CIRCULATION COPY
SUBJECT TO RECALL
IN TWO WEEKS

UCRL-83946
PREPRINT

CONSISTENT X-RAY COUNTING EFFICIENCIES FOR PLUTONIUM IN LUNGS, DERIVED BY INDEPENDENT METHODS

D. Newton
Atomic Energy Research Establishment
Harwell, Oxon, England

G. W. Campbell
A. L. Anderson
J. C. Fisher
Lawrence Livermore Laboratory
Livermore, California 94550

This paper was prepared for submittal to
Health Physics Journal

February 4, 1980

Lawrence Livermore Laboratory

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

LOAN COPY

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

CONSISTENT X-RAY COUNTING EFFICIENCIES FOR PLUTONIUM IN LUNGS, DERIVED BY INDEPENDENT METHODS

D. Newton

Environmental and Medical Sciences Division, Atomic Energy Research Establishment, Harwell, Oxon, England.

and

G.W. Campbell, A.L. Anderson and J.C. Fisher

Hazards Control Department, Lawrence Livermore Laboratory, California 94550.

INTRODUCTION

Calibration of X-ray counters for the assessment of plutonium in lungs is notoriously difficult. It has generally been attempted with phantoms containing plutonium-loaded 'lungs', less commonly by depositing independently-known quantities of some short-lived, low-energy X-ray emitter in the lungs of volunteers(Ne71; Ra72; Sh76). When X-ray detection efficiencies derived by the second of these methods were compared with those recorded for the same nuclide in a series of phantoms, large discrepancies were found(Ne78a). These may be attributed at least partly to the common use, in such phantoms, of materials whose X-ray attenuation properties are quite different from those of the tissues they are intended to represent(Ne78b).

The nuclide most frequently administered to volunteers for calibration purposes has been ^{103}Pd which, decaying by electron capture with a half-life of 17 days, provides abundant 20-23 keV K X rays. If the energies and relative intensities of these X rays had coincided with those

of the emissions from plutonium (uranium L X rays, range 13-20 keV), it would have been simple to deduce calibration factors for plutonium from the recorded count rates per unit activity of ^{103}Pd known to be present in the lungs of volunteers: the number R, defined as the ratio

$$\frac{E_{Pd}}{E_{Pu}} = \frac{\text{counts per K X ray emitted by } ^{103}\text{Pd in lungs}}{\text{counts per L X ray emitted by plutonium in lungs}}$$

would have been unity. The energies are not the same, however, and because of differential attenuation, $R > 1$. R depends strongly on the subject's chest wall thickness and on the distribution of activity in the lungs, and, to a lesser extent, on other variables such as the proportion of adipose tissue in the chest wall. Various methods(Ru69; Co73; To76; Ne78c) have been proposed to derive calibration factors for plutonium from studies with inhaled ^{103}Pd, all of them in effect providing estimates of R.

A phantom(Gr78) has been produced at Livermore which, unlike most others, is composed of materials closely tissue-equivalent in their linear attenuation of low-energy photons, and which is realistic in its anatomical structure. This development invited a comparison of the X-ray detection efficiencies for ^{103}Pd and plutonium, present in the phantom's lungs, with efficiencies previously recorded for ^{103}Pd deposited in the lungs of volunteers, or derived for plutonium from those observations. It has also enabled us to compare values of R, predicted by calculation, with those deduced from the relative detection efficiencies for ^{103}Pd and plutonium measured in the phantom; in this way we have been able to investigate the validity of such predictions more rigorously than was previously(Ne78c) possible.

contd

METHODS AND MEASUREMENTS

The phantom

The phantom[Gr78] is intended to represent a male thorax, between the 5th cervical and 4th lumbar vertebrae. Its relevant organs and 'tissues' are composed of materials which were shown to be approximately tissue-equivalent in their linear attenuation of uranium L X rays. The principal internal structures (lungs, mediastinum, liver and surrounding tissues) are contained within a shell of muscle-equivalent material, representing the chest wall. The body shell and internal components were cast in moulds prepared from the thorax and organs of a cadaver, whose sternum, rib cage and vertebrae were incorporated in the phantom's chest wall. In this, its basic, form the phantom's mean anterior chest wall thickness is 19 mm, in those regions of the upper thorax commonly viewed by detectors in monitoring for burdens of plutonium. Close-fitting overlayers of various thicknesses (Table 1), one set made of an approximately muscle-equivalent material, the other consisting of equal weights of substitutes for muscle and adipose tissue, can be added to the basic shell, to simulate chest walls of four greater thicknesses, up to a total thickness of 43 mm. In this work, three sets of lungs uniformly labelled with known amounts of activity were employed. One set contained ^{103}Pd, another ^{238}Pu, and the third ^{239}Pu with minor amounts of other plutonium isotopes and $\sim$ 18 ppm ^{241}Am by weight.

Determination of detection efficiency for the phantom

The detector was a phoswich, comprising a 1.5mm thickness of NaI(Tl) as the X-ray counter and 50mm of CsI(Tl) as the anti-coincidence counter. Both crystals were of 200mm diameter; the effective diameter, investigated by scanning the beryllium radiation window with a collimated X-ray source,

contd

was 190mm. The detector was housed inside one of the 100mm-thick, cadmium-lined lead shields used for body radioactivity measurements at A.E.R.E. Harwell.

Photon-energy spectra (range 9-120 keV) were recorded with this detector suspended in three positions over the phantom, which was placed supine on a horizontal bed:

(1) In a central position over the upper thorax, inclined so that the window was approximately parallel to the surface of the chest, i.e. viewing a region of some 200mm diameter centred on the mid point of the sternum. At its closest, the window was ~ 10mm from the surface of the phantom; this separation was maintained irrespective of the overall thickness of the phantom, which varied according to the overlayer used, by raising or lowering the detector. This arrangement will be referred to as Geometry 1.

(2) Displaced laterally by one detector-diameter from this central position, (a) to the left, and (b) to the right, without change in orientation. The sum of spectra recorded in positions (a) and (b) indicated the response to be expected for Geometry 2, i.e. for two 200mm-diameter detectors, one viewing each lung.

Measurements with the phantom in its basic form, and fitted with each of the eight overlayers (Table 1), were made under the following conditions:

(i) ^{103}Pd-labelled lungs, both geometries

(ii) ^{238}Pu-labelled lungs, both geometries

(iii) ^{239}Pu-labelled lungs, Geometry 1 only .

It was necessary to remove the phantom from the bed between (and, in some instances, during) certain of the five series of measurements, and it may not always have been repositioned identically with respect to the detector.

contd

On occasion, when we attempted to replicate particular observations after disturbing the phantom, differences in detection efficiency as large as 6% were found, indicating some sensitivity to small changes in the position and orientation of the counter over the chest.

Determination of detection efficiencies for ^{103}Pd in the lungs of volunteers

For comparison with the recorded detection efficiencies E_{Pd} for ^{103}Pd in the phantom, we shall employ data[Ne78a] derived from spectra of X rays emitted by subjects containing independently-known alveolar deposits of ^{103}Pd, measured with a similar detector in the same geometries. These subjects were volunteers who had inhaled a monodisperse aerosol of 5-μm polystyrene particles incorporating ^{103}Pd and also ^{51}Cr. ^{51}Cr emits gamma rays of energy 323 keV, and established methods of body radioactivity measurement were used to determine the residual lung content of this nuclide after early rapid clearance of the respiratory tract was complete. With this value, and the known relative concentrations of ^{103}Pd and ^{51}Cr in the aerosol, the subject's ^{103}Pd content could be deduced, independently of X-ray counting.

Prediction of the relative detection efficiency R for ^{103}Pd and plutonium in lungs

The method of predicting R makes use of a mathematical representation of the thorax and its relevant internal structures in relation to the detector[Ne78c]. One proceeds, in effect, by performing a double integration of detection efficiency over the counter window area and the lung volume, for each principal component of the X-ray spectra of plutonium and ^{103}Pd, taking into account both geometrical factors and energy-dependent attenuation

contd

effects in the lungs, in the soft tissues of the chest wall, and in the bones of the thoracic cage.

RESULTS

Detection efficiencies, etc., will be quoted for the various chest wall thicknesses represented by the combination of each overlayer and the basic phantom, and by the basic phantom alone. In each case, the value of chest wall thickness is a mean, derived from measurements with calipers in a total of 94 positions over areas of 120 mm diameter, one on each side of the sternum, tangential to the sternum and clavicle. These represent approximately the regions viewed by 120-mm-diameter detectors in standard procedures for assessment of plutonium in lungs adopted by many laboratories, including Lawrence Livermore. These areas fall within, or overlap, the larger regions covered by the 190-mm (effective) diameter phoswich used in the present investigations, and over these larger areas the mean chest wall thickness could be different. However, the mean thickness, t, over the smaller areas, has been retained for the present purposes, for two reasons. The first is that the X-ray flux emerging from the corresponding 120-mm-diameter areas on humans appeared to contribute more to the response of Geometry 2 than did emissions from outside those areas(Ne78a). The second reason is that it will permit easier comparisons between measured values of E_{Pd} for the phantom and those derived following inhalation of ^{103}Pd by volunteers, whose chest wall thicknesses, over these restricted areas, were measured by several laboratories.

X-ray detection efficiencies for ^{103}Pd in lungs

For Geometry 2, values of E_{Pd} for the phantom, with and without its various overlayers, are shown as the upper pair of curves in Fig. 1, plotted against t. The observed efficiencies for three subjects who had inhaled

contd

^{103}Pd (Ne78a) are superimposed on this plot, after adjustment to allow for a 7 per cent difference in the intrinsic efficiencies of the detectors used in the two investigations. For Geometry 1 (data not given) the corresponding curves were similar in form to those shown in Fig. 1, and the observed values of E_{Pd} in vivo were in equally good agreement with the curves; all values of E_{Pd} were of course lower than for Geometry 2, by about 40 per cent on average.

X-ray detection efficiencies for ^{238}Pu and ^{239}Pu in lungs

The photon energy spectra obtained with several µCi ^{238}Pu in the phantom's lungs showed, in addition to the prominent X-ray peak at about 17 keV, a peak at 44 keV which was attributed to the low-abundance (0.038 per cent) gamma ray of this energy. Similarly, spectra recorded with the ^{239}Pu-loaded lungs in the phantom, once the estimated contributions from the known ^{241}Am impurity had been subtracted, contained a visible contribution from 52-keV gamma rays, as well as evidence of weaker radiation of higher energy. Scatter from these radiations led to a continuum of counts in the 10-33 keV region where the signals from genuine L X rays occurred, and for large values of t it apparently added 30% or more to the X-ray contribution. For our present purposes, in comparing the detection efficiencies for plutonium in the phantom with those derived from observed values of E_{Pd} in volunteers, the X-ray contribution E_{Pu} alone is required. This was calculated by taking the total area under the observed L X-ray peak and subtracting the counts in the estimated continuum.

For Geometry 2, with ^{238}Pu-loaded lungs, values of E_{Pu} are included in Fig. 1. Values of E_{Pu} which are considered appropriate to the three subjects who had inhaled ^{103}Pd are also plotted; these were derived in each case by applying the calculated value of $R^{(Ne78c)}$ to the measured value of

contd

E_{Pd} for the subject. For Geometry 1, values of E_{Pu} (investigated both with ^{238}Pu - and ^{239}Pu-loaded lungs in the phantom) and estimates of E_{Pu} from E_{Pd} in vivo showed agreement comparable with that seen in Fig. 1.

Comparison of predicted and observed values of R for the phantom

The data of Fig. 1 imply essential consistency between the calculated value of R which is considered appropriate to each of the three experimental subjects, and the actual value of R for a phantom with the same t. In calculating the three values of $R^{(Ne78c)}$ we assumed that, in all cases, 22% of the thickness of soft tissue in the chest wall was composed of adipose tissue; this was a published average following investigations at autopsy[Do73], the actual percentages for our subjects being unknown. This assumption was not critical in securing the agreement; calculations[Ne78c] suggested that, compared with its value if muscle alone were present, R would be only marginally affected by the presence of adipose tissue, except when existing in larger than average proportions in subjects with large t. It is of interest to confirm this prediction, and to investigate the validity of our methods of calculating R when various, known thicknesses of adipose tissue are present.

Estimates of R are given in Table 1, for the basic phantom and for the phantom with each of the eight overlayers. Three sets of values were obtained from the ratios E_{Pd}/E_{Pu}:

(i) with E_{Pd} and E_{Pu} determined respectively from measurements with the ^{103}Pd - and ^{238}Pu - labelled lungs in the phantom, for which Geometry 1 was used,

(ii) by the same procedure, but with Geometry 2 used, and

(iii) by comparison of E_{Pd} and E_{Pu} determined with ^{239}Pu - loaded lungs, for Geometry 1.

contd

The three estimates are not always closely similar, the widest variations occurring when the attenuation is greatest, i.e. for large values of t. In these situations, scatter from radiations of higher energy in the spectra of ^{238}Pu and ^{239}Pu made important contributions in the L X-ray region; errors introduced in estimating these contributions would affect assessment of the genuine X-ray detection efficiency, E_{Pu}, and consequently the implied value of R. Previously-mentioned errors in re-positioning the phantom between measurements could have contributed to systematic differences between the three sets of estimates, and in some cases to random errors in individual values. Genuine differences in R, according to the region of the chest viewed by a detector, are of course to be expected, although they would not explain the differences between some of the pairs of estimates by methods (i) and (iii).

Also given in Table 1 are values of R for the phantom, derived by calculation. Initial estimates were obtained from Fig. 5 of ref. Ne78c, showing R as a function of t, calculated on the assumption that the chest wall consisted entirely of muscle. Relationships similar to those of Fig. 6 in Ne78c were derived, allowing these estimates of R to be reduced when necessary, according to the known proportion of adipose 'tissue' in the phantom's chest wall. As previously(Ne78c), R was then increased by 6% to allow for assumed filtration in the bones of the thorax. The calculated values are similar to the means of the observed values, obtained by methods (i), (ii) and (iii) above. The closeness of this agreement must in some cases be fortuitous, in view of the scatter often present in the three values from which each mean is calculated. However, it appears that our method of calculating R is not subject to major systematic error, provided that, as in this case, the assumption of a uniform distribution

contd

of activity in the lungs is valid.

CONCLUDING REMARKS

The encouraging similarity (Fig. 1), between values of E_{Pd} indicated by the phantom and those recorded for ^{103}Pd *in vivo*, suggests that the pulmonary distribution of activity in the experimental subjects was similar to that in the phantom, i.e. that it was essentially uniform. The further consistency between values of E_{Pu} for the phantom and those derived from data on E_{Pd} *in vivo*, together with the overall similarity between predicted and observed values of R (Table 1), has improved our confidence in these two quite different approaches to the problem of calibration. However, it is not to be expected, with either method, that one will always be able to deduce approximately the value of E_{Pu} correct for a particular contaminated subject, merely by reference to curves like those in Fig. 1, relating detection efficiency with chest wall thickness. Other variables, some of them indeterminate, must affect E_{Pu} markedly. The most important is probably the pattern of deposition within the lungs, which in turn must depend on the subject's respiratory pattern during intake, and on the aerodynamic characteristics of the particles.

D. NEWTON

Environmental and Medical Sciences Division
Atomic Energy Research Establishment
Harwell, Didcot, Oxon, U.K.

G.W. CAMPBELL
A.L. ANDERSON
J.C. FISHER

Hazards Control Department
Lawrence Livermore Laboratory
California 94550, U.S.A.

REFERENCES

Co73 Cohen N., Guilmette R.A. and Wrenn M.E., 1973, New York University Medical Center Progress Report COO-3382-13.

Do73 Dolguirev E., Kaidanovsky G.W., Porozov N.V. and Shamov V.P., 1973, "The methods of absolute calibration of equipment for measurements of ^{210}Pb, ^{239}Pu and ^{241}Am in human body," Proceedings of the 3rd International Congress IRPA, Washington DC, CONF 730907, part 2, p.969.

Gr78 Griffith R.V., Dean P.N., Anderson A.L. and Fisher J.C., 1978, "Fabrication of a tissue-equivalent torso phantom for intercalibration of in-vivo transuranic nuclide counting facilities," paper presented at the IAEA Symposium on Advances in Radiation Protection Monitoring, Stockholm, June 1978.

Ne71 Newton D., Rundo J. and Taylor B.T., 1971, "Progress in instrumentation and calibration techniques for the assessment of lung burdens of plutonium-239," in: Radiation Protection Problems Relating to Transuranium Elements, EUR 4612 d-f-e, pp 469-482 (Luxembourg: CID).

Ne78a Newton D., Fry F.A., Taylor B.T., Eagle M.C. and Sharma R.C., 1978, "Interlaboratory Comparison of Techniques for Measuring Lung Burdens of Low-Energy Photon Emitters," Health Phys. 35, 751-771.

Ne78b Newton D. and White D.R., 1978, "Attenuation of 13-20 keV Photons in Tissue Substitutes and their Validity for Calibration Purposes in the Assessment of Plutonium in Lungs," Health Phys. 35, 699-703.

Ne78c Newton D., Taylor B.T. and Anderson A.L., 1978, "X-Ray Counting Efficiencies for Plutonium in Lungs, derived from Studies with Inhaled Palladium-103," Health Phys. 34, 573-585.

Ra72 Ramsden D. and Waite D.A., 1972, "Inhalation of Insoluble Iron Oxide Particles in the Sub-Micron Range," in: Assessment of Radioactive Contamination in Man, pp 65-81 (Vienna: IAEA).

Ru69 Rundo J., Rudran K. and Taylor B.T., 1969, "Effective Tissue Thicknesses for External Counting of Low-Energy Emitters in Lung," Health Phys. 17, 155-157.

Sh76 Sharma R.C., Somasundaram S., Kotrappa P., Haridasan T.K., Surendran T., Kapur D.M. and Bhanti D.P., 1976, "Assessment of Chest Burdens of Plutonium," in: Diagnosis and Treatment of Incorporated Radionuclides, pp 177-201 (Vienna: IAEA).

To76 Toohey R.E., 1976 "The Use of ^{103}Pd as a Calibration Source for the Determination of ^{239}Pu in vivo," Battelle Pacific Northwest Laboratory Report, BNWL-2088, pp 53-62.

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately-owned rights.

Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Department of Energy to the exclusion of others that may be suitable.

Fig. 1 caption

Detection efficiencies for ^{103}Pd and ^{238}Pu in lungs, measured with Geometry 2. The data are based on recorded counts in the full X-ray peaks, after subtraction of estimated scatter contributions from 44-keV gamma rays in the case of ^{238}Pu. The loss of genuine X-ray signals in the pulse shape discrimination circuits was 10%.

Curves A (●) : phantom with muscle-equivalent chest wall

Curves B (⊙) : basic phantom (19-mm muscle-equivalent chest wall) with overlayers representing a 50/50 (weight %) muscle/adipose mixture; the numbers 11, 18, 23 and 28 indicate the thickness of adipose-equivalent material as a percentage of the total chest wall thickness.

Points X : measured X-ray detection efficiencies for ^{103}Pd in volunteers (upper plot) and derived efficiencies for plutonium in these volunteers (lower plot).

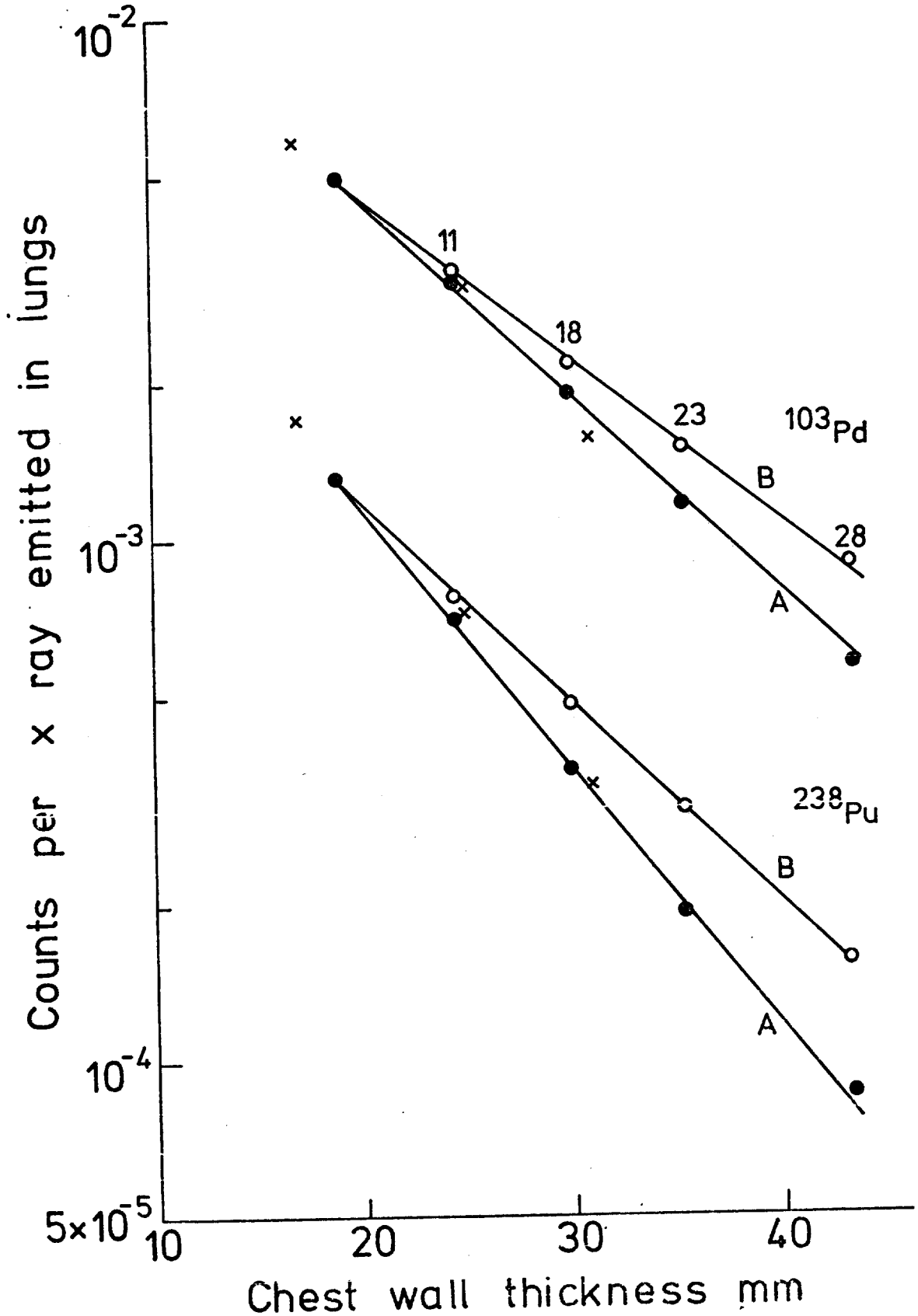
10^{-2}
10^{-3}
10^{-4}
5×10^{-5}
Counts per x ray emitted in lungs
10
20
30
40
Chest wall thickness mm
11
18
23
28
^{103}Pd
B
A
^{238}Pu
B
A

TABLE 1

ESTIMATES OF THE RATIO 'R' FOR THE PHANTOM*

OVERLAYER THICKNESS** (mm)	TOTAL CHEST WALL THICKNESS t (mm)	PERCENTAGE THICKNESS ADIPOSE/TOTAL	EXPERIMENTAL VALUES OF R†				CALCULATED R‡
			(i)	(ii)	(iii)	MEAN	
0	19.0	0	3.9	3.8	4.0	3.9	3.9
5.5 M	24.5	0	4.8	4.6	4.5	4.6	4.5
11.0 M	30.0	0	5.8	5.3	5.5	5.5	5.2
16.4 M	35.4	0	6.2	6.1	5.7	6.0	5.9
24.4 M	43.4	0	8.2	6.6	5.8	6.9	7.1
5.5 MA	24.5	11	4.4	4.2	4.5	4.4	4.3
11.0 MA	30.0	18	5.0	4.5	5.0	4.8	4.8
16.4 MA	35.4	23	5.7	4.9	4.9	5.2	5.3
24.4 MA	43.4	28	6.6	5.8	5.7	6.0	6.1

* $R = \frac{E_{Pd}}{E_{Pu}} = \frac{\text{counts per K X ray emitted by } ^{103}\text{Pd in phantom}}{\text{X-ray counts per L X ray emitted by plutonium in phantom}}$

** M = muscle-equivalent material, s.g. = 1.09; MA = material containing equal weights of muscle- and adipose-tissue substitutes, s.g. = 1.07

† (i) Geometry 1, E_{Pu} from measurements with ^{238}Pu-loaded lungs.
(ii) Geometry 2, E_{Pu} from measurements with ^{238}Pu-loaded lungs.
(iii) Geometry 1, E_{Pu} from measurements with ^{239}Pu-loaded lungs.

‡ see text and ref. Ne78c.