I. INTRODUCTION

In case of major nuclear accidents when larger amount of radioactive material is released into the atmosphere vast areas can become contaminated by the nuclear fallout. The deposited radioactivity penetrates the soil in a complex manner: dry and wet deposition lead to different initial distribution patterns which are further modified by the later transport processes in the upper layers of the soil. The distribution is influenced by various factors (physico-chemical characteristics of the radioisotopes, soil type, weather conditions, environment etc.), the resulting soil profile is hardly predictable. An important lesson we learned from the Chernobyl reactor accident is the great variability of the contamination both in the extent of the deposition and in the penetration features.

In light of some recent studies the information on the radionuclide distribution in the soil has great importance in at least two different aspects:

a./ Some transfer coefficients describing the transport of the fallout radioactivity in different compartments of the environment are strongly dependent on the distribution pattern. A deeper penetration will decrease the resuspension of the radioactive material in the ground level air while radionuclides may become more available for root uptake by some plants. These competing processes influence the contamination of the vegetation in a manner not completely clarified, providing an important area for continuing research.

b./ The contamination of an area can efficiently be determined by the method of in situ gamma spectrometry. The conversion of the spectral information (line intensities) to surface contamination data is done by using factors, which are derived from model calculation.
based on certain assumed radionuclide distribution profile. The conversion factors can be calculated and tabulated for different depth profiles but something must be known about the real distribution in order to select the appropriate factors in a given case.

The conventional way of assessing the distribution of the radioactivity is taking soil samples and analysing them layer by layer. This approach is rather time consuming, tiresome and it leads to results not necessarily representative for a larger area. The main disadvantage of the sample analysis is the long time necessary for getting the estimate of the radionuclide distribution though this parameter would promptly be needed for the proper conversion of environmental spectra measured in situ.

In recent years - following the reactor accident in Chernobyl - an increased interest for rapid methods of monitoring environmental radioactivity was expressed. The International Atomic Energy Agency initiated a research project to co-ordinate the activities carried out in various laboratories aiming at the development of rapid monitoring procedures. The Co-ordinated Research Project (CRP) G6 10 01 under the title Rapid Instrumental and Separation Methods for Monitoring Radionuclides in Food and the Environment has given a frame for 11 research programs. The Health Physics Department of the KFKI Institute for Atomic Energy Research (the former Central Research Institute for Physics) has taken a part in this CRP with a project titled: Rapid In Situ Gamma Spectrometric Determination of Fallout Radioactivity in the Environment.

The main objective of our study was to find a method to estimate the penetration characteristics of the fallout radioactivity by using only spectral information obtained by the in situ spectrometric measurement thus avoiding the need for a long and tiresome sampling and sample analysis procedure.

In our studies special attention was assigned to the two Cs isotopes ($^{137}\text{Cs}$ and $^{134}\text{Cs}$) being the predominant long-lived gamma-emitting contaminants after the Chernobyl accident: most of the considerations are based on the special nuclear characteristics of these isotopes. Nevertheless, the investigations are aimed at finding a procedure that can be generalized for the case of any isotope assessable by the method of gamma spectrometry.
II. THEORETICAL BACKGROUND AND SPECTRAL INFLUENCE

The method of in situ gamma spectrometric determination of environmental radioactivity was elaborated by H.L. Beck et al. for NaI(Tl) detectors in 1964 [1] and developed for semiconductor detectors in the early seventies [2]. Since then many applications by various authors have been published [3-5].

The evaluation of environmental spectra measured by a detector situated 1 m above the ground in a down-looking position is based on the proportionality between the environmental concentration of a given gamma emitting radionuclide \( S_A \) and the count rate of the full energy peak of any of its gamma lines in the spectrum \( \hat{N}_f \). According to Beck the calibration factor \( \hat{N}_f / S_A \) can be expressed as a product of three terms, i.e.

\[
\frac{\hat{N}_f}{S_A} = \frac{\hat{N}_f}{N_0} \frac{N_0}{\Phi_0} \frac{\Phi_0}{S_A}
\]

Here \( \hat{N}_f / N_0 \) is the angular correction factor, i.e. the angular dependence of the relative detector efficiency \( R(\theta) \) (related to the efficiency at \( \theta=0 \)) folded with the angular distribution of the photon flux, \( \phi(\theta) \). \( N_0 / \Phi_0 \) is the photopeak count rate per unit flux of incoming photons parallel to the axis of symmetry of the detector (determined purely by the characteristics of the detector). \( \Phi_0 / S_A \) is the unscattered photon flux at the height of the detector per unit activity concentration in/on the soil, \( S_A \). The photon flux is given by

\[
\phi = \int \frac{S(z)}{4\pi r^2} e^{-\mu_a r_a - \mu_s (r-r_s)} dV
\]

where \( \mu_a \) and \( \mu_s \) are the linear attenuation coefficients of air and soil, respectively, \( r \) is the distance between the detector and the volume element \( dV \), \( r_a \) is the section of \( r \) in the air and \( S(z) \) is the activity concentration of the radionuclide in question at a depth \( z \) in soil.
$\dot{N}_0/\Phi_0$ is determined by calibration of detector efficiency using point sources of various gamma emitting nuclides. The angular correction factor $\dot{N}_f/\dot{N}_0$ is obtained by a combination of calibration measurement (determination of $R(\theta)$) and model calculation (determination of $\varphi(\theta)$). The quantity $\Phi_0/S_A$ is not dependent on the detector characteristics and can be computed by using different model assumptions for the environmental source distribution.

In general calculations are performed by assuming an exponential radionuclide distribution in soil

$$S(z) = S_0 e^{-\alpha z}$$

with concentration per unit area

$$S_A = \int_{0}^{\infty} S(z)dz = S_0/\alpha$$

Using this expression, an uniformly distributed source ($\alpha=0$) and a pure surface source ($\alpha \to \infty$) can also be described. Besides the reasonable assumption of exponential distribution (which is in accordance with many measurements of depth profiles) has the advantage that $\varphi(\theta)$ as well as $\Phi_0$ can be calculated analytically. In this case the unscattered flux per unit activity concentration $\Phi_0/S_A$ is expressed as:

$$\frac{\Phi_0}{S_A} = \frac{1}{2} \left[ E_1(\mu h) - e^{\mu_s} \cdot E_1(\mu h + \frac{\alpha h \mu_s}{\mu_s}) \right]$$

where

$E_1$ is the exponential integral of the first order and $h$ is the height of the detector above the ground,

therefore the energy dependence of $\Phi_0/S_A$ can easily be computed and is shown in Fig. 1 for different $\alpha$ parameters.
Fig. 1: The primary (unscattered) flux per unit activity concentration ($\Phi_0/S_A$) in the function of the photon energy for different $\alpha$ exponential source distribution parameters.

Fig. 2: The ratio of the primary (unscattered) flux per unit activity concentration ($\Phi_0/S_A$) at 662 keV to that at 32 keV energy as a function of the exponential distribution parameter $\alpha$. 
If there is no matter between the source and the detector the shape of the spectrum is determined by the properties of the photon-detector interaction. If the space is filled with absorbing material then the gamma radiation field is modified by two different effects, viz.:

- the attenuation of the primary (unscattered) photon flux,
- the build-up of the scattered gamma radiation field.

The $\alpha$ dependence of the unscattered flux vs. energy function offers a possibility for the estimation of the distribution parameter. If we select two gamma quanta of different energies emitted by the same nuclide, then the ratio of their primary (unscattered) fluxes will be a function of the $\alpha$ value. The more distant are the energies from each other the more pronounced is the effect. As an example the ratio of the primary (unscattered) fluxes per unit activity concentration at 662 keV and at 32 keV is shown in Fig. 2.

The drawback of the method is that it requires at least two, preferably distant energy gamma lines. This is a serious limitation for both Cs radionuclides. Most of the prominent lines of $^{134}$Cs are closely situated in the 500-800 keV energy region, thus only a slight effect can be anticipated. $^{137}$Cs has, on the other hand, a single gamma line at 661.6 keV, while the 32 keV X-ray is too low in energy to be detected by normal p-type HpGe detectors. The detection of this radiation requires n-type HpGe detector or an additional low energy photon (LEP) detector has to be applied.

So far only the peak intensities have been used, other possible information has not been extracted from the spectra. As it is obvious from the considerations above not only the characteristic lines but also the continuum of the energy distribution of the environmental radiation field undergoes considerable changes due to the photon-matter interaction processes. The scattered radiation forms a continuum between zero and the characteristic line as it is shown schematically in Fig. 3. The region on the low energy side of the characteristic line is a consequence of the forward photon scattering and its elevation is a function of the depth profile. The spectrum of a given line, measured by the detector, is the convolution of the energy distribution of the environmental radiation field and the
detector response function. The total spectrum is a superposition of the different spectrum components and, in general case, the 'discontinuous' peak regions sit on the 'continuous' background regions of other lines. The information on the amount of small angle scattering (and so, on the depth distribution) is inherited to the spectrum and is embedded in the step size (i.e. the difference between the counts in the region below and above the characteristic peak). The region adjoining the peak on the low energy side (a continuum between the Compton edge and the peak) is called the 'valley' and (since the 'valley' is increasing in the 'expence' of the decreasing peak during the scattering) the quantity defined as the peak area per 'valley' height (or 'peak-to-valley', in short) can be a good measure of the photon scattering of the gamma emitters distributed in the soil.

![Schematic presentation of the spectral distortion](image)

**Fig. 3**: A schematic presentation of the spectral distortion occurring due to photon scattering taking place in the matter between the source and the detector.
This method has the advantage that it is applicable also to single line isotopes. The probability of scattering in small angles is described by the Klein-Nishina expression [6]. In principle the expectable change in the step size or its relation to the total energy peak area (i.e. the 'peak-to-valley' ratio) can be calculated. In practice, however, a proper experimental calibration is a more straightforward way of getting the necessary values.

The main disadvantage of the method is that its applicability is strongly dependent on the intensity of the gamma line and on the spectral environment of the total energy peak (whether there are interfering lines in the 'valley' or above the peak).

On the basis of the above considerations the following possible gamma spectrometric methods for the estimation of the $\alpha$ source distribution parameter were investigated:

- Ratio of several lines measured by any Ge(Li) or HpGe detector ($^{134}$Cs)
- X-ray - main gamma line ratio measured by n-type HpGe ($^{137}$Cs)
- X-ray - main gamma line ratio measured by HpGe + LEP detectors ($^{137}$Cs)
- 'Peak-to-valley' ratio measured by any Ge(Li) or HpGe detector ($^{134}$Cs, $^{137}$Cs).

The first three approaches are simply different technical realizations of the same basic method: the determination of the ratio of different energy lines in the spectrum. The fourth type of assessment is essentially different from the others and it requires some further considerations.
III. CALCULATIONS FOR THE 'PEAK-TO-VALLEY' RATIO

Let \( N(E, \alpha) \) be the net peak area measured in the spectrum peak of energy \( E \) originating from a source distributed exponentially by depth in the soil with a distribution parameter \( \alpha \). Let us denote the 'valley' i.e. the elevation of the continuum in the properly selected \( \Delta E \) energy interval on the low energy side of the peak by \( I(\Delta E, \alpha) \). In fact

\[
I(\Delta E, \alpha) = \text{Int}(\Delta E, \alpha) - \text{Int}(\Delta' E, \alpha)
\]

where \( \text{Int}(\Delta E, \alpha) \) and \( \text{Int}(\Delta' E, \alpha) \) are the measured integral values in the energy intervals below \( \Delta E \) and above \( \Delta' E \) the peak energy \( E \), so \( I(\Delta E, \alpha) \) is the net step size on the presumably constant background continuum. Thus the 'peak-to-valley' ratio \( Q(E, \alpha) \) can be defined as

\[
Q(E, \alpha) = \frac{N(E, \alpha)}{I(\Delta E, \alpha)}
\]

Since the 'valley' is not only due to the scattering in the matrices between the source and the detector but also a result of the interaction of the photons with the detector, it is necessary to define the initial value of the 'peak-to-valley' ratio, \( Q_0(E) \):

\[
Q_0(E) = \frac{N_0(E)}{I_0(\Delta E)}
\]

Here \( N_0(E) \) and \( I_0(\Delta E) \) are the relevant quantities if only a monoenergetic parallel photon beam of energy \( E \) is applied.
The peak $N(E, \alpha)$

$N(E, \alpha)$ is the result of those photon transport cases when no interactions have taken place between the source and the detector, and the whole energy is absorbed in the detector. The probability $P_0$ of not being scattered is exponentially decreasing with the distance ($r$).

If the matrix is homogeneous (e.g. air) then

$$P_0(\mu_a, r) = e^{-\mu_a r}$$

where $\mu_a$ is the linear attenuation coefficient of the air.

In case of a more realistic situation where the radionuclides are distributed in the soil as an exponential function of the depth $z$, i.e. the concentration $S(z)$ is

$$S(z) = S_0 e^{-\alpha z}$$

then the unscattered photon flux ($\phi_0$) can be calculated as

$$\phi_0(E, \alpha) = \int_{V} \frac{S(z)}{4\pi r^2} P_0(\mu_a, \mu_s, r) \, dV$$

where $r$ is the distance between the volume element $dV$ and the detector

$P_0$ is the probability of not being scattered during the transport from $dV$ to the detector.
Fig. 4: Geometric parameters of the semi-infinite space model used for the calculation of the $\Phi/S_A$ conversion factors

Using the notations of Fig. 4, $\phi_0$ can be expressed as:

$$
\phi_0 = \int_0^\infty \int_0^\pi \frac{S_0}{4\pi r^2} e^{-\alpha(r\cos\theta-h)} \cdot e^{-\left(\frac{\mu h}{\cos\theta} + \mu_s \left(\frac{r-h}{\cos\theta}\right)\right)} \cdot 2\pi r^2 \sin\theta \, dr \, d\theta.
$$

Integrating the expression above we obtain

$$
\phi_0 = \frac{S_A}{2} \left[ E_i(\mu h) - e^\frac{\alpha h}{\mu_s} \cdot E_i(\mu h + \frac{\alpha h}{\mu_s}) \right].
$$
\[ N(E, \alpha) = \eta(E) \cdot \phi_0(E, \alpha) \cdot \Delta t \] (2)

(In this calculation the slight possible anisotropy of the detector response is ignored).

b. The 'valley' \( I(\Delta E, \alpha) \)

A photon of energy \( E \) emitted by the source may give a count in the energy region \( \Delta E \) of the 'valley' if it interacted once with the matter between the source and the detector by forward scattering thus losing a small fraction of its initial energy. The probability \( P \) of interacting only once, if the matrix is air, is as follows:

\[
P_1(\mu_a, r) = \int_0^r k \cdot \mu_a e^{-\mu_a x} dx \cdot e^{-\mu_a (r-x)} = k \mu_a r e^{-\mu_a r}
\]

where \( k \) is the fraction of the interacted photons scattered into the energy interval \( \Delta E \) and

\( x \) is the distance between the source and the point of interaction.

The flux of photons \( \phi_1 \) reaching the detector having a single forward scattering during the transport from the exponentially distributed source (Eq. 1) is expressed as

\[
\phi_1(\Delta E, \alpha) = \int \frac{S(z)}{4\pi r^2} P_1(\mu_a, \mu_s, r) \, dV
\]

where \( P \) is the probability of the photon being transported from \( dV \) to the detector having a single forward scattering to the energy interval \( \Delta E \).
With the notations of Fig. 4, $P_1$ reads

$$P_1 = \int_{0}^{r} k \mu_s e^{-\mu_s x} dx e^{-\mu_s \left( r - \frac{h}{\cos \theta} - x \right)} e^{-\mu_a \frac{h}{\cos \theta}} +$$

$$+ \int_{h}^{r} k \mu_a e^{-\mu_s \left( r - \frac{h}{\cos \theta} \right)} e^{-\mu_a \left( x - r + \frac{h}{\cos \theta} \right)} e^{-\mu_a \left( r - x \right)} dx =$$

$$= k e^{-\mu_s \left( r - \frac{h}{\cos \theta} \right)} e^{-\mu_a \frac{h}{\cos \theta}} \left( \mu_s \left( r - \frac{h}{\cos \theta} \right) + \mu_a \frac{h}{\cos \theta} \right).$$

Thus $\phi_1$ is

$$\phi_1 = \int_{0}^{\infty} \frac{k S_0}{4 \pi r} e^{-\alpha (r \cos \theta - h)} e^{-\mu_s \frac{h}{\cos \theta}} e^{-\mu_a \frac{h}{\cos \theta}} \left[ \mu_s \frac{h}{\cos \theta} + \mu_a \frac{h}{\cos \theta} \right] e^{-2 \pi r^2 \sin \theta dr dV}$$

The integration results in the following expression:

$$\phi_1 = \frac{k S_0 e^{-\mu_s h}}{2 \alpha \left( 1 + \frac{\mu_s h}{\alpha} \right)}$$

(3)

By introducing the following variables:

$$A = \frac{\alpha}{\mu_s}, \quad B = \mu_a h \quad \text{and} \quad S = \frac{S_0}{\alpha}$$
Eq. (3) can be written as:

\[ \phi_1 = \frac{kS_A}{2} \frac{e^{-B}}{1 + 1/A} \]

In the special case of plane source distribution (\(a \to \infty\) i.e. \(A \to \infty\)) the flux of forward scattered photons is

\[ \phi_1 = \frac{kS_A}{2} \cdot e^{-B} \]

If the source is distributed uniformly in the ground \((S(z) = S_0)\) then \(a = 0\) i.e. \(A = 0\) and

\[ \phi_1 = \frac{kS_0}{2\mu_s} \cdot e^{-B} \]

.. c., the 'peak-to-valley' ratio \(Q(E, \alpha)\)

The counts in the selected interval \((\Delta E)\) of the 'valley' (collected during the measuring time \(\Delta t\)) originate partly from the multiple scattering of the primary photons of energy \(E\) in the detector and partly from the full energy absorption of the forward scattered photons:

\[ I(\Delta E, \alpha) = I_0(\Delta E) + \eta(E) \phi_1(\Delta E, \alpha) \Delta t \]

(In this expression the slight difference of the full energy peak efficiencies at the energy position \(E\) and at the energy interval \(\Delta E\) is neglected).

The 'peak-to-valley' ratio \(Q(E, \alpha)\) is defined as:

\[ Q(E, \alpha) = \frac{N(E, \alpha)}{I(\Delta E, \alpha)} \]
In the first step it is easier to express the reciprocal of $Q(E, \alpha)$, using Eq. (2):

\[
\frac{1}{Q(E, \alpha)} = \frac{1}{Q_0(E)} + \frac{\phi(E, \alpha)}{\phi_0(E, \alpha)}
\]

\[
\frac{1}{Q(E, \alpha)} = \frac{1}{Q_0(E)} + k \cdot \frac{e^{-B}}{1 + 1/A}
\]

(4)

In case of plane source distribution ($A \to \infty$) the following formula can be derived:

\[
\frac{1}{Q(E, \infty)} = \frac{1}{Q_0(E)} + k \cdot e^{-B}
\]

For uniform source distribution Eq. (4) can be transformed to

\[
\frac{1}{Q(E, 0)} = \frac{1}{Q_0(E)} + k \cdot \frac{e^{-B}}{e^{-B} - B \cdot E_1(B)}
\]

Up to this point the calculations were general enough to be valid for any primary energy $E$ and energy interval $\Delta E$. For the computation of $Q(E, \alpha)$ in the function of $\alpha$ for $^{137}\text{Cs}$ the numerical values of $B$, $Q_0(E)$ and $k$ are needed. Further calculations will be made for the specific energy value $E = 661.6$ keV of $^{137}\text{Cs}$, while the $\Delta E$ and $\Delta \alpha$ intervals were set for the energy regions 631–649 keV and 672–690 keV, respectively. Since $B$ is defined as $B = \mu h$ its value for the standard detector height of 1m is

$B = 0.010$
$Q_0(E)$ is the 'peak-to-valley' ratio when no scattering between the source and the detector occurs. Its presence is a consequence of the multiple Compton scattering of the photons of energy $E$ in the detector and thus is a quantity depending mainly on the geometrical characteristics of the semiconductor diode. The determination of $Q_0(E)$ is best done by experiment: a point source of $^{137}$Cs positioned about 1m distant from the detector provides the required unscattered flux $\Phi_0$ without having significant buildup of the forward scattered photon flux $\Phi_1$. A typical value for a HpGe detector with relative efficiency of 20% used for in situ gamma-spectrometry is about 70.

The value of $k$ was determined by calibration measurements. An experimental set-up, shown in Fig. 5, was used to measure $Q$ in the function of the thickness of water between the source and the detector.

![Diagram](image)

Fig. 5: S c h e m e of the calibration arrangement for determination of $Q_0$ and $k$
Since the relevant quantities in the calibration arrangement above are

\[ \phi = \frac{S_0}{4\pi h^2} \cdot e^{-\mu_d} \]

\[ \phi_1 = \frac{kS_0}{4\pi h^2} \mu_w d e^{-\mu_w d} \]

thus

\[ \frac{1}{Q} = \frac{1}{Q_0} + \frac{\mu_d}{Q_0} \]

Using the measured values of Q (shown in Fig. 6) and the linear attenuation coefficient for water:

\[ \mu_w = 0.085 \, \text{cm}^{-1} \]

the following parameters were obtained by regression analysis:

\[ Q_0 = 73.5 \]

\[ k = 0.052 \]

Fig. 6: 'Peak-to-valley' ratios (Q) in the function of water thickness, measured in the geometry of Fig. 5.
Substituting the numerical values of $B$, $Q_0$ and $k$ in Eq. (4) $Q = Q(661.6 \text{ keV}, \alpha)$ can be calculated for any value of $A$, i.e. $Q$ can be plotted in the function of $\alpha$ (for the calculations the value $\mu_s = 0.12 \text{ cm}^{-1}$ was used). $Q$ is a monotonously increasing function of $\alpha$ between the extreme values of 14.6 for the uniform distribution ($\alpha \to 0$) and 37.0 for the plane distribution ($\alpha \to \infty$). (See curve (a) of Fig. 10 for the values of $Q$).

d., Modification of the exponential distribution model

Soil column measurements and compartment model calculations have shown that the simple exponential distribution is just a rough estimation of the real concentration profile. The departure from exponentiality is a function of time, soil structure, chemical composition, weather etc. Both theory and experiments prove that the migration of the radionuclides in the soil affects the distribution so, that the maximum concentration moves downwards and the uppermost layers become more and more depleted. Fig. 7 shows typical concentration profiles obtained by analysis of soil columns collected in the contaminated areas of the USSR, 4 years after the Chernobyl accident.

Fig. 7: Concentration profiles of $^{137}\text{Cs}$ in the upper layers of soil in some samples originating from the contaminated areas of the USSR
Since the top layer of the soil plays an essential role in the attenuation of the unscattered flux and in the build-up of the scattered radiation field the model used for the calculation of $Q$ has to be modified.

To allow for the depletion of the top layers while keeping the assumption of exponentiality for the underlying soil the following radionuclide concentration profile is proposed:

$$S(z) = S_0 e^{-az} - S_1 e^{-\beta z} = S_0 e^{-az} \left(1 - \frac{S_1}{S_0} e^{-\left(\frac{\beta}{\alpha} - 1\right)az}\right)$$ (5)

In this model (beside the original parameters of $S_0$ and $\alpha$) two additional parameters ($S_1/S_0$ and $\beta/\alpha$) are introduced: $S_1/S_0$ setting the proportion of depletion and $\beta/\alpha$ describing the dynamics of approaching exponentiality.

Fig. 8 demonstrates the influence of the new parameters on the distribution pattern, while Fig. 9 shows the matching between measured data and values calculated on the basis of the modified exponential model.

Performing the calculations of $\phi_0$ and $\phi_1$ for the modified exponential distribution the following expressions can be obtained:

$$\phi = \frac{S_0}{2a} \left[E_1(\beta) e^{\alpha B} - E_1(\beta + B) - \frac{S_1}{S_0} \frac{\alpha}{\beta} \left[E_1(\beta) e^{\alpha B} - E_1(\beta + B + B)\right]\right]$$

$$\phi_1 = \frac{kS_0}{2a} e^{-B} \left(\frac{1}{1 + \frac{1}{A}} - \frac{1}{S_0} \frac{1}{\beta/\alpha + \frac{1}{A}}\right)$$

By substituting $\phi_0$ and $\phi_1$ into Eq. (4) one gets:

$$e^{-B} \left(\frac{1}{1 + \frac{1}{A}} - \frac{S_1}{S_0} \frac{1}{\beta/\alpha + \frac{1}{A}}\right)$$

$$Q = \frac{1}{Q_0} + k \cdot \left(E_1(\beta) e^{\alpha B} - E_1(\beta + B) - \frac{S_1}{S_0} \frac{\alpha}{\beta} \left[E_1(\beta) e^{\alpha B} - E_1(\beta + B + B)\right]\right)$$
Fig. 8: Typical profiles of the modified exponential distribution ($\alpha = 0.5$), (a) for different $S_1/S_0$ values at $\beta/\alpha = 2.0$ and (b) for different $\alpha/\beta$ values at $S_1/S_0 = 0.9$.

Fig. 9: Comparison of the measured depth distribution of the Cs-137 concentration with the predicted values of the simple (a) and the modified (b) exponential model.
The functions of Q versus $\alpha$ for different values of $S_1/S_0$ and $\beta/\alpha$ are plotted in Fig. 10 (curves (b)-(d)). A significant difference can be observed between the curves of the modified exponential distribution and that of the simple exponential model (curve (a)), especially in case of low $\alpha$ values. The proportion of depletion ($S_1/S_0$) has a primary importance, the influence of $\beta/\alpha$ is much more a smaller perturbation of the basic function characterized by the $S_1/S_0$ parameter.

![Fig. 10: Calculated values of the 'peak-to-valley' ratio (Q), for different $S_1/S_0$ parameters, in comparison with some experimentally determined Q values ($\beta/\alpha$ is set to 2.0)](image)

It is to be emphasized that it is, in principle, impossible to determine $\alpha$ from a single measurement of Q, as long as $S_1/S_0$ and $\beta/\alpha$ are free parameters. It gives the possibility, however, that a range for possible values of $\alpha$ can be given. If further investigation of the transport processes of radionuclides in the soil can establish correlation between $S_1/S_0$, $\beta/\alpha$ and $\alpha$, then a more accurate estimation of these distribution parameters will be obtainable.
To check the applicability of the model a set of measured data is also shown in Fig. 10. In these cases Q values obtained by in situ spectrometry are plotted versus $\alpha$ determined by laboratory gamma-spectrometric analysis of soil samples collected at the spot of the in situ measurements. Most of the measured data correlate well with the curves of perfect (100%) or substantial (90%) depletion, and even those data (e.g. set No. 12 in Fig. 7) which were close to exponentiality fell below the curve of the simple exponential model (curve (a)). A possible explanation for this phenomenon is that the relatively inactive vegetation acts as a depleted layer shifting the measured Q values towards the predictions of the modified exponential model.

Another consequence of the discussed departure from exponentiality is the slight change of $\Phi_0/S_A$ which is an essential quantity in calculating the $N_f/S_A$ conversion factors. Here

$$S_A = \frac{S_0}{\alpha} \left( 1 - \frac{S_1}{S_0 \beta} \right)$$

is the total activity per unit area, the quantity we are normally looking for. Thus

$$\frac{\Phi}{S_A} = \frac{E_1(B) - e^{AB}E_1(AB+B) - \frac{S_1}{S_0 \beta}}{2 \left( 1 - \frac{S_1}{S_0 \beta} \right)} \frac{\alpha}{S_1} \left[ E_1(B) - e^{\alpha A} E_1(B_{AB+B}) \right]$$

This expression will give the usual definition of $\Phi/S_A$ if $S_1/S_0 = 0$ or $\beta/\alpha \rightarrow \infty$, i.e. the distribution is approaching exponentiality. Values of $\Phi/S_A$ for different $S_1/S_0$ values are presented in Fig. 11.

As it was pointed out only a range of possible $\alpha$ values can be given (if no other information on the distribution is available) on the basis of the Q measured. It is imperative, however, that whatever $\alpha$ is selected, the $\Phi/S_A$ value for the conversion is to be used accordingly. There is, fortunately, a compensating tendency in this procedure: though $\alpha$ may vary considerably depending on the $S_1/S_0$ and
\(\beta/\alpha\) parameters the \(\Phi/S_A\) factors will not differ so much if properly selected. As an example: if \(Q=20.0\) is measured then \(\alpha\) varies between 0.20 cm\(^{-1}\) (simple exponential case) and 0.55 cm\(^{-1}\) (modified exponential, \(S_i/S_0 = 1.0, \beta/\alpha = 2.0\)) meanwhile the \(\Phi/S_A\) values are 0.46 and 0.53, respectively. It means that the uncertainty of the distribution does not affect strongly the in situ spectrometric determination of the contamination. There is, however, a potential risk to slightly overestimate the total deposition if the simple exponential model is used.

![Graph](image)

**Fig. 11:** \(\Phi/S_A\) conversion factor for different \(S_i/S_0\) parameter values (\(\beta/\alpha\) is set to 2.0)

It is to be emphasized that the selection of the model function Eq. (5) of the modified exponential distribution is not based on theoretical consideration. Its use is justified since it can account for the top layer depletion and has the advantage of easy mathematical treatability. Eq. (5) has, however, a serious disadvantage: it cannot describe a deeper penetration of the maximum with high \(\alpha\) value. With the passing of the time the migration of the concentration maximum may necessitate the further modification of the model.
IV. APPLICATIONS

During the recent years two expeditions were made to areas with high radioisotope contamination of Chernobyl origin:

a./ A measuring trip was organized to some selected parts of Austria, where the contamination was significantly higher than in Hungary and the distribution of the radionuclides in the soil was well known even before the measurements.

b./ In situ gamma spectrometric measurements were made in the USSR during an experts' mission to the most affected areas organized by the IAEA. In parallel with the in situ measurements soil samples were taken in order to determine the distribution profile by later laboratory analysis.

In the two missions two different measuring equipments were used so the calibrations for the spectrometers had to be performed twice. Both detectors were of the same type (reversed HpGe with extended sensitivity in the X-ray energy region) which condition enabled us to test both methods described in Section II.

As mentioned in Section III soil sample analyses showed that in most cases the exponential distribution model is just a rough approximation of the real soil profile. There are two major characteristic features which result in significant departure from exponentiality:

- In deeper layers of the soil the decrease of the radionuclide concentration is 'slower', a second exponential component with smaller $\alpha$ value below 10-15 cm is to be used to get a better fit to the distribution.

- A time-dependent desorption effect causes the concentration maximum to migrate from the surface to the deeper layers. The speed of the displacement is a function of the environmental parameters (soil structure and composition, chemical form of the contaminant, annual rainfall etc.) and it results in a 'hump' on the originally exponential distribution.

The first factor does not influence too much the in situ determination of the fallout radioactivity. The proportion of radioactivity below 15 cm to the total deposition is only a few
percent and its contribution to the primary flux measured by in situ gamma spectrometry is negligible in the whole energy region of interest. The second phenomenon, however, is certainly an explanation why the method of X-ray to main-gamma-line ratio did not work under real conditions. The X-ray line intensities were significantly less than expected and in some cases peak at 32 keV could not be detected at all, mostly because of the intense attenuation of the X-rays in the uppermost layers. The deviation of the 'X-ray to main gamma line' ratio from the expected values, however, can be an indication of the 'hump' in the soil profile so even the failure of the method can provide some information about the radionuclide distribution.

On the other hand the method of 'peak-to-valley' ratio provided results in good agreement with the soil sample measurements. Fig. 10 shows examples for the matching between the data of the two types of analysis, if the modified exponential distribution model is used.

The procedure works for superscript 137Cs (and as long as the isotopic ratio of the original contamination holds also for superscript 134Cs) with some limitations, however. At the present values are in the range of 0.1 - 1.0 cm\(^{-1}\) (relaxation length 1 - 10 cm) and we have no experience beyond this range. Interference of other nuclides can disturb the determination of the 'valley' (636 keV line of superscript 125Sb, for instance) or of the peak (658 keV line of superscript 110mAg, etc). A further limit of the application is the level of contamination: pulse pile-up problems caused by high intensity radiation lead to distortions in the spectrum. A significant line broadening is expected in fields where the superscript 137Cs contamination is in the order of 10\(^3\) kBq/m\(^2\) when detectors of about 20\% relative efficiency are used.

It is to be noted that the in situ measurements in the USSR provided information about the probable distribution much earlier than the soil sample analysis and this information was used to calculate a corrected value for the total deposition. This way these measurements can be considered as the first application of the methods developed under real field conditions and the method of the 'peak-to-valley' ratio proved to be very efficient. For this reason it is not worthless to give some further details of the measurements made during Mission 5 of Task 2 (devoted to the assessment of environmental contamination) within the framework of the International Chernobyl Project, coordinated by the IAEA [7].
Measurements in the affected areas of the USSR

The objectives of Mission 5 (part of the efforts to perform independent radiological survey to determine the contamination on selected sites of the affected areas in the USSR) gave an excellent opportunity to test and apply the method of 'peak-to-valley' ratio under realistic conditions. During the mission (in the summer of 1990) the international team of 8 experts visited 3 settlements in areas contaminated by the Chernobyl fallout: Bragin (BSSR) and Polesskoje (UkrSSR) in the controlled zone of higher contamination (15-40 Ci/km²) and Daleta (UkrSSR) in a less contaminated area ( < 5 Ci/km²). (Former activity units are used in accordance with the official data provided by the USSR authorities). Two complete HpGe gamma spectrometric systems were operated by two groups of experts independently. Some measurements were made in the same place to give an opportunity for the comparison of the results but the groups usually worked separately. The experts of Working Group 1 (WG1) (I. Winkelmann and M. Schweigert from the Bundesamt fuer Strahlenschutz, Institute fuer Strahlenhygiene, Neuherberg and H. Sitter from Kernforschungszentrum Karlsruhe) operating one of the systems took soil samples for the determination of by later laboratory analysis, while Working Group 2 (WG2) (P. Zombori from the Central Research Institute for Physics, Budapest and H. Lettner from University of Salzburg) used the in situ spectra measured during the mission to determine the parameters of the radionuclide distribution. Some soil samples for laboratory analysis were also taken by WG2. The distribution parameters determined by the two different methods as well as the contamination values obtained by the two groups were later compared.

Nine spectra were available for the study of the radionuclide distribution by the method of 'peak-to-valley' ratio. Table 1 summarizes the Q values determined and sets of possible distribution parameter at different proportion of depletion $S_1/S_0$, in comparison with the values provided by I. Winkelmann on the basis of his soil sample analyses. This comparison confirmed the concept of the modified exponential distribution: in general, parameters measured by sample gamma spectrometry were closer to those values derived from the 'peak-to-valley' ratio by assuming substantial depletion in the top soil layer.
Knowing the proper distribution parameters the corresponding value of $\Phi/S_A$ can be selected, thus the most appropriate conversion factor $N_f/S_A$ is available. On the basis of the analysis outlined above the activity concentration and the dose rates at the selected measurement sites were determined (see Table 2) and the data were compared to the results of I. Winkelmann and his group (Fig. 12).

Fig. 12: Comparison of the Cs contamination data measured by two in situ gamma spectrometric working groups (WG) of Mission 5 in selected sites of the contaminated areas of the USSR

Concerning the isotopic composition and the quantitative characterization of the contamination the following statements can be made:

As expected, $^{137}$Cs was predominant in the composition, while $^{134}$Cs was always found in a concentration 6-7 times less at the time of measurement (July-August 1990). Due to their nuclear characteristics $^{134}$Cs had a relatively higher dose rate contribution (38 %), while 59% of the dose rate was attributable to $^{137}$Cs. Because of the longer half life this relative contribution will increase in the future.
Beside the Cs-isotopes some other radionuclides (\(^{60}\text{Co},^{106}\text{Ru},^{125}\text{Sb},^{144}\text{Ce}\)) could also be detected in the environment, but their contribution to the total dose rate was in the order of 1% or less. The ratio of \(^{144}\text{Ce}\) concentration to \(^{137}\text{Cs}\) concentration was not constant indicating a different type of contamination: while volatile Cs isotopes are more evenly distributed in the environment, \(^{144}\text{Ce}\) may be contained in particles of the fuel element and so, distributed in a more discontinuous manner. (Fig. 13 shows a typical example for the isotopic composition of the contamination)

![Diagram showing relative contributions and activity concentrations of different isotopes.](image)

<table>
<thead>
<tr>
<th>Isotopes</th>
<th>Dose rate</th>
<th>Act. conc.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs-137</td>
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<td>76.984%</td>
</tr>
<tr>
<td>Cs-134</td>
<td>38.261%</td>
<td>11.548%</td>
</tr>
<tr>
<td>Ce-144</td>
<td>0.174%</td>
<td>5.065%</td>
</tr>
<tr>
<td>Ru-106</td>
<td>1.565%</td>
<td>5.065%</td>
</tr>
<tr>
<td>Sb-125</td>
<td>0.87%</td>
<td>1.317%</td>
</tr>
<tr>
<td>Co-60</td>
<td>0%</td>
<td>0.1%</td>
</tr>
</tbody>
</table>

Fig. 13: Isotopic composition of the contamination measured by field gamma spectrometry in the contaminated areas of the USSR

\(^{137}\text{Cs}\) contamination data measured by in situ gamma spectrometry were again compared to the official data shown on the USSR contamination maps. In Fig. 14 measured \(^{137}\text{Cs}\) activity concentrations generally lie within or below the range given by the soviet officials. Considering the great variability of the fallout and the random nature of the measurement site selection the agreement is very good.
Fig. 14: Comparison of $^{137}\text{Cs}$ contamination data obtained by field gamma spectrometry with official USSR data (|: in situ measurements, □□□□ map categories)

V. SUMMARY

The distribution of fall-out radioactivity in the upper layers of the soil has a strong effect on the ecology of the radionuclides and influences the conversion of the spectral information (peak intensities measured in situ) to surface contamination and dose rate contribution. The radionuclide concentration profile is usually determined by the time consuming soil sampling and sample analysis procedure which hinders the rapid evaluation of a larger scale environmental contamination. A growing interest in speeding up the radioanalytical work made this issue a major objective of coordinated research projects.

To obtain a fast and reliable estimation of the actual distribution pattern some new $\gamma$-spectrometric methods (based on the spectrum shape deformation due to scattering and absorption in matter) were investigated. The observation that deeper penetration correlate with higher 'valley' (i.e. the continuum on the lower energy side of a
characteristic peak in the in situ spectra) and some theoretical
considerations and calculations concerning the small angle (forward)
scattering of the photons provided a novel procedure (called the
method of 'peak-to-valley' ratio) which proved to be efficient in
accomplishing the objectives above.

The ratio of 'peak-to-valley' \( (Q) \) as a function of the
radionuclide depth distribution parameter \( \alpha \) was initially defined and
calculated for the case of simple exponential concentration profile.
Some calculations and experimental studies revealed, however, that the
real distribution is different from the simple exponential one, and
the distribution model used for the calculations had to be changed.
The modified exponential model, given in the paper, has the potential
to describe the real concentration profiles typical for most of the
soil types a few years after a major nuclear accident (4-5 years after
Chernobyl in the present case). Formulae and plotted graphs are given
in the paper to convert the experimentally determined \( Q \) values to
different sets of parameters.

The practical implementation of the method is very easy. Both peak
areas and 'valley' integrals can be obtained immediately after the
measurement and the 'peak-to-valley' ratios can be converted to \( \alpha 
\) distribution parameters using the formulae or \( Q(\alpha) \) plots given. The
application can be disturbed, however, if the 'valley' or the peak
interfere\( \frac{1}{\text{E.g.}} \) with other peaks, or if the intensities are too low to get
statistically significant counts, especially for the 'valley'. It is
also to be noted that only a range of possible parameter values can be
given, but the consequent use of the distribution parameters and the
corresponding conversion factors reduces the error of the
contamination measurement by in situ \( \gamma \)-spectrometry.

The new method has been tested in measurements over areas of known
high activity concentration in Austria and applied under realistic
conditions during a mission to the contaminated areas of the Soviet
Union. When compared to independent analyses a good agreement both for
the distribution parameters as well as for the total contamination was
obtained.
ACKNOWLEDGEMENT

The authors of the paper are grateful to Dr. H. Lettner for his co-operation and Dr. I. Winkelmann for providing his valuable data for the purpose of the comparison.

REFERENCES


Table 1
Comparison of the distribution parameters determined by the 'peak-to-valley' ratio and obtained by soil sample analysis (β/α parameter is fixed to be 2.0)

<table>
<thead>
<tr>
<th>Site No.</th>
<th>Description</th>
<th>Parameters</th>
<th>( S_{1}/S_{0} )</th>
<th>( \alpha )</th>
<th>( \beta )</th>
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<td></td>
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<td></td>
<td></td>
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<td>-</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>0.9</td>
<td>&lt; 0.03</td>
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Table 2

Results of the in situ gamma spectrometric measurements made in Bragin/Daleta/Polesskoye during Mission 5 (dose rates are compared to data measured by a dose rate meter SSM1)

<table>
<thead>
<tr>
<th>Spectrum code</th>
<th>Location</th>
<th>Site description</th>
<th>Act. conc. (kBq/m²)</th>
<th>Dose rate (μSv/h)</th>
</tr>
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<tbody>
<tr>
<td>BRAGO001</td>
<td>Bragin</td>
<td>Sportsground</td>
<td>Cs-134: 54</td>
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<td>Ru-106: 44</td>
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