<td>120</td>
<td>63</td>
</tr>
<tr>
<td>4a</td>
<td>15.8</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
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</table>

<table>
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<tr>
<th>Second tracer experiment</th>
<th>Designation of Station</th>
<th>Distance from the Injection point (km)</th>
<th>Distance between neighbouring stations (km)</th>
<th>Elapsed time of flow at each interval (min)</th>
<th>Mean flow velocity (cm/sec)</th>
</tr>
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<tbody>
<tr>
<td>0b</td>
<td>1.19</td>
<td></td>
<td>1.19</td>
<td>118</td>
<td>17</td>
</tr>
<tr>
<td>1b</td>
<td>1.19</td>
<td></td>
<td>1.64</td>
<td>129</td>
<td>21</td>
</tr>
<tr>
<td>2b</td>
<td>2.83</td>
<td></td>
<td>2.22</td>
<td>152</td>
<td>24</td>
</tr>
<tr>
<td>3b</td>
<td>5.05</td>
<td></td>
<td>2.72</td>
<td>212</td>
<td>21</td>
</tr>
<tr>
<td>4b</td>
<td>7.77</td>
<td></td>
<td>1.00</td>
<td>43</td>
<td>39</td>
</tr>
<tr>
<td>5b</td>
<td>8.77</td>
<td></td>
<td>4.73</td>
<td>150</td>
<td>52</td>
</tr>
<tr>
<td>6b</td>
<td>13.50</td>
<td></td>
<td>3.38</td>
<td>72</td>
<td>78</td>
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<tr>
<td>7b</td>
<td>16.88</td>
<td></td>
<td>1.33</td>
<td>85</td>
<td>26</td>
</tr>
<tr>
<td>8b</td>
<td>18.21</td>
<td></td>
<td></td>
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</table>
The second tracer experiment was made about three weeks later than the first one; its object was the use of NH₄Br solution for comparison of the utility of Br⁸⁰ with that of Br⁸² in activation analysis.

After the sampling of water fractions at each observation station, the specific activity of each fraction was determined at the laboratory by the use of a specially designed apparatus (Fig. 7).

The capacity of the vessel mounting around a NaI scintillation crystal is about 2l, which affords a good counting efficiency (about 60 times larger than that of a scintillation crystal of the well type). There is also another advantage of this arrangement, i.e. the water sample which fills the vessel works as a shielding material against the background radiation.

The sensitivity limit of measurement of the specific activity in the water samples in this vessel is about \(2 \times 10^{-3} \mu c/l\); that is, we can determine the dilution rate up to \(10^7\) in the case of the "spot" injection of a radioactive solution whose initial specific activity is \(2.46 \times 10^4 \mu c/l\).

The dots along the solid lines (a) in Fig. 8 represent the values measured by this method. The dilution rates higher than \(10^7\) were measured by the
"evaporation" method, the maximum sensitivity by means of a low-background flow counter being about 30 pc/l. We can see that the measured values obtained by this method are represented by the circles along the solid lines (a) in Fig. 8.

For determination of these specific activities, the calibration of the counting efficiency of each apparatus had been made beforehand by comparison of their counting efficiencies with that of a 4π gas-flow counter.

Neutron irradiation for 3 min produced γ-ray spectra, an example of which can be seen in Fig. 9.

Fig. 10 shows, on the other hand, a typical example of the decay curves whose components were determined by calculations.

In the spectrum shown in Fig. 9 several photoelectric peaks, whose energies are 0.51 MeV, 0.62 MeV and 1.37 MeV can be seen.

We assume then that the radionuclides Br$^{80}$, Br$^{80m}$, Br$^{82}$, Cl$^{38}$ and Na$^{24}$ exist. From the decay curve corresponding to this γ-ray spectrum we can also confirm the presence of these nuclides.

We can deduce the following equation from Eq. (1):

$$A e^{-0.0385t} + B e^{-0.000321t} + C e^{-0.00257t} + D e^{-0.00077t} + E e^{-0.0186t} = Z,$$

(2)

where $A$ = initial radioactivity of Br$^{80}$, $B$ = initial radioactivity of Br$^{82}$,
C = initial radioactivity of Br$^{80m}$, D = initial radioactivity of Na$^{24}$, and E = initial radioactivity of Cl$^{38}$.

The units of the times $t$ and $T$ in Eq. (1) must be the same, and in Eq. (2) the unit of the disintegration time $t$ is represented in minutes. The half-lives of disintegration of these radionuclides are 18 min (Br$^{80m}$), 36 h (Br$^{82}$), 4.5 h (Br$^{80m}$), 15.0 h (Na$^{24}$) and 37.3 min (Cl$^{38}$).

Since a particular decay curve gives the "gross" activity $Z$ at any time, we can solve equation (2) by applying the measured values of $Z$.

Each decay curve is divided into two or three parts, and we apply equation (2) to each separate part, neglecting the unnecessary terms of this
Gamma-ray spectrum of a water sample irradiated for 3 min and then cooled for 16 min
Sample 3
Neutron flux density, $3 \times 10^{14}$n/cm²s

Decay curve of the sample corresponding to Fig. 9 and those for component radionuclides determined by mathematical treatment of the measured gross activity.
equation for each part. We thus obtain the concentration of bromine by comparing the calculated activity with that of the standard sample of bromine.

In Fig. 8 we see the dilution configuration of bromine from the curves (b), which show dilution rates about 100 times lower than those obtained by the radioactive tracer sodium-24.

In other words, the dilution rates in the case of 'point' injection are 100 times greater than those of "continuous" injection.

After 6 h of irradiation of the similar water samples in a neutron flux of the same intensity, γ-ray spectrometry allowed us to determine the content of bromine. In this case we measured the activity of Br\(^{82}\) after 40 h of cooling of those samples. The curve in Fig. 11 shows an example of γ-ray spectra of the water fractions irradiated for 6 h and then cooled for 40 h. In these spectra we can see four photoelectric peaks at 0.55 MeV, 0.77 MeV, 1.04 MeV and 1.33 MeV, and the decay curves obtained from these peaks showed the well defined half-life of 36 h.

In Fig. 8 are shown the values obtained by activation analysis in which the results of shorter and longer irradiation are in good accord (curve (b)).
Growth and the decay curves for neutron irradiation of elements found in the river water. These activities were calculated by the estimated composition of these elements; however, the curves for bromine correspond to the tracer solutions after dilution of $2 \times 10^5$ times.

(a) 3 min irradiation followed by cooling
(b) 6 h irradiation followed by cooling
The practical lower limits of the dilution rate which can be determined by these two methods are $10^7$ and $3 \times 10^5$ respectively.

From these results we conclude that the 6-h irradiation time is more favourable in view of the accuracy and the simplicity of the method.

The concentrations of the elements in the river water which disturb the activation analysis of the tracer elements were determined to be $20 \sim 30$ ppm Na, $20 \sim 30$ ppm Cl and $0.1 \sim 0.5$ ppm Mn.

In Fig. 12(a) we see the growth curves of the radioactivities of these elements during 3 min of irradiation by neutrons and the subsequent decay curves. On the other hand, Fig. 12(b) shows similar curves but with much longer irradiation (6 h). In this example we assumed the existence of 20 ppm Na, 0.4 ppm Mn, 20 ppm Cl and 2 ppm Br.

If we use a solution of 200 l containing 100 kg of NH$_4$Br (including 83 kg of Br) as a non-active tracer, the 2 ppm of bromine which is found in the downstream water corresponds to a dilution rate of $2 \times 10^5$.

All in all, we thus determined the dilution configuration and the flow velocity along 18.2 km of this river.

IV. CONCLUSION

As the result of our experiment we can say that this activation method is quite useful even on a river containing much pollution and sea water.

The use of the short period irradiation imposes not a few problems, because the analysis of the decay curves is rather tedious and inexact; and, moreover, chemical separation cannot be applied for practical reasons.

The method which involves a rather long-period activation followed by cooling of the short-lived radioactivities is much more favourable, and in the case of bromine we believe that irradiation of $30 \sim 50$ h gives a better sensitivity.

If the pollution in the river is not noticeable, the sensitivity of the activation analysis increases; but in some cases it must be kept in mind that the concentration of tracer chemicals should not exceed their permissible concentration for drinking water.

We neglect on purpose the mathematical treatment of the results, because in our case the conditions of the river do not satisfy the diffusion equation of flow.

We should like to call attention to the fact that the diffusion rate augments very rapidly at the first stage of flow and then increases slowly. As to self-purification of the tracer elements, we ascertained, after simple experiments, that there was no physical, chemical or biological reaction, other than dilution, which affects practically the results of the tracer experiment.

ACKNOWLEDGEMENTS

The authors are indebted to the research members of our institute, S. Maeda, Y. Horiguchi, S. Nagatsuka, T. Sawai, K. Nakajima and others,
who co-operated with us. This work was supported by the Public Safety Control Division of the Tokyo Metropolitan Government and the Tokyo Metropolitan Fisheries Laboratory and was performed with the co-operation of their engineers, Y. Koido, T. Ito, T. Kajinuma, H. Furuse and others.

We also wish to thank the research members of the Atomic Energy Research Institute of Tokai and those of the Nuclear Research Institute of Saint Paul's University, who kindly operated their nuclear reactors for the activation analysis.

REFERENCES


DISCUSSION

G.H. LEAN: How does the accuracy of the dilution method used with non-active tracers compare with that obtained with active ones?

M. KOBAYASHI: I would say that the activation method is just as effective as the active-tracer method. It does, of course, give rise to various technical and economic problems. In certain special cases, e.g. when measurements are made in rivers in the neighbourhood of crowded cities, it is naturally preferable to the active-tracer method. It is certainly quite adequate for measuring low dilution rates.

J.F. CAMERON: It is possible to give a figure for the weight of tracer, e.g. ammonium bromide, which would be required to measure a flow rate of, say, 10 m$^3$/s to any accuracy of 1 to 2%? At what flow rate would the cost of the tracer be prohibitive?

M. KOBAYASHI: In our experiments we measured the velocity of the centre of the flow and the dilution rates. I do not think it is possible to establish any relationship between the flow rate and the dilution rate.

I am not sure what the economic limits of the method are but it can certainly be used in the case of small rivers such as are found in the area around Tokyo. With large rivers like the Tone, I should say that one or two tons of ammonium bromide would have to be used.

J.C. VOGEI (Chairman): The flow rate of the Tone river is, of course, much larger than 10 m$^3$/s mentioned by Mr. Cameron, so you would probably have to divide the value of one or two tons by 10 or 20, or whatever the right figure is.
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