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INTERNAL RADIOACTIVE CONTAMINATION IN SELECTED GROUPS OF CRNL EMPLOYEES

by

D.W.S. EVANS

Chalk River Nuclear Laboratories

Chalk River, Ontario

October 1975

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Contamination radioactive interne chez certains groupes d'employés de Chalk River

par

D.W.S. Evans

Résumé

Ce rapport décrit en détail le développement et l'exécution d'un programme de 30 mois conçu pour caractériser l'importance et la répartition de la contamination radioactive interne chez certains groupes d'employés des laboratoires nucléaires de Chalk River au moyen d'un anthroporadiamètre à écran partiel.

Les résultats montrent que les niveaux de contamination chez ces employés sont très faibles et aucun contaminant n'était présent en quantités excédant 10% de la charge corporelle maximale admissible, à l'exception d'un radionucléide (sélénium 75) administré pour fins médicales.

On fournit également des précisions sur les périodes effectives de quelques charges corporelles.

L'Energie Atomique du Canada, Limitée
Laboratoires Nucléaires de Chalk River
Chalk River, Ontario
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ABSTRACT

This report details the development and execution of a 30 month program designed to characterize the magnitude and distribution of internal radioactive contamination amongst selected groups of employees at Chalk River Nuclear Laboratories, using a shadow shield whole-body counter.

The results show that the levels of contamination in these employees are very low, and no contaminant was present in amounts exceeding 10% of the maximum permissible body burden, with the exception of a medically administered radionuclide (selenium-75).

Details of the time course of some of the body burdens are also furnished. (author),

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In 1972, a shadow shield whole-body counter was constructed at CRNL in order to facilitate the assessment and control of internal contamination of CRNL employees with radionuclides. Previously the distance between CRNL and the large whole-body counter 12 miles away at Deep River had caused considerable difficulties in conducting an adequate monitoring program. As a primary objective, a detailed study of the extent of the contamination amongst those considered to be at highest risk was initiated in September, 1972, and concluded in March, 1975, with the transfer of the counter to a permanent location. A total of more than 1300 whole-body counts were made, and it is appropriate at this stage to evaluate the apparatus, procedures in current use and to draw some conclusions from the experience gained.

EQUIPMENT

The shadow shield counter is patterned after that of Palmer and Roesch (1) and in the period covered by this report, was located in the basement of the Electronics Building (600). The detector assembly consists of a 29.2 cm x 10.2 cm thallium activated sodium iodide crystal clad in stainless steel, and optically coupled to 7 matched photomultiplier tubes. The 10 cm thick plate supporting the lead castle (which was used instead of a thinner plate because of ready availability) houses the crystal in a central hole, and the lower surface of the crystal is almost flush with that of the plate. The clearance between the crystal face and upper surface of the padded trolley is about 30 cm, a dimension sufficient to permit the passage of all but the grossly obese. The counter length is also sufficient to permit the scanning of bodies of up to ~75 inches height.

A whole-body scan is made by passing the subject, lying on a padded trolley, at a constant speed beneath the shielded crystal. Counting begins when the subject's feet interrupt a photobeam 15 cm away from the crystal axis and ends when the head ceases to obstruct a second photobeam in a similar position on the opposite side of the crystal. Each subject is counted in a prone position in one direction and supine in the other, the time involved being approximately 20 minutes.

The detector signals are fed into a multi-channel pulse height analyzer and the data is stored in one quarter of the memory (256 channels at 10 kev/channel). Accessory equipment including a magnetic tape memory and teletypewriter with punched tape output permits spectrum stripping, computer printout of the spectra and other manipulation of the data.

More recently, a row of 7 collimated 5 cm x 5 cm sodium iodide crystals and photomultipliers has been installed adjacent to the lead castle. A count rate meter coupled to an x-y plotter records the output and facilitates the longitudinal localization of radioactivity in or on the body. The degree of collimation can be varied as required.

CALIBRATION

For purposes of radiation protection, the demands for accuracy in the measurement of internal radioactive contamination are not stringent, and an accuracy of ± 25% is stated to be acceptable (2).

Calibration of the shadow shield counter was effected by scanning solutions of various radionuclides homogeneously distributed in a polyethylene bottle phantom (~170 cms height, ~70 kg water filled). The radionuclides included ^{203}Hg, ^{51}Cr, ^{137}Cs, ^{54}Mn, ^{60}Co and ^{40}K, encompassing a γ energy range of 0.279 to 1.460 MeV. Calibration constants for other radionculides encountered were derived where necessary by interpolation on the calibration graph. The sensitivity of the counter for potassium, based upon scans of the bottle phantom containing 140 g potassium and counting times of about 20 minutes is ~0.83 (c/m)/g* potassium. The background count rate in this period is approximately 186 c/m, yielding a standard deviation of ~4%. For caesium-137, the sensitivity is approximately 11300 (c/m)/μCi. The minimum detectable activity, defined as that amount of activity producing a total count which differs from the background count by three times the standard deviation of the latter is about a nanocurie for radionuclides with 100% γ yield/disintegration.

The practical assumption of a constant counting efficiency at a given γ energy is a major source of error in the quantitation of internal contamination, since the wide range in size and shape of bodies containing radioactivity distributed spatially in an ill-defined or unknown manner necessarily is associated with variation in the counting efficiency of the detector. To illustrate the human variation, the height of the subjects examined ranged between 152 and 188 cm, the weight between 44 and 145 kg and the antero-posterior dimension at the xiphisternum between 19 and 31 cm. To some extent, the effect on counting efficiency of displacement of radioactivity from the body axis in vertical and horizontal planes is reduced in the shadow counter by the prone-supine counting procedure, and the large diameter of the sodium iodide crystal (29.2 cm) relative to the breadth of the average man. It is also relevant that the trunk is the region of greatest concern.

A crude estimation of the variation in counting efficiency with source location was obtained by suspending a "point" source of cobalt-60 at the centroids of various parts of the water-filled phantom, and counting in the usual manner. The results, normalised to the counting efficiency obtained with the source located in the thoracic section of the phantom are shown in the following Table, and indicate a relatively small range of values.

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*c/m is used in this report for counts/minute

TABLE I: Variation of Counting Efficiency with Source Location

LOCATION OF SOURCE	RELATIVE COUNTING EFFICIENCY
Head	0.88
Thorax	1.00
Abdomen	0.87
Arm	0.77
Thigh	1.10
Leg	0.92

Similar results were obtained in the absence of a scattering medium; lateral displacement of a "point" source on the floor of the counter 15 cm from the crystal axis resulted in a 17% decrease in counting efficiency.

An example of the uncertainty introduced by the use of calibration factors uncorrected for geometry and absorption is afforded by the results of measurements of body potassium in 60 subjects grouped in three different ranges of body weight, shown below in Table II.

TABLE II: Body Potassium and Body Mass

n	MEAN WEIGHT AND S.D. kg	MEAN BODY K AND S.D. g	BODY K g/kg
20	55.5 ± 5.1	107.0 ± 13.3	1.92
20	75.0 ± 6.5	124.6 ± 10.5	1.66
20	96.7 ± 7.1	126.9 ± 9.1	1.31

There is no statistically significant difference between the apparent potassium content of subjects with an average weight of 75 kg and those about 22 kg heavier, although the ratio body K/body weight clearly reflects differing body composition.

As a more realistic check on the accuracy of the shadow counter measurements, several employees who had occasion to visit other nuclear establishments were counted in various types of whole-body counters. The results of these intercomparisons, shown below in Table III, indicate satisfactory agreement between the measurements of potassium in 5 men weighing between 65 and 86 kilograms. Agreement between measurements of the body caesium-137 was good at "high" levels (between two shadow counters), but poorer at levels of a few nanocuries, near the detection limit.

It is relevant that the 95% confidence limits (counting statistics only) in a 20 minute count for a body burden of 10 nCi of caesium-137 in the presence of 140 g potassium are approximately ± 9%, and for 1 nCi of caesium-137, the approximate detection limit, ± 80%.

TABLE III: Intercomparison of Calibration for Potassium-40 and Caesium-137

Age (a)	Weight (kg)	K (g/kg)	^{137}Cs (nCi)	Counter
35	65.75	2.19	3.8	(1)
		2.27	2.1	CRNL
39	61.70	1.87	2.4	(1)
		1.87	1.8	CRNL
51	67.60	1.87	3.4	(1)
		1.88	2.2	CRNL
50	72.50	1.85	3.4	(2)
		2.02	3.4	(3)
		1.81	3.9	CRNL
46	86.20	1.95	7.1	(3)
		1.78	4.9	CRNL
57	76.20	--	148	(4)
		--	134	CRNL
41	81.67	--	342	(4)
		--	336	CRNL

(1) Multidetector stretcher geometry, steel room, Brookhaven National Laboratory, Upton, New York.

(2) Single detector linear scan, shadow shield, Pacific Northwest Laboratory, Richland, Washington.

(3) Single detector chair geometry, steel room, Pacific Northwest Laboratory, Richland, Washington.

(4) Single detector linear scan, mobile shadow shield, Ontario Hydro.

It is concluded that despite systematic and other errors, the relatively crude calibration of the counter is adequate for purposes of radiation protection.

BACKGROUND

The background counting rate of the shadow counter is relatively high and variable, a consequence of the geology and topography of the area, and the argon-41 emission from the reactors. Practically, counting proved possible only when the wind was in a direction appropriate to remove argon-41 from the counter site. In still air, interference occurred unpredictably, particularly under inversion conditions, and on occasion, the argon-41 interference associated with stable weather patterns rendered counting impossible for periods of up to three weeks.

The Table below illustrates the relatively high background counting rate of the CRNL counter compared with two other shadow shield counters with similar detectors (29.2 cm x 10.2 cm sodium iodide crystals) and crystal - stretcher distances.

TABLE IV: Background Counting Rate of 3 Shadow Shield Counters

	Counter A (Ref. 3)	Counter B (Ref. 4)	CRNL Counter
$(c/m)/cm^3$ Crystal Volume (0.1 - 2.0 MeV)	0.439	0.520	0.580 (0.255)
$(c/m)/^{40}K$ Energy Band (1.36 - 1.56 MeV)	137	173	186 (156)
$(c/m)/^{137}Cs$ Energy Band (0.60 - 0.72 MeV)	---	196	179 (117)

The bracketed figures in the table were measured during construction of the counter, with the crystal and photomultipliers totally enclosed by 4 inches of lead. It is also of interest that the count rate in the potassium-40 energy band is about twice as high as that of the iron room counter in Deep River (~98 c/m).

Figure I shows a 40,000 second background count. Apart from the prominent ^{40}K peak, some of the various peaks associated with members of the uranium-238 and thorium-232 decay series are visible. Annihilation radiation is also present, and possibly a trace of cobalt-60. In the ab-

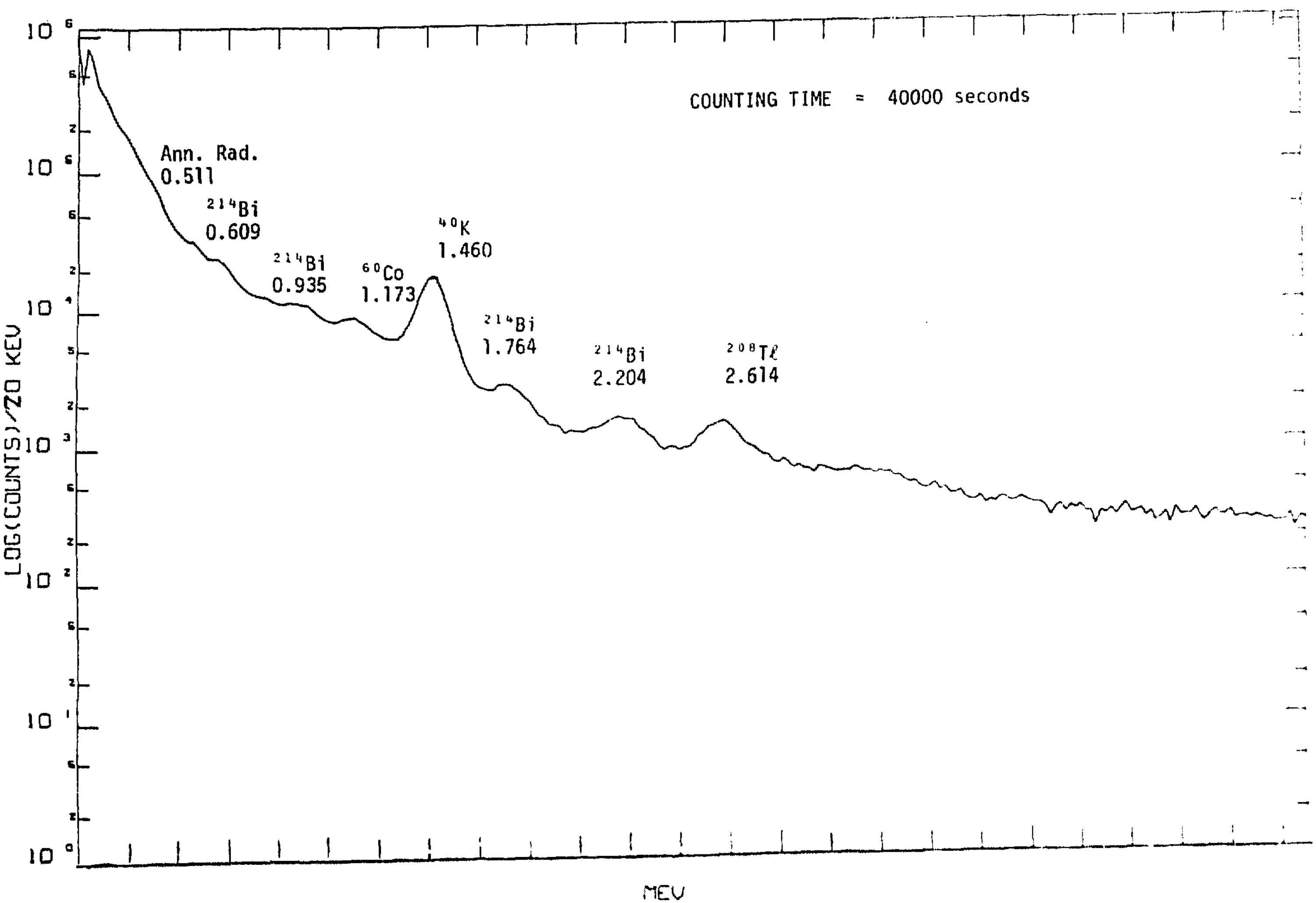

FIGURE I The Shadow Shield Background to 5.0 MeV sence of argon 41, the background count rate remained stable for prolonged periods: a sequence of 15 4000 second counts within the energy range 0.20 - 2.00 MeV yielded a mean count rate of 3019 c/m and standard deviation of 10.5 c/m (0.35%), the expected value of the latter being 6.7 c/m (0.22%). Practically, a morning count could suffice for the day's counting in the absence of a wind shift.

More prolonged observation of the background counting rate in two energy bands (1.36 - 1.56 MeV and 0.60 - 0.72 MeV) over a period of several years suggested the presence of secular variation, with a tendency for the background to rise in the winter months. In 1972, the increase was particularly marked, beginning in early September, peaking in early November at 30% higher than normal in the lower energy band and declining gradually over the next 4 months. This increase did not correlate with the barometric pressure and is largely unexplained. Subsequent variation has been small, and throughout 1974 the background has remained adequately stable, the counting rate in the caesium-137 energy band (weekly 20 minute counts) averaging 159 c/m with a standard deviation of 4.3 c/m (2.7%). Electronically, the stability of the counter proved adequate, and the standard deviation of the weekly counts of a caesium-137 source over a seven month period was ± 1.5%.

Because of the design of the counter, the marked increase in the background below ~0.3 MeV practically restricts assessment of radionuclides to those with γ emissions greater than this energy.

PROCEDURES

Before counting, all employees were required to shower and change into clean "whites", measures which experience has shown to be necessary if contamination of the counter is to be avoided, and if differentiation between internal and external contamination is to be made with any degree of assurance. In terms of radiation protection, such differentiation is pointless unless the apparent magnitude of the internal contamination approaches an investigation level. It is nevertheless of interest and where possible, it was attempted when the detected activity appeared to exceed ~ 20 nCi.

Some indication of the location of contamination is suggested by a marked difference between the prone and supine counting rates, but this proved an unreliable guide. The recently installed collimated array of sodium iodide crystals also provides an indication of the longitudinal location of the activity, but ultimately the time course of the detected radioactivity is the best determinant; most skin and hair contamination declines with a half life measured in days compared with weeks or months where the contamination is internal. Practically all the larger contaminations were examined on several occasions, mainly in order to ascertain the effective half lives of the contaminants.

RESULTS

In the period 18 September, 1972, to 25 March, 1975, when the counter was transferred to the new Low Background Building (560), 1351 whole-body scans were made on 895 employees, about 45 employees per month. This low average is much below the capacity of the system, and was due to extensive interference from argon-41 and electronic problems largely engendered by the lack of temperature control in the counting room. Because of the presence of caesium-137 derived from radioactive fallout in most people, it is convenient for the present purpose to define a normal body scan as one indicative of less than 10 nCi of caesium-137 and no other detectable radioactive contaminant in the body, using the calibration procedure described above. On this basis, about 55% of the total scans were classified as normal, and in the remaining 45%, the detected levels of internal contamination were for the most part trivial; fewer than 2% exceeding 100 nCi (Table V). In none of the cases were the Recommendations of the International Commission on Radiological Protection on the maximum permissible doses exceeded.

TABLE V: Maximum Body Burdens Detected in 895 Employees

Radionuclide	Employees with >100 nCi	Maximum Burden (μCi)	% MPBB*
^{137}Cs	9	0.65	~2
^{60}Co	1	0.12	~1
^{203}Hg	6	0.40	10
^{110m}Ag	0	0.08	<1
^{65}Zn	0	0.04	<0.1
^{95}Zr-^{95}Nb	0	0.03	<0.2
^{75}Se	1	29	32

The following radionuclides were identified:

Commonly	^{137}Cs, ^{60}Co
Infrequently	^{203}Hg, ^{134}Cs, ^{95}Zr-^{95}Nb, ^{46}Sc, ^{131}I, ^{160}Ru Annihilation Radiation
Rarely	^{51}Cr, ^{110m}Ag, ^{198}Au, ^{75}Se, ^{99}Mo-^{99}Tc, ^{65}Zn, ^{59}Fe, ^{140}Ba-^{140}La

Other activity at various energies was detected on many occasions at levels too low to permit identification at a reasonable level of confidence.

* Maximum permissible body burden

CAESIUM-137

The commonest contaminating radionuclide, caesium-137 was detected in the majority of the employees to the extent of a few nanocuries, but only about 6% had body burdens greater than 25 nCi and about 1% greater than 100 nCi. The distribution of the contamination is shown below in Table VI.

TABLE VI: Distribution of Caesium-137 in Employees

Body Burden ^{137}Cs (nCi)	No. Employees
10 - 24	10
25 - 49	30
50 - 99	13
100 - 249	6
250 - 499	2
500 - 999	1

The maximum burden found was approximately 0.65 μCi. This burden was acquired by a pipefitter during the early days of changing of the calandria of the NRU reactor a month or two before the whole-body scan.

In general, it proved difficult to conduct long-term follow up of employees who had acquired body burdens of sufficient magnitude to warrant study, because of repetitive contaminations, internal and external, but where possible such studies were made. The mean biological half life of caesium-137 in 9 employees whose body burdens appeared to show exponential decay was ~ 126 days, with a range of 86 to 150 days. Caesium-134 was also detected on numerous occasions in association with the larger caesium-137 contaminations, but the activity in all instances did not exceed a few percent of the latter.

COBALT-60

The second commonest contaminant, cobalt-60 was detected in about 30% of the counts in very small quantities, and only 29 (3.2%) of the 895 employees examined had burdens in excess of 10 nCi, as shown in the following Table.

TABLE VII: Distribution of Cobalt 60 in Employees

Body Burden ^{60}Co (nCi)	No. Employees
10 - 24	24
25 - 49	4
50 +	1

The time course of the largest body burden (~122 nCi) was followed for over 600 days, by which time further contamination had taken place. A linear regression fit to a logarithmic transform of the counter data within the period 105-586 days after the first count yielded an effective half life (whole-body) of 379 days, corresponding to a biological half life of about 473 days. The date of contamination was not known, although the shape of the retention curve on the semi-log plot suggested that it had been recent, with an indication of a component with a half life of very roughly 20 days. Cobalt-60 was not detected in urine samples (detection limit for a 1500 ml sample about 500 (d/m)/day* depending upon the background counting rate). Consistently, the counting rate in the prone position was about 15% higher than that in the supine position, but localization studies were not practicable at that time. The 473 day biological half life is consonant with other reports that a small amount of cobalt-60 is retained for very long periods (5). Available evidence suggested that in this case, the burden was acquired by inhalation.

MERCURY-203

Mercury-203 was detected on 45 occasions in 29 employees, but in amounts under 100 nCi in all but 6, with a maximum burden of about 0.40 μCi. These internal contaminations resulted from inhalation of mercury-203 on two separate occasions (July, 1973 and June, 1974) following the withdrawal from the NRX reactor of a ruptured or defective isotope capsule containing mercuric oxide.

A 27 year old employee became internally contaminated, probably in the second half of June, 1974. A urine specimen showed the presence of mercury-197 and-203 on 2 July, 1974. Subsequently, 9 whole-body counts were made over a 5 month period and the initial measurement suggested a body burden of ~0.23 μCi. On a semi-log plot, the retention curve appeared to contain 2 components with half lives of ~8 and ~27 days. Whether these represent a distributional change of mercury-203 within the body and/or counting errors is a moot point.

* (d/m)/day = (disintegrations/minute)/day

A linear regression fit to a logarithmic transform of the counter data within the period 33-144 days after the first count on 11 July 1974 (50-161 days after the most probable date of contamination) yielded an effective half life (whole-body) of 27.2 days (90% confidence limits of 24.8 and 28.5 days). This value is in good agreement with recent published data, but not with that cited in ICRP Publication 2 at 8.2 days.

Within the period 8 July to 10 September (14 - 78 days after the most probable date of contamination) the mercury 203 excreted in the urine accounted for approximately 22% of the total whole body loss within the same period of time, a value similar to that found by Brown et al (6) after an inhalation exposure. The activity in the urine ((d/m)/day* based upon creatinine correction of urine sample volumes) declined with an effective half life of about 26 days.

SILVER 110^m

Approximately 80 nCi of silver 110^m were detected in a [illegible]ar old employee. Enquiries suggested that this burden had been acquired by inhalation a week earlier during the rebrazing of a silver soldered plug freshly withdrawn from the reactor. Silver-110^m was not detected in an overnight specimen of urine. Subsequent examinations showed that after an initial rapid clearance, the effective half life over the next four months was about 42 days (biological half life about 50 days), after which the remaining quantity had become too small to measure accurately.

Few measurements of the effective half life of silver 110^m in man have been reported in the literature. Newton and Holmes (7) cited an inhalation case where the half life was shown to be about 43 days, a value similar to that reported for liver clearance. The silver-110^m appeared to be cleared from the lung very rapidly and was apparently excreted only in the faeces. Sill (8) reported 4 examples where the half life ranged between 8 and 69 days. Silver-110^m was detected in or on a second employee on one occasion, but in much smaller quantities.

SELENIUM-75

A 52 year old man was found to have a body burden of about 29 μCi of selenium-75, the result of an intravenous injection of 211 μCi of selenium 75 selenomethionine for medical purposes. Over a period of 100 to 235 days post injection, the effective (whole-body) half life approximated 80 days, corresponding to a biological half life of about 238 days.

OTHER CONTAMINANTS

The amounts detected in all cases were less than 100 nCi and mostly under 25 nCi.

COMMENTS

The expected finding that in a 30 month period of monitoring, the levels of internal contamination in (mainly) selected groups of em-

* (d/m)/day = (disintegrations/minute)/day

ployees adjudged to be at especial risk because of the nature of their work are for the most part negligible, attests the efficacy of the control measures for the containment of radioactivity at Chalk River. In no case did the level of contamination found exceed 10% of the maximum permissible body burden. It should also be noted that the frequency of internal contamination disclosed represents an upper limit because of the manifest difficulty of distinguishing between internal and external contamination. For the most part, complex spectra were uncommon and were usually indicative of external contamination with fresh fission products. Caesium-137 and cobalt-60 were the most commonly associated internal contaminants.

Broadly, the few larger contaminations were associated with a major non-routine operation, the changing of the calandria of the NRU reactor, or with known incidents such as the rupture of an isotope capsule containing mercuric oxide on two separate occasions, both incidents being initially signalled by the air monitors in Building 163. Routine operations were responsible for the bulk of the lesser contaminations. There remained the sporadic or unsuspected internal contamination, the result of faulty operational practice or unknown equipment failure, incidents which are unpredictable and which are brought to light only by routine monitoring.

The results of this survey would suggest that too many employees are being monitored at too frequent intervals. Guidance on the design of monitoring programs is contained in ICRP Publication 12 (9), which also notes the importance of the acquisition of data relating to the human metabolism of radio-elements (paragraph 107). An additional and important reason in my view for maintaining a monitoring program at a level higher than that prescribed by physical and biological considerations is the assurance afforded to those potentially exposed to contamination,who may be unacquainted with these parameters, that surveillance is adequate and significant exposures are not taking place. That such assurance is important is suggested by the ready acceptance of the present program despite the inconveniences caused by the unsuitable location of the counter. With the transfer of the shadow counter to a permanent location several miles away, the present monitoring program will be re-examined in detail.

No major procedural changes in the screening of employees are envisaged. The need to maintain Building 560 as free of contamination as possible dictates a continuance of the procedures outlined earlier, and greater control will be possible. Previously employees were required to shower at their worksite prior to counting, and it was not always possible to ensure that this had been done, with consequent uncertainty about the location of any detected contamination. At the present time, consideration is being given to the use of disposable paper clothing on grounds of economy.

The shadow counter is more than adequate for the purposes of radiation protection in terms of sensitivity for the commonly encountered contaminants with γ emissions at energies greater than approximately 0.3 MeV. For rapid screening of personnel in the event of a radiation emergency, the high sensitivity of the system could be a considerable asset, permitting very short counting times - a few minutes with the subject centrally placed beneath the crystals. Although unnecessary for purposes of radiation protection, a better assessment of the errors in the quantitation of internal contamination is desirable in the future, as well as detailed studies of the localization, distribution and fate of inhaled radionuclides.

Dimensionally, the clearance between the crystal and surface of the padded trolley at 30 cm would seem to be the minimum compatible with a high acceptance of screening by employees. In all, 4 employees were too obese to pass beneath the crystal, another 3 were too apprehensive to be counted, and a few others expressed unease.

All measurements of spectra were made manually using spectrum stripping methods where necessary to identify and quantify the contaminants. In future, analysis on only those spectra considered to be of interest or dosimetrically significant will be conducted, assisted by a computer program under development, and the rest assessed as being trivial in magnitude on the oscilloscope will not be analysed.

The competent assistance of D.F. Autayo is gratefully acknowledged.

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APPENDIX

The equipment includes:

Nuclear Data Series 2200 Pulse-height Analyzer with Data Reduce/Integrate and Magnetic Tape Data Reduce modules.

Keithley 246 High Voltage Supply.

Tektronix RM 503 Oscilloscope.

Model 33 Teletype.

Harshaw NaI (Tℓ) 29.2 cm x 10.2 cm Crystal Type 46MBS16/B and 7 5 cm Bicron Crystals, Model P14.

Ortec Single Channel Analyzer and Ratemeter.

Bodine Speed Reducer Motor 1/70 H.P.

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