AECL-5621

ATOMIC ENERGY OF CANADA LIMITED

L'ÉNERGIE ATOMIQUE DU CANADA LIMITÉE

MEASUREMENT OF TRANSURANIC ELEMENTS IN VIVO AT CRNL

by

J.R. JOHNSON

Presented at "The Workshop on the Measurement of Heavy Elements in Vivo", Battelle Seattle Research Centre, 24-25 June, 1976

Chalk River Nuclear Laboratories

Chalk River, Ontario

November 1976

ATOMIC ENERGY OF CANADA LIMITED

MEASUREMENT OF TRANSURANIC ELEMENTS IN VIVO AT CRNL*

by

J.R. JOHNSON

*Presented at "The Workshop on the Measurement of Heavy Elements in Vivo", Battelle Seattle Research Centre, 24-25 June, 1976.

Medical Research Branch
Chalk River Nuclear Laboratories
Chalk River, Ontario
November 1976

Mesure des éléments transuraniens in vivo à Laboratoires Nucléaires de Chalk River*

par

J.R. Johnson

*Rapport présenté au "Workshop on the Measurement of Heavy Elements in Vivo" Battelle Seattle Research Centre, 24-25 juin 1976.

Résumé

On décrit les détecteurs, l'électronique et le blindage employés dans les Laboratoires Nucléaires de Chalk River pour mesurer in vivo les isotopes radioactifs dont les seules émissions capables de se diriger vers l'extérieur du corps sont des photons à basse énergie. On décrit également les méthodes employées pour étalonner les détecteurs et les composants électroniques ainsi que les incertitudes liées à ces étalonnages pour les isotopes transuraniens ^{238}Pu, ^{239}Pu, ^{240}Pu et ^{241}Am.

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

Novembre 1976

AECL-5621

ATOMIC ENERGY OF CANADA LIMITED

MEASUREMENT OF TRANSURANIC ELEMENTS IN VIVO AT CRNL*

by

J.R. JOHNSON

*Presented at "The Workshop on the Measurement of Heavy Elements in Vivo", Battelle Seattle Research Centre, 24-25 June, 1976.

ABSTRACT

The detectors, electronics and shielding used at Chalk River Nuclear Laboratories to measure in vivo radioactive isotopes, whose only emissions capable of penetrating to the outside of the body are low energy photons, are described. Also described are the methods used to calibrate these detectors and electronic components, and the uncertainties associated with these calibrations, for the transuranic isotopes ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Am.

Medical Research Branch
Chalk River Nuclear Laboratories
Chalk River, Ontario
November 1976

AECL-5621

TABLE OF CONTENTS

MEASUREMENT OF TRANSURANIC ELEMENTS IN VIVO AT CRNL*

by

J.R. Johnson

1. INTRODUCTION

In vivo measurements at Chalk River Nuclear Laboratories (CRNL) are made in a new low background building situated about 5 miles from the laboratories. Prior to the completion of this building, whole body counting was done in the plant area itself[1], and the performance of the counters suffered from the effects of a variable and frequently high background, mainly caused by ^{41}Ar. When it was decided that experimental mixed oxide fuel bundles would be fabricated at CRNL, a chest counting facility with low stable background was required to complement area monitoring and bioassay sampling in evaluating possible inhalation contamination cases.

The site for the low background building was chosen for its low ambient gamma background and for its direction from CRNL in relation to wind direction probabilities estimated from records kept at the laboratory since the late forties. (A higher than normal background due to ^{41}Ar has occurred at this site only once in 14 months that have elapsed since monitoring began.) The building was constructed with materials tested for radioactivity, and whenever a reasonable choice existed, the material with the lowest radioactivity was selected.

The counting facilities in the low background building include a shadow shield counter, a 3 m x 5.5 m dolomite cement room, and a 2 m x 2 m iron room. The dolomite room has 0.5 m walls and ceiling and a 1 m floor but does not have a graded Z lining. The iron room has 20 cm of selected steel plate for walls, floor and ceiling and has lead and copper lining.

* *Text of a paper presented at "The Workshop on the Measurement of Heavy Elements in Vivo", held at Battelle Seattle Research Centre, June 24-25, 1976.*

Chest counting is done in the iron room with two 12.7 cm phoswich[2] detectors. Future plans include lining the dolomite room with lead and copper, and if the expected background for the phoswich detectors is achieved this room will become the area used for routine chest monitoring, and the iron room will be used for development work.

The isotopes of interest as potential inhalation hazards to persons working in the mixed oxide fuel fabrication plant area are mainly ^{238}Pu, ^{239}Pu, and ^{240}Pu, and with time, ^{241}Am, as it grows in from the decay of ^{241}Pu. The plutonium isotopes do not emit any radiation during decay capable of penetrating to the outside of the chest from the lungs in measurable quantities other than the L X-rays of uranium[3]. These X-rays are of low relative abundance and are highly attenuated as they pass through the chest wall. A measurement of a subject's chest for lung burdens of these isotopes at or below the maximum permissible lung burden* of 16 nCi essentially consists of measuring the subject's background in the L X-ray energy region and comparing the measured value to a predicted value. A calibration factor that depends on the isotopic ratios of the deposited radioactive material, the pattern of the deposition, and the attenuation of the L X-rays in the tissue between the deposited material and the detectors is then applied to the difference between the measured and predicted background to obtain an estimate of the lung burden for this subject.

The main experimental effort since moving into the new building has been toward reducing the background of the phoswich detectors, and finding statistical parameters that can be used to describe the background for uncontaminated subjects, which can then be used as aids in predicting the background of suspected contaminated subjects.

This report describes the supporting equipment used with phoswich detectors, the backgrounds obtained, and the calibration factors used at

* *The lung burden, if retained for a year, would result in a dose to the lung during the year of approximately 15 rem.*

CRNL. Included is a brief discussion of origins of the uncertainties in the estimated lung burdens.

2. DETECTORS AND ELECTRONIC COMPONENTS

The detectors used for chest monitoring are two 12.7 cm diameter phoswich detectors (see Figure 1). The NaI(Tℓ) layer is 0.1 cm thick and the CsI(Tℓ) is 5 cm thick. Pulses from these detectors are sorted according to their shape using constant fraction timing discriminators (CFT SCA) and a time-to-amplitude converter (TAC) (see Figure 2). Portions of the TAC output are selected using single channel analysers (SCA) which gate the inputs of the gated analog router (GAR) to the multichannel analyser (MCA).

The MCA can also be used to examine the spectrum of pulse shapes, with the object of obtaining the best resolution possible between the pulses originating only in the NaI(Tℓ) layer and pulses originating completely or in part in the CsI(Tℓ) layer. This maximum resolution is required if we are to obtain the maximum advantage of the phoswich detectors over simple thin NaI(Tℓ) detectors. The advantage is the much lower background obtainable with a phoswich detector which is achieved by discriminating against pulses from events shown schematically as 2 in Figure 1, that is, the Compton scattering events.

Adjustments that have been found to affect the timing resolution for a given detector-electronic system are high voltage setting, length of co-axial cable between the photomultiplier tube and the preamplifier, RC time constants, pole-zero adjustments on the main amplifier, d.c. adjustments on the main amplifiers, constant fraction settings on the CFT SCA's and the walk adjustments on the CFT SCA's. The best timing resolution obtained to date was with the start CFT SCA set at 40%, the stop CFT SCA at 50% and all other adjustments made to achieve maximum resolution. A timing spectrum with these settings is shown in Figure 3.

3. BACKGROUND MEASUREMENTS

The backgrounds obtained in the L X-ray energy region (13 to 25 keV) in the iron room for both detectors are summarized in Table 1.

TABLE 1

Background in Iron Room

Mean value of backgrounds in counts per minute for two phoswich detectors in the iron room in the energy region 13 to 25 keV

Background (No phantom)	3.8	cpm
Background with water-filled phantom	4.7	cpm
Background with normal subjects	8-12	cpm
Defined background for reference phantom	10	cpm

The increase in the background with the introduction of a water-filled phantom from 3.8 to 4.7 cpm is thought to be entirely due to the scattering into this energy region of higher energy photons originating outside the phantom and being scattered by the phantom material. The increase in backgrounds to between 8 and 12 cpm for normal uncontaminated subjects is due to the subjects' normal complements of ^{40}K and ^{137}Cs. The variation in the background between subjects is thought to result from the variations of these radioactive materials between subjects, and to a lesser extent, from the variation in scattering material with different body builds.

Enough background measurements have been taken of phantoms and of three individuals to show that for a given configuration of scattering material and for each individual the measured backgrounds are symmetrically distributed about their mean values, and that the sample standard deviations are, within the statistical precision so far obtained, identical to those one expects of a Poisson statistic, namely the square root of the mean background. It was felt at the outset that a study of this nature was required to be able to describe statistically the fluctuations in subject background. Otherwise no confidence could be given to the predicted value of the background of suspected contaminated subjects.

The sample standard deviation also sets the minimum value of the activity that can be measured with this combination of shielding, detectors and electronics. That is, if the true average background is known exactly, and the calibration factor (sensitivity) is also known exactly, then the minimum detectable activity at the 95% confidence level (2σ MDA) is

$$2\sigma \text{ MDA} = \frac{2\sigma}{S} \text{ nCi}^* \qquad (1)$$

where $\hat{\sigma}$ is the best estimation of the standard deviation in the background in cpm and S is the sensitivity in cpm/nCi. If $\hat{\sigma}$ has been shown by a statistical study to be

$$\hat{\sigma} \simeq \frac{1}{t} \sqrt{B.t} \qquad (2)$$

where B is the background in cpm and t is the counting time in minutes, then Eq. 1 becomes

$$2\sigma \text{ MDA} \simeq 2 \frac{\sqrt{B.t}}{S.t} \text{ nCi} \qquad (3)$$

Note that $\hat{\sigma}$ could have a larger value than that given by Eq. 2 but it cannot be smaller. The fact that Eq. 2 is valid for our system indicates that the true average background for each individual studied, and for each configuration of scattering material studied, is essentially constant.

4. CALIBRATION

Calibration of detectors for the measurement in vivo of plutonium is difficult. Two approaches that have been used are measurements on volunteer subjects that have inhaled known amounts of radioactive material that emit photons with energy similar to that of the uranium L X-rays, and measurements made of realistic phantoms with known amounts of activity in the phantom lungs.

* *SI Equivalent: nCi = 37 Bq*

Calibrations of the phoswich detectors at CRNL are based on measurements performed by the author while visiting the Atomic Energy Establishment, Winfrith (AEEW), as Attached Staff in 1973 and 1974. These measurements are described in reference 4 and calibration factors derived from these measurements are summarized in Table 2. The measurements were made on the Winfrith chest phantom[5], which is thought to be a realistic approximation to subjects with activity evenly distributed in the lungs and with a mean soft tissue thickness (MSTT) overlying the lungs of 2.1 cm.

TABLE 2

Reference Phantom

Sensitivity for L X-rays	=	1.7×10^{-3} Counts/photon
S nsitivity for 60 keV gammas	=	5.6×10^{-2} Counts/photon
Background in 13-25 keV range	=	10 cpm
Background in 40-75 keV range	=	30 cpm
Mean Soft Tissue Thickness	=	2.1 cm

The backgrounds defined in Table 2 are the approximate midpoints of values for uncontaminated subjects measured with two phoswich detectors in the iron room at CRNL.

The MSTT for subjects will vary considerably from the 2.1 cm value used for reference phantom, and this variation will have a large effect on the sensitivity of the detectors, particularly for the L X-ray sensitivity. The magnitude of this effect can be estimated by estimating the subject's mean soft tissue thickness and correcting the sensitivity for the different attenuation factors in this soft tissue. At present we are using a correlation function between MSTT and the ratio of weight (W) in kg to height (H) in cm as derived by Ramsden et al[6] to estimate the MSTT of subjects. This function is

$$MSTT = 15.3\ W/H - 3.55\ cm \qquad (4)$$

We have recently purchased an ultrasonic unit for performing tissue thickness measurements and once we gain confidence in our ability to interpret the results of measurements using this instrument, Eq. 4 will be replaced either by our own correlation function or by direct measurement of a subject's MSTT. Once the MSTT for a subject has been estimated the sensitivity can be corrected by using measured attenuation curves for the L X-rays and/or the 60 keV gamma in soft tissue. These correction factors are given in Figure 4.

The plutonium that will be used at CRNL is expected to have the isotopic content at the time of separation given in Table 3.

TABLE 3

Reference Plutonium at Time of Separation

Isotope	Specific Activity Ci/g	Relative Abundance by Weight	Relative Abundance by Activity	17 keV[7] Photons per α
Pu-238	17.4	0.2	0.438	0.112
Pu-239	0.0614	60.	0.448	0.046
Pu-240	0.227	33.	0.913	0.100
Pu-241	111.5	7.	98.2	0
*Am-241	3.24	0	0	0.348

*^{241}Am also emits a 60 keV γ with an abundance of 0.40 γ/α

In practice the isotopic content of the contamination will be estimated from radiochemical analysis of nose blows or fecal samples. The sensitivity of the two phoswich detectors for each of these isotopes, and the 2σ MDA can be calculated for the reference phantom using Table 2 and Eq. 3. The results of these calculations are presented in Table 4.

TABLE 4

Sensitivity and 2σ MDA for Reference Phantom

Isotope	Sensitivity, cpm/ηCi (MSTT = 2.1 cm)	2σ MDA, ηCi (2000 s)
Pu-238	0.42	2.6
Pu-239	0.17	6.4
Pu-240	0.37	3.0
Am-241	50.5*	0.038*

* *Calculated for the 60 keV γ and using a background of 30 cpm*

The 2σ MDA for reference plutonium in reference phantoms is shown in Figure 5 as a function of time since the plutonium was separated.

Calibrations are continuing. We have purchased a skeleton and are examining materials for their suitability for use as tissue for building a "secondary standard" phantom for tissue thickness and distribution studies. We hope to be able to take measurements in the near future on one of the USERDA sponsored phantoms being designed and built at Lawrence Livermore Laboratory to corroborate these studies.

5. BACKGROUND PREDICTION

As pointed out earlier, the ability to predict, with well-defined confidence limits, the background in the L X-ray energy range is of utmost importance when estimating the lung burden of suspected contaminated subjects. It is beyond the scope of this report to present all the results obtained at CRNL and at AEEW for deriving prediction functions for this background. Only the methods will be discussed. There are four methods, or combinations of methods, that have been used, and each has its advantages and disadvantages depending on the contamination and measurement history of the individual being evaluated for possible lung contamination. These methods are commented on separately below.

(a) Previous Measurements

If a subject can be measured before being exposed to the risk of an inhalation contamination, these measurements can be used to estimate the individual's mean background and the standard deviation in this background. This method requires that contamination by any other activity that could change the background in the L X-ray energy region be identified, and if the subject being evaluated has been contaminated with any of these activities, this method cannot be used directly.

(b) Matched Subjects

A method that does not have the requirement that the individual be measured prior to the suspected exposure is to measure the background of an individual whose body build is matched to that of the subject. If the subject is contaminated by other activities this method cannot be used unless a matched subject is found with the same contamination (by activity and distribution) as the subject being evaluated.

The two methods described above have the advantage of being direct measurements. The next two are indirect and require the development of correlation functions, which need many measurements of individuals covering a wide range of body builds.

(c) Background at Higher Energies

Useful correlations have been obtained, both here and at AEEW, between the background at higher energies and the background in the L X-ray energy region. The correlations appear to be valid for high energy gamma emitters that are evenly distributed throughout the body but introduce large errors if the primary photon energies of the interfering activities are below 200 keV, or the activity is concentrated in the chest region.

(d) Sum of Contributions

This method starts with the background obtained for a water-filled phantom and adds to it contributions from identified activity in the subject. Correlation functions involving body build must

be derived for each isotope that may be present in the subject if this method is to be useful. The assumption that the interfering activity has the same distribution in the subject as that used to develop the correlation functions is also required.

All individuals that are expected to be at risk to an inhalation exposure to plutonium in the fuel fabrication plant at CRNL will be measured before plutonium is introduced into the facility. These uncontaminated backgrounds will be used as the prediction values unless measurements at higher energies, either with the CsI(Tℓ) layer of the phoswich detectors or the shadow shield counter, indicate exposure to other activities has occurred. Because unmeasured persons will undoubtedly be in the plutonium facility from time to time, and the possibility of contamination with other activities than plutonium does exist, we are continuing to accumulate spectra of uncontaminated (with plutonium) subjects to be used as matched subjects and for the improvement of correlation functions.

6. UNCERTAINTY IN LUNG BURDEN ESTIMATES

There are two sources of uncertainty in the estimation of lung burdens of individuals. The first is the uncertainty in the predicted counting rate for uncontaminated subjects, which affects the precision of the estimation. The second is the uncertainty in the value used for the sensitivity of the detectors which affects the accuracy of the estimation.

(a) Precision

One method of quantifying the uncertainty in the lung burden estimate due to the uncertainty in the predicted background is to estimate the uncertainty in the difference (D) between the measured (M) and predicted (P) counts in a set time (t).

$$D = P - M \qquad (5)$$

The measured count is known exactly. If the estimation of P is based on n previous measurements of the subject or on n measurements of a matched subject then the uncertainty in predicted value is

$$\Delta P = \left[\hat{\sigma}^2 + \frac{\hat{\sigma}^2}{n}\right]^{\frac{1}{2}} \tag{6}$$

where $\hat{\sigma}$ is the sample standard deviation of the previous n measurements.

$$\hat{\sigma} = \left[\frac{1}{n-1} \sum_{i=1}^{n} (X_i - \bar{X})^2\right]^{\frac{1}{2}} \tag{7}$$

The X_i are the results of the individual measurements and $\bar{X}$ their average value. If $\hat{\sigma}$ has been shown to be approximately the square root of $\bar{X}$, then Eq. 6 becomes

$$\Delta P = [P + P/n]^{\frac{1}{2}} \tag{8}$$

P can also be estimated from a correlation function of the form

$$P = \sum_{i=1}^{m} a_i Y_i \tag{9}$$

where the a_i are the correlation parameters and the Y_i the estimated backgrounds at higher energies or amounts of other activities for the subject being measured. If the assumption is made that the a_i and the X_i are all independent, then the variance in the predicted mean value of P is[8]

$$\sigma_p^2 = \sum_{i=1}^{m} \left[\left(\frac{\partial P}{\partial a_i} \Delta a_i\right)^2 + \left(\frac{\partial P}{\partial y_i} \Delta Y_i\right)^2\right] \tag{10}$$

and the overall uncertainty in the value of P is

$$\Delta P = [P + \sigma_p^2]^{\frac{1}{2}} \tag{11}$$

In Eq. 11, P is the variance associated with underlying statistical distribution of P about its mean value, which is taken to be the minimum value as was done in Eq. 8. Δa_i are the uncertainties in the correlation parameters. ΔY_i are the fluctuations in the measured values of the background at higher energies or the measured activities and are taken to be their minimum values, $(Y_i)^{\frac{1}{2}}$.

(b) Accuracy

The lung burden (L) is calculated using the formula

$$L = D/(S.t) \qquad (12)$$

The uncertainty in the sensitivity (S) is difficult to assess accurately. It depends on the method of calibration and includes such things as the uncertainty in the original calibrated radioactivity, uncertainty in the attenuation in chest wall tissue, uncertainty in the effects of distribution of activity in the lungs, amount of activity in the ribs and tracheobronchial lymph nodes, etc. Work is continuing at CRNL to improve the value used for the sensitivity. The uncertainty in this value is not taken into account when the uncertainty in a lung burden is calculated. The values of the sensitivities used are those given in Table 2 suitably modified by the correction factors given in Figure 4.

While this approach may not be acceptable to some investigators it is essentially not different than that used for calculating the dose to other organs from internally deposited radioactivity. For example, the dose to the thyroid from radioiodine is calculated from fairly accurate measurements of the μCi burden and its effective half-life in the thyroid, and a defined value of 20 g for the mass of the thyroid of adult males[9]. The measured values[9] of thyroid mass for adult males is 17.5 ± 6.8 (1σ)g. It is difficult to estimate in vivo the true mass of the thyroid for the individual being evaluated and only rarely is the uncertainty in the mass included in a dose calculation.

(c) Overall Uncertainty

Accepting, as noted above, some uncertainty associated with the value used for S, the overall uncertainty in the lung burden is

$$\Delta L = \frac{\Delta D}{S.t} \qquad (13)$$

with ΔD depending on the method used to predict the value of P. Expressions for ΔD similar to Eq. 8 or 10 can be derived to suit the method of prediction used. The value of ΔL from Eq. 13 can be viewed as the approximate 68% confidence limits on the estimate of L if the usual assumptions regarding the normalcy of the distribution of L are made.

7. SUMMARY

The new facility at CRNL for measuring lung burdens in individuals suspected of having inhaled plutonium has been described. Measurements taken in this facility to date indicate that it will be possible to measure lung burdens of plutonium below the maximum permissible lung burden of 16 nCi. The limit of detectability is critically dependent on maintaining a low stable background. This limit also varies considerably with the physical habitus and state of contamination of the individual being measured.

Experiments are continuing at CRNL with the object of devising better methods for predicting an individual's background, and of obtaining better values for the sensitivity of the phoswich detectors for measuring transuranic elements in vivo.

FIGURE 1

Schematic of a phoswich detector and the shape of pulses from three different types of interactions of photons in the detector.

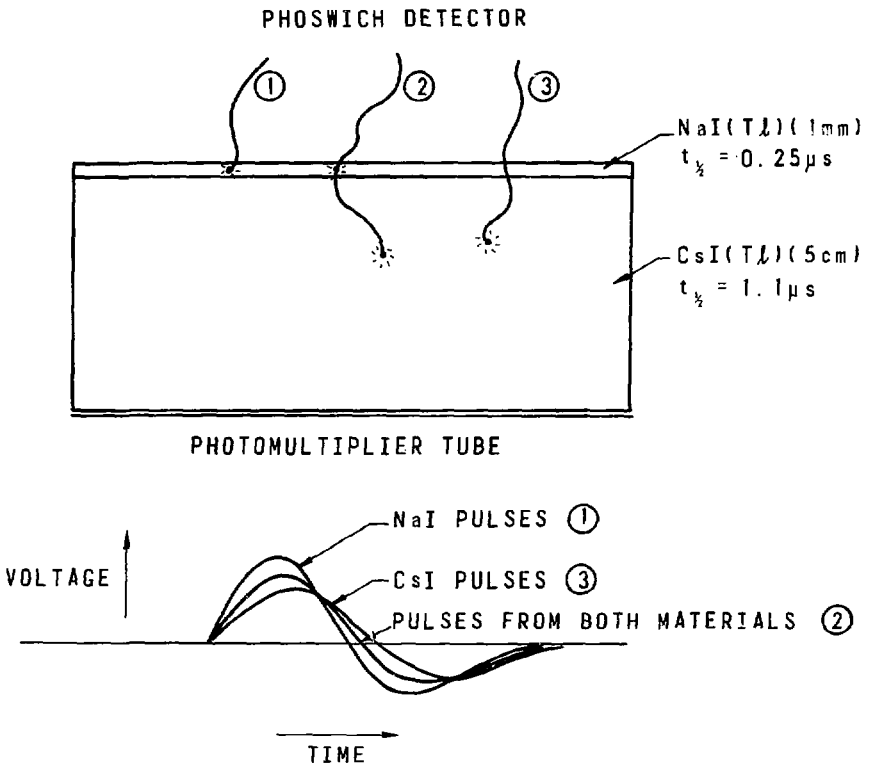


FIGURE 2

Block diagram of the electronics used for pulse shape discrimination with the phoswich detectors.

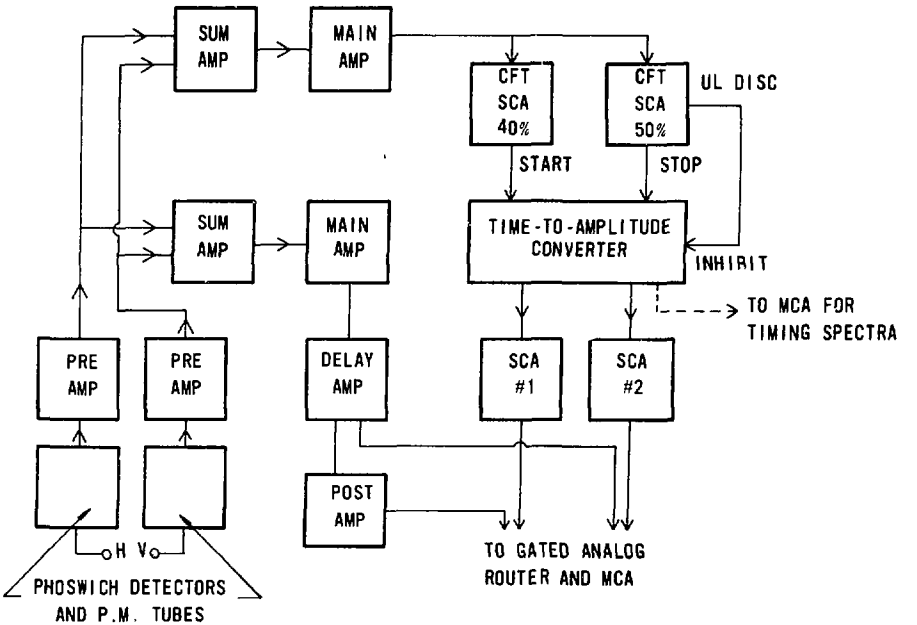


FIGURE 3

Timing spectrum obtained using the time-to-amplitude converter. This resolution was the best obtained with the system used. FWHM - full width at half maximum

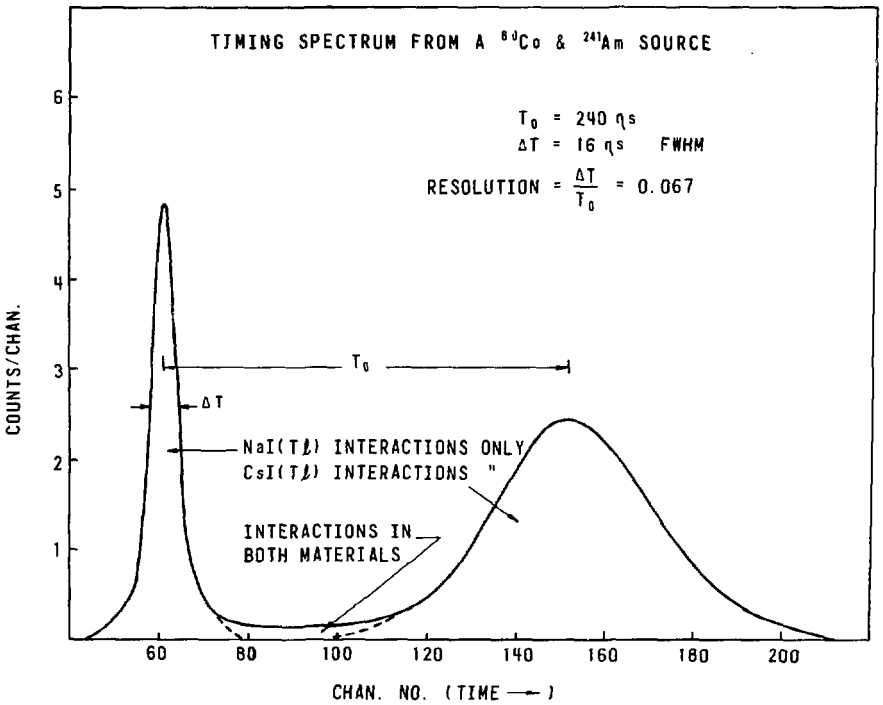


FIGURE 4

Factors to correct the sensitivities given in Table 7 for the effects of different body builds. (Sensitivities are divided by these factors.)

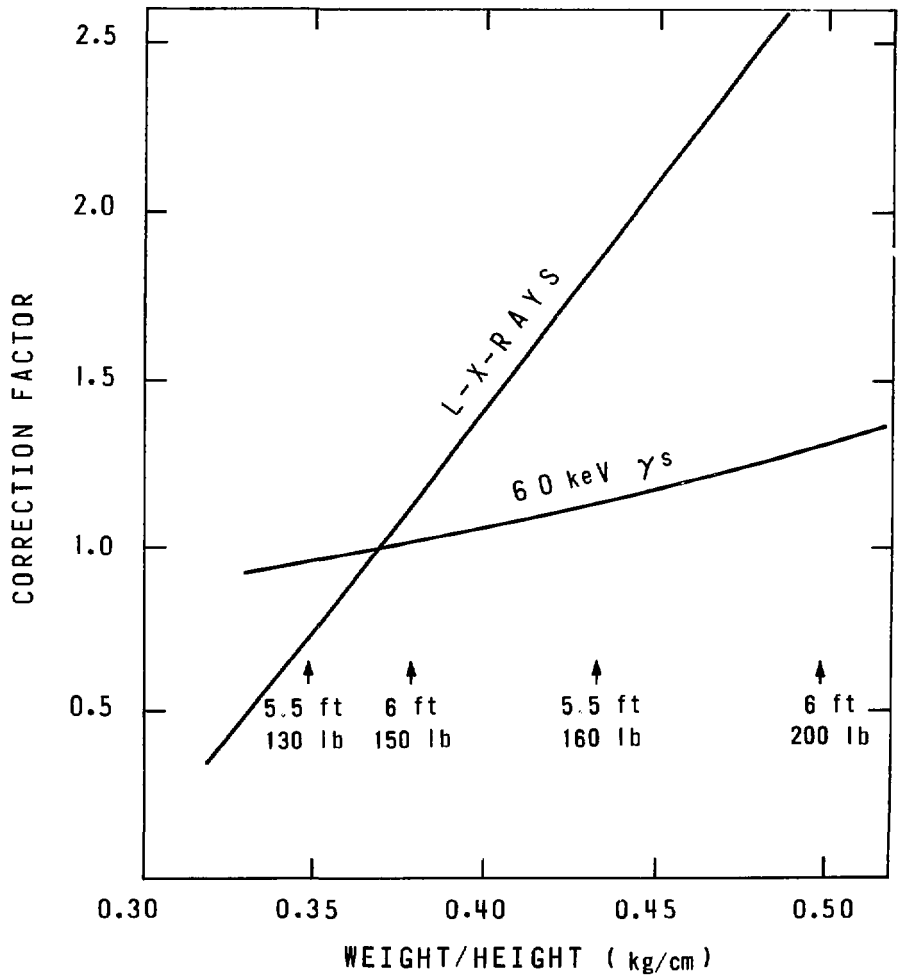
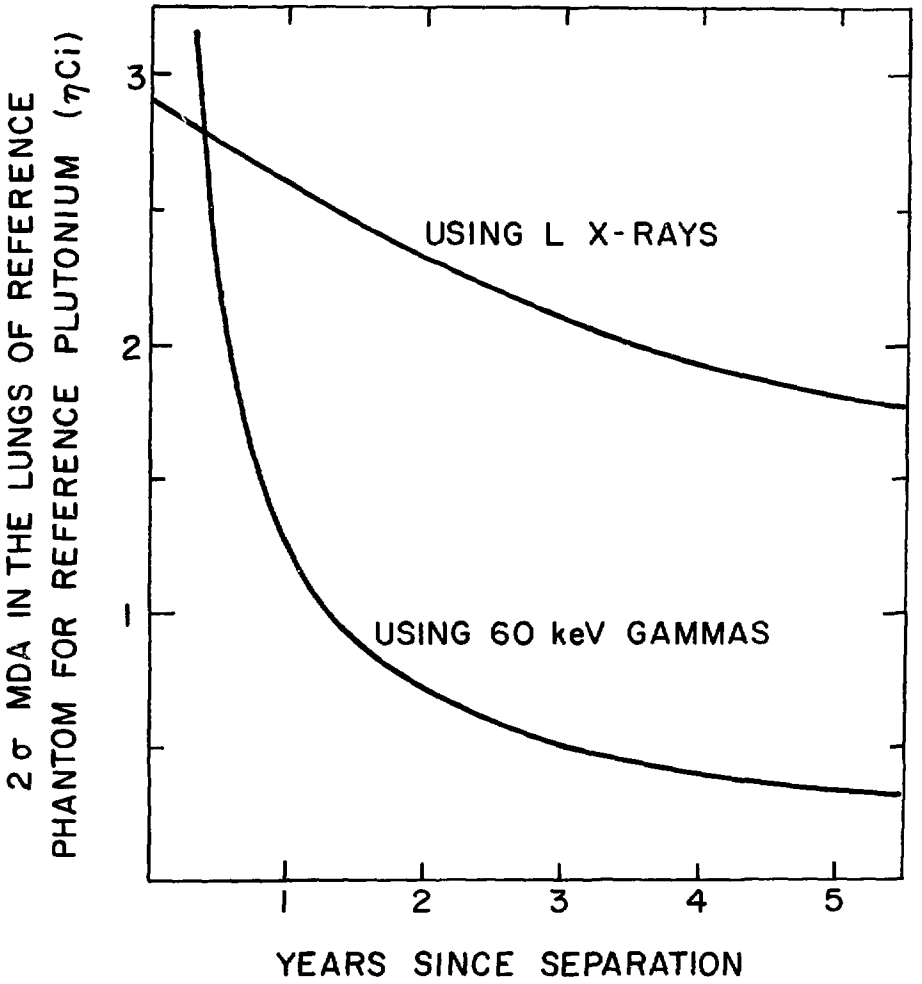


FIGURE 5

Minimum detectable activity for reference plutonium (defined in Table 3) in reference phantom (defined in Table 2) as a function of time since the plutonium was separated from other activities.



REFERENCES

1. Evans, D.W.S., Internal Radioactive Contamination in Selected Groups of CRNL employees. Atomic Energy of Canada Limited Report AECL-5255 (1975).

2. Laurer, G.R., and M. Eisenbud. In Vivo Measurements of Nuclides Emitting Soft Penetrating Radiations. Diagnosis and Treatment of Deposited Radionuclides p. 189-207. Exerpta Medica Foundation (1968).

3. Lederer, G.M., J.M. Hollander, and I. Perlman. Table of Isotopes John Wiley & Sons (1967).

4. Johnson, J.R. and D. Ramsden. Investigations into a Phoswich Detector for Plutonium In Vivo Monitoring. AEEW-M1304 (1975).

5. Speight, R.G., C.O. Peabody and D. Ramsden. An Improved Chest Phantom for Studies of Plutonium and Americium in Human Lungs. Assessment of Radioactivity in Man; Vol. 1 p. 115-130. IAEA (1964). STI/PUB/84.

6. Ramsden, D., C.O. Peabody and R.G. Speight. The Use of Ultrasonics to Investigate Soft Tissue Thicknesses on the Human Chest. AEEW-R 493 (1967).

7. Toohey, R.E., Argonne National Lab. Private Communications.

8. Freund, John E. Mathematical Statistics. Prentice-Hall Inc. (1962).

9. Report of the Task Group on Reference Man. ICRP Publication 23. Pergamon Press (1975).

The International Standard Serial Number

ISSN 0067–0367

has been assigned to this series of reports.

To identify individual documents in the series we have assigned an AECL–number.

Please refer to the AECL–number when requesting additional copies of this document from

Scientific Document Distribution Office
Atomic Energy of Canada Limited
Chalk River, Ontario, Canada
K0J 1J0

Price - $3.00 per copy

1969-76