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INTERLABORATORY COMPARISON OF TECHNIQUES FOR MEASURING LUNG BURDENS OF LOW-ENERGY PHOTON-EMITTERS*

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Abstract—An interlaboratory exercise has been conducted to assess techniques of detection and calibration in the direct measurement of lung contamination with plutonium and other nuclides emitting only low-energy X-rays. Three volunteers, of small, intermediate and large physique, inhaled an aerosol incorporating ^{103}Pd, a 20-keV X-ray emitter, and visited 13 other laboratories in the U.K., Europe and North America. Participants in the exercise were asked to estimate each subject's lung content, using their procedures for assessing burdens of plutonium, and their estimates were compared with values derived independently from measurements of ^{51}Cr, also incorporated in the inhaled particles, by gamma-ray spectrometry. Laboratories' calibration procedures were in most cases based on elaborate thorax phantoms, and these generally led to underestimates of the subjects' contents, in some instances by a factor of three or more; only one such laboratory produced estimates in satisfactory agreement with the independently known values. The "phoswich" detectors, employed by most participants, appeared to be more sensitive than gas counters. If a standard configuration were required, offering the highest sensitivity in most situations, the choice would be a pair of 12-cm diameter phoswich detectors viewing the left and right anterior surfaces of the upper thorax. No improvement in sensitivity would result from increasing the size, although larger units may offer other advantages.

INTRODUCTION

TWO FUNDAMENTAL difficulties complicate the assessment of ^{239}Pu in lungs by external counting of low-energy X-rays (Table 1). In both cases they result from the severe attenuation which these radiations undergo within the lungs and in their passage through the chest wall: at 17 keV, for example, the half-value thickness in muscle is only ~ 6 mm. One of the ensuing problems is in the provision of equipment sufficiently sensitive to detect the weak flux of X-rays escaping from the body, even when more than the maximum permissible lung burden (16 nCi) is present. The other difficulty is one of calibration. The relationship to be assumed, in translating a recorded count-rate into an estimate of internally deposited plutonium, depends on a number of factors, many of them indeterminate and/or peculiar to an individual subject, e.g. the pattern of distribution of the activity within the lungs and the nature and thickness of the soft tissues of the chest wall. Similar problems arise, to a greater or lesser extent, in estimating lung burdens of several other transuranic nuclides.

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Table 1. Principal X-ray emissions from ^{239}Pu and ^{103}Pd, and attenuation coefficients in muscle at relevant energies

Nuclide	X-ray group	Energy (keV)	Intensity* (%)	μ† (cm^{-1})
^{239}Pu	U L_α	13.6	1.67	2.13
	U L_β	17.2	2.38	1.13
	U L_γ	20.2	0.55	0.77
^{103}Pd	Rh K_α	20.2	67	0.77
	Rh K_β	22.8	13	0.60

*Relative intensities from measurements at Harwell employing a 300-mm² Si(Li) detector with a thin beryllium window, normalised to assumed total X-ray intensities of 4.6% for ^{239}Pu (Sw70a) and 80% for ^{103}Pd (Ma76).

†Total attenuation coefficient; values are based on calculations by White (Wh76), using published methods (Wh74), with the elemental composition of muscle assumed to be as given in Int64.

Methods of detection and calibration in the assessment of ^{239}Pu in vivo

Large-area proportional counters (Lan64; Ta69; Ra69; To70; Sh72) or thin scintillation detectors (Lau68; Is69; Sw70b; C173a; Ga77a), often with elaborate anticoincidence arrangements for background reduction, have been developed for these purposes. The performances quoted for these instruments suggest that the best of them are capable of detecting lung burdens of less than 16 nCi in subjects of average physique.

For calibration, most laboratories have relied on phantoms varying in their complexity and anatomical accuracy and containing in almost every case a uniform distribution of plutonium in their "lungs" (Sp64; Bou68; Is69; Wa70; Sw76). Presupposing that a phantom is realistic in its design and in the attenuation properties of its materials, one must adjust the calibration derived in this way to cater for the large majority of subjects whose dimensions do not conform closely to those of the phantom, and these adjustments are most commonly based on estimates of the subject's chest wall thickness by ultrasonic methods (Ra67; Ne72; De73; Ga77b). As an alternative to a phantom for calibration, some limited use of cadavers has been reported (Eh64; Bu69), but apart from its obvious disadvantages, this approach shares one of the major defects of methods employing phantoms, viz. assumptions must be made concerning the distribution of activity within the lungs, and these may not be realistic.

A third possibility, which in principle offers many advantages, is to adopt an *in vivo* calibration method (Ta65; Ru68; Ne71; Ne72; Ra72; Sh76). Aerosols emitting X-rays of suitable energies are inhaled by volunteers selected to represent a variety of physiques, with the aim of establishing correlations between the calibration factor (i.e. response per unit activity present in the lungs) and a simple physical parameter such as weight/height ratio or chest wall thickness, for use in the interpretation of spectra from contaminated subjects. Because attenuation coefficients for X-rays vary markedly with energy in the 13–23-keV region (Table 1), one would employ ideally a nuclide whose X-ray spectrum was similar, in the energies and in the relative intensities of its components, to that of the uranium L X-rays which follow the decay of ^{239}Pu. On this criterion both ^{233}Pa (Bou67; Ne71) and ^{237}Pu (Ra72) are suitable, but in other respects both have serious disadvantages. The use of ^{233}Pa, a beta emitter of half-life 27 days, involves more than a trivial radiation dose to the lungs, if it is administered in sufficient quantities to permit calibration studies over a lengthy period. Plutonium-237 (half-life 45 days) decays by electron capture, and in principle could be administered in microcurie quantities without subjecting volunteers to unacceptable radiation doses. In practice problems arise in producing ^{237}Pu free of alpha-emitting impurities which increase irradiation of the lungs substantially. Protactinium-233 and ^{237}Pu share an additional disadvantage: the emission of abundant radiation (K X- and gamma-rays) in the 60–120-keV region. Particularly when phoswich detectors are used, these quanta give rise to an intense low-energy continuum, on which the L X-ray contributions are superimposed, and this complicates interpretation of the spectra.

The *in vivo* technique which has been employed in most calibration studies at Harwell (Ru68; Ne72) employs 5-μm polystyrene microspheres, incorporating the nuclides ^{103}Pd (half-life 17.0 days) and ^{51}Cr (27.8 days) in known relative concentrations. Palladium-103, decaying by electron capture, emits the K X-rays of rhodium, whose principal (K_α) energy of 20.2 keV coincides with one of those in the uranium L X-ray

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spectrum (Table 1). The ^{51}Cr label, emitting gamma-rays at 323 keV, enables the retention of the particles, and hence the ^{103}Pd content of the subject's lungs at a given stage, to be derived using largely established methods of body radioactivity measurement. The amount of ^{51}Cr in the inhaled mixture can be chosen so that it is easily detected by these methods, but is not so large that its scattered quanta interfere seriously with measurements of the X-rays from the ^{103}Pd. Experience has shown that leaching of activity from the particles *in vivo* is negligible and that, after an early rapid clearance of material deposited in the ciliated regions, elimination from the lungs is a slow process [biological half-life $\gtrsim$ 100 days (Boo67)]. For acceptable levels of radiation dose to the lungs—65 mrem/μCi ^{103}Pd deposited in those regions from which clearance is slow (Sn75), and 13 mrem/μCi for ^{51}Cr (Sn74)—investigations are possible for a period of several months. A disadvantage of ^{103}Pd as a simulator for ^{239}Pu is that only the 20.2-keV component of the uranium L X-ray spectrum is reproduced in its emissions. Thus, in deriving a calibration factor for ^{239}Pu *in vivo*, assumptions have to be made concerning the contributions from the 13.6- and 17.2-keV X-rays relative to that from the 20.2-keV component. A further adjustment, to allow for the effect of the 22.8-keV K_β X-rays from ^{103}Pd, is also required. Uncertainties in the energy-dependence of attenuation within the subject (Ne78b) limit the accuracy with which these adjustments can be made.

The interlaboratory comparison and its objectives

Calibration studies involving the administration of radioactivity to volunteers absorb a substantial technical effort and entail a small radiation dose to the subject. Consequently there is a strong case for increasing the yield of an experiment by making its subjects available for investigation at more than one laboratory. A study of reports in the literature, and enquiries made among laboratories whose methods were unpublished, suggested that it would be particularly useful to circulate subjects containing known amounts of a low-energy photon emitter among a number of institutes equipped for the assessment of plutonium *in vivo*. The techniques of calibration which we encountered in this preliminary survey all differed in some important respect, and it seemed inevitable that the assessments of different laboratories investigating the same subject contaminated with plutonium would cover a wide range. These differences in calibration also prevented a realistic comparison of the sensitivites offered by different equipment and techniques of detection. Indeed, it appeared likely that, in some instances where two laboratories made conflicting claims for the minimum activity detectable with essentially similar equipment, the discrepancies reflected differing assumptions about the effects of attenuation within the body.

Accordingly, during 1972, as an extension of calibration studies carried out at Harwell using the $^{103}Pd/^{51}Cr$ technique, the three subjects involved, representing individuals of small, intermediate and large physique, were made available for investigations elsewhere, with the following objectives:

(i) to compare, on a common realistic basis, the sensitivities of various detectors and methods for the assessment of low-energy photon emitters in lungs.

(ii) to evaluate the calibration techniques employed by participants, or, in cases where no established technique existed, to provide, within the limitations mentioned above, data on calibration and on its variation with body size, for use in the assessment of internal contamination.

(iii) to enable each laboratory to investigate other factors relevant to its procedures, such as the optimum positioning of its detectors relative to the subject.

Participating laboratories

Fourteen organizations participated in the project: Aktiebolaget Atomenergi, Studsvik, Sweden; Argonne National Laboratory, IL, U.S.A.; Atomic Energy Establishment, Winfrith, U.K.; Atomic Energy Research Es-

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tablishment, Harwell, U.K.; Centre d'Etudes Nucléaires, Fontenay-aux-Roses, France; Centro di Studi Nucleari, Casaccia, Italy; Dow Chemical Co. (Rocky Flats Division), CO, U.S.A.; Inhalation Toxicology Research Institute, NM, U.S.A.; Laboratoire du Service Médical du Travail du CEA, Montrouge, France; Lawrence Livermore Laboratory, CA, U.S.A.; Los Alamos Scientific Laboratory, NM, U.S.A.; Mound Laboratory, OH, U.S.A.; New York University Medical Center, U.S.A.; Radiation Protection Bureau, Department of National Health and Welfare, Ottawa, Canada.

METHODS EMPLOYED IN THE INTERCOMPARISON

An exercise entailing visits by our three subjects to 13 other laboratories would inevitably extend over many weeks, during which reduction of the inhaled activities would occur through radioactive decay and biological clearance. Consequently, in planning the organization and experimental details we had to take into account a number of factors and requirements:

(i) Each subject would be at the disposal of individual laboratories for only 1–2 days, and so ideally sufficient ^{103}Pd activity should be present in his lungs to enable each laboratory to make a large number of investigations in this short time. This, together with the need that, for our own purposes at Harwell, the activity should remain easily detectable in two of the subjects for three months or longer, indicated that preferably several microcuries of each nuclide should be deposited in those regions of the lungs from which clearance was slow.

(ii) The retention of the radioactive particles would need to be assessed, by measurements of the remaining ^{51}Cr, at intervals before, during and preferably also after the programs of visits to other laboratories, with the same detector and method employed on each occasion. From interpolated estimates of the ^{51}Cr and the known radioactive composition of the inhaled mixture, the ^{103}Pd content of each subject at the time of his visit to each of the other laboratories could be deduced on a common basis.

(iii) During the course of the exercise it would be necessary to monitor the flux of X-rays at the surface of the chest for any change which could not be attributed to radioactive decay and particle clearance. For example, it was conceivable that attenuation of the X-rays within the lungs might be affected by some changes in the distribution of the particles. This monitoring would entail, for each subject, measurements with the same X-ray detector at intervals during the experiment, to provide a basis for any necessary adjustments before the data of different laboratories were compared.

Generation of the aerosol and administration to volunteers

A monodisperse aerosol of 5-μm polystyrene particles, incorporating the activities ^{51}Cr and ^{103}Pd, was generated by a spinning disc technique as previously described (Boo67; Ru68). A suspension of these particles in ethanol was atomized with air at the top of a column 76 cm tall. The cloud was mixed with diluting air so that the liquid evaporated and at the base of the column the aerosol consisted of dry particles. At this point one horizontal side-arm terminated in a mouthpiece, while a second tube enabled part of the aerosol to be drawn through a filter for sampling purposes; particles not taking either of these routes passed to waste via a third outlet. The equipment was essentially the same as that described in more detail elsewhere (Ho71).

Three subjects (Table 2) inhaled the aerosol through the mouthpiece. A "normal" breathing pattern (one which the subject found natural and comfortable in a resting state, i.e. tidal volume ~0.5 l.) was maintained. As we had found previously (Ne72), the proportion of the retained activity which was deposited in the ciliated regions, and was thus subject to early rapid clearance, was largely unpredictable. A series of exposures,

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Table 2. Biometric data for subjects

Subject†	Age (yr)	Weight (kg)	Height (cm)	Chest circumference (cm)	Chest wall thickness‡ (cm)	Lung function* FEV_1 (l.)	FVC (l.)
DN	35	62	170	92	1.7	4.5 (3.6)	5.4 (4.5)
JR	47	81	178	100	2.5	3.6 (3.5)	4.5 (4.6)
KFB	50	88–92	174	117	3.1	3.7 (3.4)	4.9 (4.6)

*Recorded 2 yr (DN and KFB) and 5 yr (JR) after the interlaboratory exercise; figures in parentheses are typical values for subject's age and height (Ba71). FEV_1 is the volume of air expelled in 1 sec with maximum effort; FVC is the maximum volume expelled after full inspiration.
†All three subjects were male.
‡By ultrasonic methods; mean of estimates by six laboratories.

extending over two days, was necessary in order to achieve the required amount of activity deposited in those regions from which clearance was slow [biological half-life ≥100 days (Boo67)]. Filter samples of the aerosols administered to each subject were obtained as indicated above, and these were measured by the methods of scintillation counting to be described, in order to establish the $^{103}Pd/^{51}Cr$ ratio in the inhaled particles.

Organization of the project, and supporting radioactivity measurements

The ^{51}Cr content of each subject's chest was measured periodically at Harwell until 5–8 days after intake, to ensure that clearance of those particles deposited in the ciliated regions was complete, and to define the early part of the long-term clearance pattern (Fig. 1); several measurements of X-ray emission from the chest were also made. Experimental details are given below. At this point two of the subjects (DN and KFB) were made available for 3 weeks to permit investigations at laboratories in N. America, while the third (JR) spent 10 days visiting establishments in Europe and elsewhere in the U.K. After these visits the subjects returned to Harwell for several days, during which further assessments of their ^{51}Cr contents and X-ray emissions were made. They then left to complete their programs of visits in Europe (DN and KFB) and in N. America (JR). On their subsequent return to Harwell, it was possible to resume these measurements on subjects DN and KFB. Laboratories had each of the three subjects at their disposal for at least 1 day, and the program was so arranged as to give each organization access to at least one subject before radioactive decay of the ^{103}Pd might

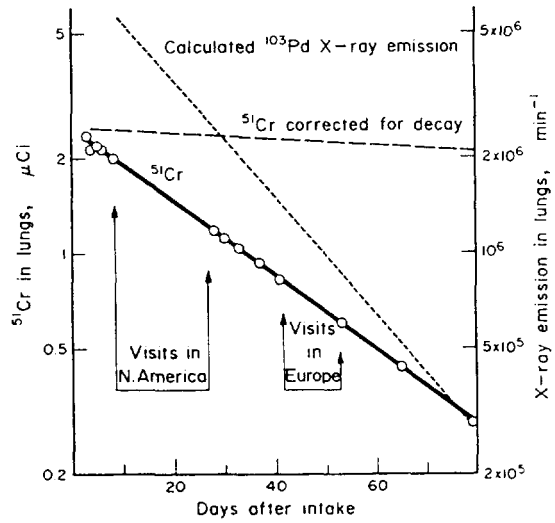

FIG. 1. Radioactivity and clearance of polystyrene particles in subject DN. Plotted points and full line (scale on left): observed ^{51}Cr in lungs, from serial measurements at Harwell, and best-fitting single exponential function of time. Dashed line (scale on left): fitted function corrected for radioactive decay of ^{51}Cr, i.e. biological clearance of particles. Dotted line (scale on right): X-ray emission from ^{103}Pd in lungs, calculated from particle clearance and known radioactive composition of the inhaled aerosol.

prejudice accomplishment of the agreed objectives; in practice, all laboratories found that the X-ray emissions from each of the three subjects were fully adequate for their purposes.

To ensure that all laboratories worked to common standards of activity, each was supplied with sources of ^{103}Pd and ^{51}Cr, deposited on filter paper which was sealed between thin sheets of polyvinyl chloride. The X- and gamma-ray emissions of these

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sources had been recorded at Harwell, by methods to be described below.

Estimation of retained ^{51}Cr. The serial measurements of ^{51}Cr at Harwell were made inside a 10-cm thick lead shield (In70) using a sodium iodide crystal (diameter 23 cm, thickness 15 cm), to record the 323-keV gamma-rays. With the subject in a supine position, two measurements were made on each occasion. For one of these, the detector was positioned 25 cm below the bed, under the mid-point of the subject's sternum, while during the other measurement the crystal occupied a corresponding position over the chest, 47 cm above the bed. The arithmetic mean of the two results was used to estimate his ^{51}Cr content; this mean response would not be materially affected by any changes in the distribution of particles in the lungs during the period of study, which might otherwise distort the observed retention pattern. For calibration purposes, a chest phantom was employed constructed of hardboard (similar to "Presdwood"), containing cavities filled with compressed foam rubber to represent lung tissue. The effect of a distributed source was simulated by measuring a point source of ^{51}Cr in numerous positions inside the "lungs". Differences in detection efficiency between subjects and phantom were minimized by recording the response over a wide energy range (40–415 keV), rather than in a region restricted to the photoelectric peak at 323 keV. Corrections for these differences, which were due primarily to differences in attenuation, were applied when necessary, based on the ratio of count-rates in the peak and forward scatter regions of the subject's spectrum, compared with the same ratio recorded from the phantom.

In addition, the other laboratories were asked to make their own assessments of ^{51}Cr in the subjects. Their methods included fixed sodium iodide crystals (generally 20 × 10 cm or larger) viewing both anterior and posterior surfaces of the chest, similar to the method adopted at Harwell; fixed detectors viewing the anterior surfaces only (chair technique, or crystals in contact with the thorax); whole-body scanning of the supine subject; arc techniques (radius 1.0 or 1.5 m); and use of a 4π-liquid scintillation counter. Calibration procedures included the use of chest phantoms containing "lungs" incorporating ^{51}Cr as at Harwell; whole-body phantoms containing activity widely dispersed; measurements of point sources in air, with semi-empirical adjustments to allow for the effects of attenuation and differences in geometry; and the use of data from other subjects containing ^{51}Cr inhaled under controlled conditions, or other isotopes administered by various routes. Corrections were generally, although not invariably, made to allow for differences in attenuation between subject and phantom, the most common approach being based on considerations of spectral shape.

Monitoring subjects' X-ray emissions at Harwell. Measurements of X-ray emission from the thorax were made at intervals before, during and, in two subjects, after their programs of visits to other laboratories. A summary of the experimental details may be found in Table 4.

Measurement and standardization of sources. Measurements of X- and gamma-ray emissions of all sources employed in these studies, including the filter samples of the inhaled particles and sources provided for other laboratories, were made with a NaI(Tl) detector (diameter 15 cm, thickness 9 cm), at a distance of 44 cm. For our purposes it was not necessary to know the ^{51}Cr activities accurately in absolute terms, provided estimates of the subjects' ^{51}Cr contents made at Harwell and elsewhere were expressed relative to the nominal activity of a filter sample or of a source supplied by us. A calibration factor for ^{51}Cr was however known for this source-detector arrangement, with an estimated accuracy of ± 5%, and, as a matter of convenience, the ^{51}Cr activities quoted in this report will be expressed in microcuries on that basis.

Calibration of the NaI(Tl) detector for ^{103}Pd, with an estimated accuracy of ± 2%, was accomplished by counting a source provided for us by the Lawrence Livermore Laboratory. This source had been standardized by comparing its X-ray emission with photon emissions from absolute standards of

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^{241}Am, ^{137}Cs and ^{57}Co, using both GeLi and SiLi detectors. Other laboratories receiving similar sources from Livermore generally reported agreement with the quoted activity to within ±3% (An74).

Since all ^{103}Pd sources employed in this work were standardized by X-ray counting, activities will be expressed in terms of photon output rather than as microcuries. This will avoid ambiguities which might otherwise occur because of the discrepancies in published estimates of X-ray abundance for ^{103}Pd (Ne78a).

ESTIMATES OF ^{103}Pd IN VIVO FROM MEASUREMENTS OF ^{51}Cr IN LUNGS

Substantial fractions of the inhaled particles were cleared from the upper airways and ciliated regions during the first 2 days after intake, as in previous experiments (Ne72). This early clearance is of no relevance in the present context, and we are concerned solely with the subsequent pattern of retention.

Data on the long-term retention of the particles and their associated radioactivity are summarized in Table 3, and illustrated in Fig. 1 in the case of subject DN. In this figure, the plotted points indicate Harwell's estimates of the ^{51}Cr content, from which an initial value of 2.50 μCi and an effective half-life of 25.7 ± 0.2 days may be deduced. Corrected for radioactive decay of the ^{51}Cr label, this represents a biological half-life of 340 days, indicating the rate at which the polystyrene particles were cleared from the lungs (dashed line in Fig. 1). If we combine this value with the 17.0-day physical half-life of ^{103}Pd, an effective half-life of 16.2 days for this nuclide is indicated, and this enables the subject's X-ray emission at any time during the experiment to be deduced (dotted line in Fig. 1), given the measured ratio of the two nuclides in the inhaled aerosol.

In point of fact, the estimates of X-ray emission given in the final column of Table 3 were derived on a slightly amended basis which took into account the estimates of retained ^{51}Cr supplied by other laboratories, as well as our own. Additional estimates of the initial ^{51}Cr content were obtained by adjusting each laboratory's assessment, according to the effective half-lives in Table 3. The mean of all such values for each subject is included in Table 3 with its standard error and the range of individual values, which was considerable. The methods employed for estimating ^{51}Cr were summarized in a previous section, and some were manifestly more satisfactory than others. We have been unable to formulate defensible criteria on which to calculate a weighted mean of these data, and we have therefore based our calculations of X-ray emission in the subjects on the simple arithmetic mean ^{51}Cr contents in Table 3.

Error estimates given in the final column of Table 3 take into account (i) a quoted uncertainty of ±2% in the activity of the ^{103}Pd which we received from Livermore for standardization purposes (An74), (ii) the standard errors of the mean estimates of ^{51}Cr *in vivo* given in Table 3 and (iii) additional uncertainties of ±6 and ±3% applying to subjects DN and KFB respectively. In these two subjects there were doubts about whether the composition of activity on filter samples relating to small fractions of the inhaled

Table 3. Data on deposition and long-term retention

Subject	Harwell data*				All laboratories			
	Initial ^{51}Cr (μCi)	Half-lives (days)				Initial ^{51}Cr (μCi)		Assumed initial ^{103}Pd (photons/min ÷ 10^6)
		Eff. ^{51}Cr	Biol.	Eff. ^{103}Pd	No.†	Mean ± S.E.	Range	
DN	2.50 ± 0.02	25.7 ± 0.2	340	16.2	18	2.52 ± 0.05	2.15 – 3.05	8.34 ± 0.55
JR	1.22 ± 0.02	23.3 ± 0.9	144	15.2	16	1.15 ± 0.04	0.86 – 1.40	4.89 ± 0.22
KFB	2.65 ± 0.02	25.2 ± 0.3	269	16.0	16	2.37 ± 0.10	1.64 – 3.10	10.62 ± 0.60

*Derived by fitting a single exponential function of time to the serial estimates of ^{51}Cr in lungs, using the computer program FATAL (Sa72). Error estimates are based on scatter about the fitted function, and those attached to the initial ^{51}Cr contents do not include systematic errors introduced in the calibration procedure. We are indebted to Dr. J. Rundo for these analyses.

†Number of independent determinations by different procedures; some laboratories reported individual values based on two or more methods.

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aerosol was representative of the material deposited in the lungs. The errors quoted under (iii) reflect these uncertainties.

STABILITY OF X-RAY DETECTION EFFICIENCIES

X-ray detection efficiencies, observed at Harwell for subjects DN and KFB, are shown plotted against time in Fig. 2, expressed in effect as recorded count-rate per unit activity of ^{103}Pd remaining in the lungs at the time of measurement. For both subjects, neighboring values shown an appreciable scatter which may be attributed largely to difficulties in reproducing the subject-detector geometry precisely on these occasions. In the case of KFB this scatter is superimposed on a noticeable downward trend: when a least-squares analysis of the data was performed, with a linear relationship with time assumed, the lower of the two lines in Fig. 2 was obtained, predicting a reduction of 13% in the efficiency between days 4 and 55. This decline is satisfactorily explained by changes in the subject's weight, which increased by 4 kg during the experiment. A correlation of the recorded efficiencies with body weight is shown in Fig. 3, and a function of the form

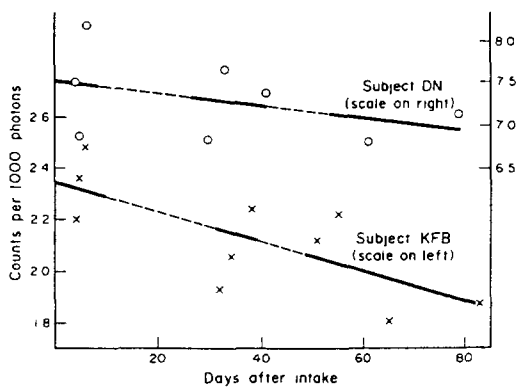

FIG. 2. X-ray detection efficiencies for subjects DN and KFB (recorded counts/1000 photons emitted in lungs), determined at Harwell for two 19-cm diameter phoswich detectors (Table 4). Measurements were made with the subjects supine, and with arms raised and folded behind the head. The linear functions of time providing the best fits to these data are shown, the broken portions indicating the periods during which other laboratories made their measurements.

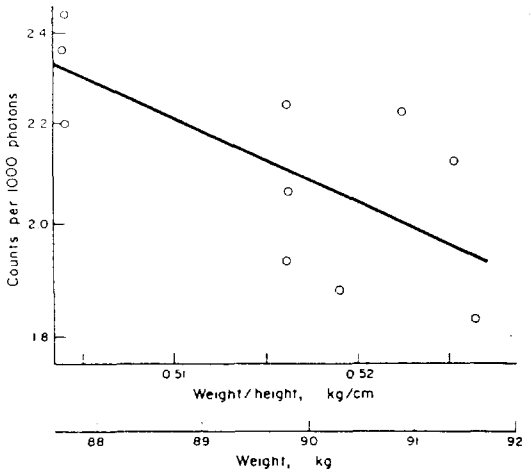

FIG. 3. X-ray detection efficiencies for subject KFB measured at Harwell, correlated with his body weight. The line is the best-fitting function with the form of equation (1). Note that the vertical scale is logarithmic.

$$C(W/H) = C(0.50)\exp[-a(W/H - 0.50)] \quad (1)$$

was fitted to these data by least squares. In equation (1), $C(W/H)$ is the counting efficiency (in counts/photon) for a given value of the ratio weight/height (in kg/cm), and a plot of the calculated function is included in Fig. 3. The best-fitting values for the two parameters were:

$$C(0.50) = (2.40 \pm 0.12) \times 10^{-3} \text{ counts/photon}$$
$$a = 8.0 \pm 3.1 \text{ cm/kg}.$$

Previous data (Ru68), correlating calibration factors for ^{103}Pd with W/H in seven subjects ranging in weight from 61 to 107 kg, were subjected to the same method of analysis, and the result

$$a = 8.2 \pm 1.3 \text{ cm/kg}$$

was obtained, similar to the estimate given above. Thus it would seem reasonable to assume that the decline in detection efficiency for KFB was due entirely to his increasing body weight. Equation (1), with $C(0.50) = 2.40 \times 10^{-3}$ and $a = 8.0$, was used to adjust the data for KFB in Fig. 2 for these changes. Least-squares analysis then indicated a

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change of $0 \pm 5\%$ in the adjusted efficiencies between days 4 and 55.

For subject DN, the slightly negative slope of the straight line best fitting the data of Fig. 2 was not significant, predicting a reduction in efficiency of $4.7 \pm 4.3\%$ between days 4 and 55. During the experiment, values of this subject's weight recorded at Harwell varied only within the range 60.9–62.0 kg, in a seemingly random manner.

Thus, in subjects DN and KFB, there was no evidence of any important changes in X-ray emission from the body such as might result from a change in the distribution of particles within the lungs, or from the preferential leaching of either nuclide from the polystyrene. The data for JR do not extend beyond day 17 and are insufficient to permit such conclusions, but there is no reason to suspect that the particles would have behaved differently in him. No systematic variations in his body weight occurred during the interlaboratory exercise.

RESULTS OF INTERLABORATORY COMPARISON

Following their measurements on the three subjects, which took place between October and December, 1972, laboratories provided us with a summary of their methods and findings for inclusion in this paper, and several have in addition reported independently on their experience in the project (Ru73; C173b; An76; To76).

A provisional report, based on a preliminary analysis of incomplete data, was prepared in 1974 and subsequently issued by the IAEA (Ne76). Many of the statements and data in that account have been amended in the preparation of this paper, following revisions to or clarification of the material originally submitted to us. In some instances, however, there remain ambiguities in the interpretation of participants' reports which we have been unable to resolve satisfactorily. For this reason, and also in response to specific requests from some laboratories for anonymity—on entirely legitimate grounds—we have not, except in the case of Harwell, divulged the origin of each laboratory's results. Instead, we have adopted a system of code letters to refer to participants.

Detection efficiencies and sensitivities for ^{103}Pd *in lungs*

Each laboratory was asked to measure the response from the X-rays, employing as far as possible the equipment and techniques which it used, or would envisage using should the need arise, to estimate internal contamination with plutonium. Most laboratories used two detectors, and with one exception (lab. K) the counter(s) viewed the anterior surfaces of the thorax. Details of these detectors, their arrangement relative to the thorax, and the response per unit activity, E, for each subject, are included in Tables 4–6. Each value of E was derived by dividing a laboratory's observed count-rate, after certain adjustments, by the contemporary X-ray emission-rate calculated from the data of Table 3. Some laboratories provided details of investigations made by more than one method, and these additional data have been included in cases where they are useful in illustrating points made in the text.

Phoswich detectors. These data are summarized in Tables 4 and 5. Laboratories specified a variety of energy ranges in reporting their count-rates, and the basis on which these had been selected was not always clear. In some instances the chosen band extended up to only 25 keV, and this choice was probably intended to optimise the sensitivity for detecting plutonium. However, it would impair somewhat the efficiency for measuring ^{103}Pd, in view of the poor energy resolution of phoswich detectors and the presence of K_β radiation at 22.8 keV. The percentages of the total peak area falling in the specified ranges were estimated by reference to spectra of ^{103}Pd *in vivo* supplied by the laboratory concerned, or by another employing detectors of similar size and supposedly similar energy resolution, and these percentages appear, in parentheses after the energy ranges, in Table 4. According to these percentages, the reported count-rates were adjusted before the values of E were calculated, so that in each case the latter reflect the approximate full peak response and enable more valid comparisons to be made between the data of different laboratories. Laboratories contributing the data in

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Table 4. Performance of two-phoswich systems

Lab	Detector dimensions*	Geometry, etc.	Energy range, keV (and % of total peak recorded)	Subject DN (62 kg) E†	Subject DN (62 kg) F†	Subject JR (81 kg) E†	Subject JR (81 kg) F†	Subject KFB (90 kg) E†	Subject KFB (90 kg) F†
A	12.7 × 0.10	One viewing each lung, circumference tangential to sternum and clavicle, angled for maximum contact with body	14–25 (86)						
		Arms behind head		1.09	47	0.53	11	0.27	3
B	12.7 × 0.30	Detectors as for A	12–29 (97)						
		Arms beside torso		3.59	510	1.87	138	0.96	36
C	12 × 0.16	Detectors as for A	15–27 (96)						
		Arms beside torso		3.10	425	1.84	150	133	78
D	12.7 × 0.10	Detectors as for A	10–25 (89)						
		Arms beside torso				2.10	174	1.25	62
		Arms behind head		4.30	730	2.75	298	1.33	70
E	12.7 × 0.10	Detectors as for A	15–25 (90)						
		Arms beside torso		3.28	425	2.68	283	1.69	113
F	15 × 0.30	(1) Detectors similar to A, but recessed in perspex cylinders, window 1.5 cm from body	12–29 (97)						
		Arms beside torso		4.63	607	2.10	125	1.53	66
		(2) As for (1), but detectors displaced 3 cm towards vertex		2.59	190	1.90	102	1.38	54
Harwell	19 × 0.30	One viewing each lung, at level of mid-sternum‡	10–32 (100)						
		Arms beside torso		5.40	514	2.87	145	1.47	38
		Arms behind head		8.20	1186	3.71	243	2.35	97

*Diameter × thickness (cm) of NaI(Tl). In some cases where the detector canning obscured the periphery of the window, an estimate of the effective diameter has been made.

†E = counts in full X-ray peak per 1000 photons emitted in lungs; the values for subject KFB have been adjusted to compensate for changes in his body weight (see text). F (figure of merit) = E^2 ÷ detector area expressed in m^2.

‡Detectors displaced laterally by 11.5 cm to left and right of the central position described in Table 5, without change in orientation.

Table 5. Performance of alternative phoswich systems

Lab	Detector dimensions*	Geometry, etc.	Subject DN (62 kg) E†	Subject DN (62 kg) E†	Subject JR (81 kg) E†	Subject JR (81 kg) F†	Subject KFB (90 kg) E†	Subject KFB (90 kg) F†
Harwell	19 × 0.30	Single detector, centrally over mid-sternum of supine subject; rotated about an axis orthogonal to mid-sagittal plane to follow surface contours of upper thorax; separation from body ~1 cm						
		Arms beside torso	3.32	388			1.17	48
		Arms behind head	4.28	646	2.30	186	1.34	63
G	12.5 × 0.05	Single detector over right lung, between ribs 2 and 5						
		Arms behind head	1.73	243	0.85	59	0.56	26
H	10.2 × 0.40	Four detectors in contact with front of chest (left upper, left lower, right upper, right lower)						
		Arms beside torso	5.13	805	1.38	58	1.23	46

* and † See corresponding footnotes to Table 4.

Table 5 reported full peak count-rates and here no adjustment was needed.

A further source of instrumental variation affecting the results concerned the efficiency of pulse shape discrimination (PSD) circuits in admitting signals resulting from X-ray interactions in the NaI(Tl) element of a phoswich. Laboratories providing this information (C, E and G) reported efficiencies in the range 90–95%, and we have assumed this to apply to all laboratories except Harwell, where at the time of these experiments, the efficiency was known to be only 80% for the X-rays emitted by ^{103}Pd. The performance of our equipment has since been improved, and at 90% the PSD efficiency is now typical of that applying elsewhere. Consequently, in calculating values of E for the Harwell counter, an additional multiplying factor of 90/80 has been applied to the observed count-rates.

The data for KFB from all laboratories have been further adjusted to take account of the increase in X-ray attenuation which occurred in this subject during the experiment (Figs. 2 and 3). A normalizing factor, derived from the linear function fitted to the data in

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Table 6. Performance of proportional counters

Lab	Detector and shield	Geometry§, etc.	Background counts/min Detector	Background counts/min Subject‖	Efficiency, E† for subject DN (62 kg)	JR (81 kg)	KFB (90 kg)
J	CPX180.** Shield 40 cm sand + 6 cm Pb	Centrally above thorax; subject measured supine and prone¶	7 (17–27 keV)	4	0.42	0.33	0.30
K	CPX180. Shield 10 cm Pb	(1) Centrally above sternum of supine subject	10	5	0.72	0.53	0.31
		(2) In right armpit, viewing lateral surface (preferred method)			1.82	0.55	0.46
L	CPX180. Shield 20 cm Fe + 0.3 cm Pb + 0.08 cm Cd. 90 cm concrete + 340 cm earth above laboratory	Centrally above sternum of supine subject	4 (16–24 keV)	2	1.02	0.50	0.26
M	Twin Ar/CH_4 flow counters, area 27.5 × 15 cm^2. Shield 16 cm Fe + 0.3 cm Pb	One over each lung	1.7 (2 counters, 11–25 keV)	0.7	0.22*	0.11*	0.04*
N	Sealed detector, area 350 cm^2. Filling $Ar/Xe/CH_4$. Shield 10 cm Pb, partially lined with 0.2 cm Cu	Centrally over thorax of supine subject‡	1.7 (14–25 keV)	2.0	0.49	0.22	0.17
Harwell	Ar/CH_4 flow counter, 30 × 30 × 10 cm^3. Shield 10 cm Pb + 0.1 cm Cd	Centrally above thorax of supine subject	0.4 (14–24 keV)	0.4	0.47	0.27	0.11

*The response was in all cases higher with the subject's arms raised and folded behind the head, by an average factor of 1.6.
†See corresponding footnote to Table 4.
‡Lab. N estimates that a pair of such detectors, one viewing each lung, would give 1.7 times the response shown for a single counter.
§Efficiencies quoted are for measurements with subject in "arms beside torso" posture.
¶Detector arranged at approximately constant height above bed; hence the geometry for subject DN (overall chest thickness 21 cm) was less efficient than for JR (24 cm) and KFB (26 cm).
‖Typical contributions from ^{40}K and ^{137}Cs at 1972 levels.
**Sealed counter with Xe/CH_4 filling, window 18-cm diameter, manufactured by *Compagnie Générale de Radiologie*, France.

Fig. 2, was applied so that the values of E show the expected result, had the measurements of all laboratories been made on day 45, when the subject's weight was ~ 90 kg.

One of our important objectives was to compare the performances of phoswich detectors of various sizes, and in various configurations relative to the body, in terms of some figure of merit for sensitivity in detecting ^{103}Pd in lungs. Provided the pattern of X-ray emission from the various regions of the chest is not strongly energy-dependent in the 17–20-keV region, these relative performances should broadly speaking apply in the assessment of plutonium *in vivo*. A quantity commonly employed as a figure of merit in comparing low-level counters is S^2/B, where S is the detector count-rate contributed by a reference source and B is the background response of the detector. In the present context, B would include effects arising from the body's influence on the ambient radiation field and also from scattered quanta from natural ^{40}K and fallout ^{137}Cs *in vivo*, as well as the intrinsic counter background. For sufficiently weak samples, giving a count-rate $C \ll B$, S^2/B is inversely proportional to the total counting time, spent in determining $(C + B)$ and B, which is necessary to achieve a given percentage statistical precision in C(Lo51). In the routine monitoring of workers for the presence of plutonium *in vivo*, the condition $C \ll B$ will generally be met. Of course, in experimentally comparing S^2/B for different counters with a reference source, we do not need to make $S \ll B$, since the ratio of count-rates S_1/S_2 for two detectors functioning correctly will be independent of source strength. Thus for these purposes our subjects, for whom $S \gg B$, may be regarded as suitable reference sources. To allow for the reduction of activity in their lungs between their visits to the various laboratories, through radioactive decay and biological elimination, we may employ a figure of merit, M, based on the values of efficiency, E in Tables 4 and 5, instead of on the observed count-rates, S, i.e.

$$M = E^2/B. \quad (2)$$

However M is unsuited to our purposes, since the background B will depend on factors which in this study varied from laboratory to laboratory, such as the nature

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and thickness of materials employed for shielding. To make the desired comparison, a means must be found of eliminating these variables. Consequently, we have assumed that the background response (as we have defined it above) would, inside a given shield, be proportional to the surface area of a phoswich, and we shall employ, as a figure of merit,

$$F = E^2/\text{area}. \quad (3)$$

The background would also be influenced to some extent by the thickness of scintillator employed to detect X-rays, which varied among laboratories from 0.5 to 4.0 mm. However, throughout most of this range the efficiency for 20-keV radiation would not be materially affected by these differences, and F can be used to indicate how the sensitivities of detectors of a given thickness are affected by the crystal diameter and by their geometrical arrangement relative to the subject. Values of F are included in Tables 4 and 5.

The entries in Table 4 for labs A–E relate to essentially similar pairs of phoswiches, of ~12-cm diameter, yet there were large differences in their detection efficiencies E for a given subject. In particular, lab A reported values which were very much lower than for any of the others, in all three subjects, despite having imposed on them a posture which ought to have increased E (see below); possibly over-rigorous criteria in this laboratory's PSD circuits impaired the detection efficiency. The variations in E for labs B–E (with the subjects adopting the same arms-beside-torso posture) probably reflect differences in what was nominally a very similar geometry, since some laboratories found the response to depend critically on the position and orientation of the detector. Overall, however, the performances of labs B–E did not vary widely, and certainly there was among them no laboratory which returned values of E which were higher or lower than those of the others in all three subjects. With the exception of lab A, the same is true of all of the laboratories in Table 4 when the figure of merit, F is considered. On this criterion the larger detectors of lab F or of Harwell appear to offer no advantage.

From the data for Harwell in Tables 4 and 5, we may deduce that a single 19-cm diameter detector, centrally placed over the upper thorax, would on the whole be two-thirds as sensitive as a system employing two of these phoswiches. It would also in general be less sensitive than the two-phoswich systems of labs B–E, despite having a surface area some 25% larger. The four-phoswich arrangement investigated by lab H (Table 5) was relatively insensitive for the larger subjects, probably because the count-rates from the two lower detectors would have been relatively small. If just two of these 10-cm diameter detectors had been employed, viewing only the upper parts of the thorax, the sensitivity would in all probability have been similar to those of the 12-cm units of labs B–E.

Proportional counters. Six laboratories reported measurements with gas-filled proportional counters (Table 6). In three cases the commercially available CPX 180 detector was employed, while the other three used counters of their own design and construction.

The figure of merit, F, based on surface area as an index of background response (equation 3), which was adopted to compare the sensitivities of phoswich detectors, cannot be applied to proportional counters. For phoswich detectors E can be assumed for our purposes to be independent of the thickness of the NaI(Tl) scintillator, but with proportional counters both E and the background will depend strongly on the nature and depth of the filling gas. Consequently our data do not enable us to deduce quantitatively how the sensitivities of different counters and procedures would compare if they were employed inside a given shield. However, some general conclusions on the relative performances of phoswiches and gas counters are possible.

If the figure of merit M (equation 2) is adopted, the Ar/CH_4 counter (Ta69) at Harwell can be shown from the data in Table 6 to be somewhat more sensitive than the other gas counters. Data (In70) on the background response of thick NaI(Tl) detectors at these laboratories suggest that the differences in M are not related primarily to differences in

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room shielding. For subject JR, the data for Harwell in Table 6 indicate

$$M = 9.1 \times 10^{-8}\ (\text{counts/photon})^2/(\text{counts/min}).$$

By contrast, for two 19-cm phoswiches in the same shield (background 47 counts/min in range 10–32 keV, including contributions from ^{40}K and ^{137}Cs in the subject), we have for subject JR (Table 4), in the same units

$$M = 1.8 \times 10^{-7}.$$

Thus, on this criterion the most sensitive of the gas counters was 2.0 times less sensitive than Harwell's two-phoswich system, which in its performance was fairly typical of the systems included in Table 4. The comparison was somewhat less favorable to the proportional counter for the other two subjects, and the average ratio of sensitivities for all three was 2.4. The values of M can of course be influenced by changes in the energy bands for which data are given in Tables 4 and 6; if these bands were adjusted to optimise M for each detector, the assessment would improve further in favor of the phoswich. These conclusions relate of course to detection of the 20.2- and 22.8-keV photons from ^{103}Pd. For the lower energy components (13.6 and 17.2 keV) in the spectrum from ^{239}Pu, the intrinsic efficiency of the gas counter is greater, while that of the phoswich is essentially unchanged. Consequently, for the detection of ^{239}Pu in lungs, the better proportional counters may compare more closely in sensitivity with phoswich detectors than our data have suggested. Indeed, in certain instances where their much superior energy resolution is useful in minimizing interference from other low energy photon emitters, gas counters may prove to be the more sensitive instruments.

Emission of X-rays from the surface of the thorax

Several participants took advantage of the easily detectable X-ray emissions from the subjects to study the possibilities of employing detectors in alternative, or additional positions which might result in higher sensitivity than was available with established practices. The results were generally disappointing; their findings are summarized below, together with other relevant observations made in the course of these investigations.

Anterior surfaces. In most cases these studies involved a series of measurements with a single detector, most commonly a 12-cm diameter phoswich, occupying various positions in contact with the chest. Harwell and also labs D and G found that at all levels on the thorax, and in all subjects, a higher response was obtained from the right half of the chest compared with that from a corresponding position on the left side; the results of lab. G, which reported increases of 91% (DN), 26% (JR) and 59% (KFB) on transferring a detector from the upper left regions of the thorax to a similar position over the right lung, were typical. A lower response is of course anticipated from the left side because of the presence of the heart. However, another laboratory (Ru73) made a more detailed study, using a collimated detector to record both the 20-keV X-rays from ^{103}Pd and the 323-keV gamma rays from ^{51}Cr emitted by subjects JR and KFB. It concluded that a difference in response between right and left sides was also found in regions anatomically superior to the heart, and that in these regions it arose from a larger deposition of activity in the right lung, perhaps because of its greater size, rather than from increased attenuation on the left side.

The highest response in all three subjects was recorded with detectors mounted high on the thorax, i.e. in the positions employed (Table 4) by labs A–F. In this region the response of smaller detectors (15 cm diameter or less) was often reported to depend critically on position and orientation, especially for the smallest of the subjects (DN). This is evident in the results of Table 4 for lab F, showing that a movement of only 3 cm produced a change in the response approaching a factor of two, and similar effects were reported by lab C for this subject. Lab D reported that, in one instance, simply removing the detector from subject KFB and replacing it in what was believed to be an identical position increased the count-rate by 62%.

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The response of the 19-cm diameter detector system studied at Harwell (Table 4) could certainly be influenced by substantial changes of orientation, but to a considerably smaller extent than was found with smaller units, and for a given geometry E could be reproduced satisfactorily. In Table 4, the adjusted value of 8.20×10^{-3} counts/photon for subject DN was calculated from the mean of eight observations between days 4 and 79 after intake, which showed a standard deviation of only $\pm 6.2\%$. Similarly, for subject KFB, the standard deviation of the 10 values in Fig. 3 about the fitted line was $\pm 6.3\%$.

At many laboratories, measurements were made with the subject adopting two postures, (i) in a "normal" supine position, i.e. with the arms resting beside the torso and (ii) in a modified position with the arms raised and folded so that the hands rested under the head. Most laboratories reported a substantially increased response for the modified posture, and this can be seen in the data for Harwell and for lab. D in Tables 4 and 5. The effect is at least partly associated with a redistribution of the surface tissues of the thorax (Ne72).

Lab C made serial measurements covering a period of ~30 min using the arrangement referred to in Table 4, and reported for all three subjects a progressive reduction in response of 10–20%. Part of this could be explained by the increase in the distance between subject and detector, associated with changes in the shape of the thorax in a supine subject. However, repositioning the detectors to restore the separation to its original value did not fully restore the initial efficiency, suggesting that internal rearrangements of organs and tissues may have affected attenuation and/or geometry. A related matter may be the observation at Harwell that the response from subject DN, during a period of breath-holding following deep inspiration, was 78% greater than during a period of sustained deflation. For subject JR a similar effect (69% increase) was observed. No data were available for KFB.

Posterior surfaces. Several laboratories explored the possibility of increasing the overall response using additional counters viewing the rear of the thorax. The results were not encouraging. From measurements reported by lab G it appeared that a pair of 12.5-cm diameter phoswiches viewing the lungs from the most efficient positions behind the thorax would give 72% (DN), 73% (JR) and 84% (KFB) of the response of similar counters positioned for maximum sensitivity over the anterior surfaces of the upper thorax. However, to achieve this result it was necessary for the subject to flex his shoulders so as to maximise the separation of the scapulae, and this would be incompatible with the posture required to maximize X-ray emission from the frontal surfaces. Lab G's findings were substantially confirmed by investigations at Harwell, both with the present three subjects and during previous experiments (Ne72). On the other hand, two other laboratories, while confirming the reduced X-ray emissions from the posterior surfaces of subjects DN and JR, reported that the response for subject KFB, using 12-cm diameter detectors viewing the back of the upper thorax, was 40% (lab C) and 56% (lab D) higher than from corresponding locations on the frontal surfaces (the positions described in Table 4). It seemed that the emission from the posterior surfaces was highly non-uniform, and while localized regions might exist in which the flux would exceed that from the frontal surfaces of the upper thorax, predicting the location of these areas in an individual would be exceedingly difficult.

Lateral surfaces. Two laboratories (D and G) investigated the value of additional 12.7-cm diameter phoswiches, one viewing each of the lateral surfaces; their data showed that, except for the smallest subject, the extra pair would add less than 30% to the efficiency of two such detectors viewing the frontal aspects, so that the sensitivity of the envisaged combination of four would be poorer.

Lab K, using an 18-cm diameter proportional counter, found the right armpit a more efficient location than a central position over the sternum for all three subjects (Table 6), and if only one detector is available regards this as a preferred procedure.

Estimates of ^{103}Pd *in lungs*

Prior to receiving the subjects, laboratories

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had been informed of the approximate ^{103}Pd content of each, relative to that of the source provided by Harwell, based on our measurements of ^{51}Cr retention during the first few days after intake. They were asked to make independent estimates of each subject's content from their recorded X-ray count-rates, employing as far as possible their established calibration methods, but with ^{103}Pd substituted for plutonium. Some laboratories (D, F, H and L), either having no formal commitment to the assessment of low energy X-ray emitters *in vivo* or regarding the measurement of associated ^{241}Am as the most reliable method of estimating plutonium in the lungs, had not developed any such techniques; they were nevertheless invited to provide estimates on any basis available to them.

The methods of calibration adopted by each laboratory, and their assessments of ^{103}Pd in the subjects, are summarized in Table 7; the methods are described in more detail elsewhere (Ne78c). The estimates are expressed as a fraction P of the ^{103}Pd activity at the time of measurement, deduced from the initial values and effective half-lives in Table 3. Thus, P represents, for each subject, the ratio of a laboratory's estimate, based on its measurements of X-ray emission from the chest, to that derived from the subject's ^{51}Cr content and the relative concentrations of the two nuclides in the inhaled aerosol.

Only two laboratories employed methods which did not involve elaborate phantoms. Lab H, using a method of semi-empirical calculation originally devised for use with gamma-ray emitters of higher energy, produced values of P all substantially greater than 1, although for individual subjects the excess was within, or comparable with, the large uncertainty quoted. The

Table 7. Laboratories' estimates of ^{103}Pd in lungs, each expressed as a fraction P of the value derived from the subject's ^{51}Cr content and the ^{103}Pd/^{51}Cr ratio in the inhaled mixture

Lab	Basis of calibration† for ^{103}Pd in lungs	Adjustments for subject's physique	Estimated ^{103}Pd Content, P		
			Subject DN (62 kg)	Subject JR (81 kg)	Subject KFB (90 kg)
G*	Phantom, with ^{103}Pd-loaded lungs	None	1.02	0.50	0.33
K*	Alderson Remcal phantom, with point source of ^{103}Pd at centre of right lung	None	0.88	0.27	0.24
M*	Phantom (similar in size to subject DN), with ^{239}Pu-loaded sawdust as lung substitute	None	0.69	0.35	0.12
A*	Alderson Remcal phantom, with ^{239}Pu-loaded lungs		0.67 ± 0.12‡	0.56 ± 0.11	0.57 ± 0.10
B*			0.30	0.32	0.21
C*	Alderson Remab phantom, with ^{103}Pd-loaded lungs	Based on ultrasonic measurements of chest wall thickness	0.53 ± 0.13§	0.59 ± 0.14	0.60 ± 0.15
D			1.52	1.71	1.35
E*			0.33	0.53	0.45
F	Alderson Rando phantom, with ^{103}Pd-loaded lungs	Published correlations (Ne72) of detection efficiency for ^{103}Pd with chest circumference	0.39	0.29	0.27
J*	Alderson Rando phantom, with ^{239}Pu-loaded lungs	Based on transmission of 60-keV γ-rays through thorax	0.44	0.62	1.12
N*	Specially designed phantom, with ^{103}Pd-loaded Temex foamed lungs	Based on mean thickness of soft tissue overlying rib cage, predicted from published correlations with anatomical variables (Ra67)	0.94	1.11	1.27
H	Semi-empirical calculation, allowing for geometrical factors and attenuation in lungs, overlying soft tissue and bone. The lungs were represented by four spheres, corresponding to the regions viewed by each of the four detectors (Table 5)		1.4 ± 0.9	1.8 ± 1.0	2.7 ± 1.6
L	Recorded efficiency for a point source of ^{103}Pd in air, adjusted according to published data on attenuation of 20-keV X-rays *in vivo* (Ru69)		0.83	0.93	0.86

*The method of calibration adopted was similar to the laboratory's procedure employed in the assessment of plutonium in lungs, or envisaged for this purpose.

†Equipment and methods of detection were as set out in Tables 4–6. Where a phantom was employed for calibration, the distribution of ^{103}Pd or ^{239}Pu in its lungs was uniform, or essentially uniform, except where otherwise stated. Where Alderson phantoms were used, the lungs were prepared from Alderson lung substitute. All phantoms incorporated the thoracic components of a human skeleton.

‡The origin of these uncertainties was unspecified, but they include possible errors of 10% in the ^{239}Pu content of the phantom.

§The principal contributions to these estimated errors were stated to be uncertainties in chest wall thickness (responsible for ± 13%), in distribution pattern of activity in subject's lungs (± 15%), and in validity of Remab phantom as a match for humans (± 15%).

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reason was possibly that, with the assumed spherical distributions of activity, the average depth of deposition below the surface of the body was overestimated. Lab. L used correlations which in effect enable one to predict the attenuation of 20-keV X-rays in the lungs and chest wall, and quoted values of P all within 20% of unity. In as much as the correlations (Ru69) were themselves derived from studies (Ru68) in which ^{103}Pd had been inhaled by volunteers, lab. L's success should perhaps be regarded largely as a useful demonstration of essential consistency between the results of the present and previous investigations.

In calculating their estimates of ^{103}Pd *in vivo*, three laboratories (G, K and M) each applied a single calibration factor to the count-rates they observed from all three subjects, on the grounds that no established basis had been developed for adjusting the results for differences in attenuation, etc. between subjects and phantom. The progressive reductions in their estimates of P for the larger subjects consequently reflect similar trends evident in the values of E for these laboratories in Tables 5 and 6.

The other laboratories (A–F, J and N) in Table 7 each adjusted the response from a phantom according to the subject's physique, in order to derive an appropriate calibration factor. Except for lab. N's phantom, these were Alderson products, in some cases with modifications to the manufacturer's design. Calibration with these proprietary phantoms generally led to low values of P, in the range 0.2–0.7. The major exception to this was lab. D (P between 1.3 and 1.7); there is no wholly satisfactory explanation for this anomaly, but it may be connected with lab D's statement that the structure of its phantom had become distorted during storage. The strongly upward trend of lab J's values in the larger subjects is at least partly explained by the geometrical factors outlined in a footnote to Table 6; these would have had a minimum effect for subject JR, for whom P was 0.62, i.e. within the range 0.2–0.7 quoted above. The results shown for lab A are not wholly valid in this context. In deriving calibration factors appropriate to ^{103}Pd in our subjects, lab A employed the count-rate from ^{239}Pu in its phantom, adjusted for (i) differences in the X-ray abundances of the two nuclides and (ii) the estimated transmission of Rh K X-rays through the subject's chest wall, relative to that of U L X-rays through the chest wall of the phantom. There was no allowance for the differential attenuation of the two X-ray spectra within the lungs, and if it had been possible to take this into account, the resulting values of P for lab A would have been lower.

The estimates of labs A–F show no marked or consistent trend according to physique, suggesting that their procedures (most of them entailing ultrasonic measurements of chest wall thickness) correctly predicted the variability of the calibration factor between subjects, but that their basic standards, i.e. the phantoms, were inappropriate. It seems to us likely that the implied errors in calibration for laboratories using these Alderson phantoms arose (i) because the phantoms were constructed of materials which were not closely tissue-equivalent at 20 keV, and/or (ii) because they were not sufficiently realistic anatomically and/or (iii) because the almost universal assumption of a uniform distribution of activity in the lungs was not justified. Errors in allowing for attenuation within the chest wall, arising from the use of inappropriate materials, were probably not responsible for major inaccuracies, except possibly in two cases. Labs F and J used an Alderson Rando phantom, incorporating a nominally muscle-equivalent material as a substitute for the soft tissues; the mass attenuation coefficient of this substitute at 17.4 keV is reported (Wh74) to be only 76% of that for real muscle, although in practice the material may be more suitable than this comparison would suggest, if the presence of adipose tissue in the chest wall is taken into account (To75). In the Alderson Remab and Remcal phantoms used by labs A–E, the "soft tissues" of the chest wall consisted essentially of water, a much more valid substitute for muscle, contained between thin plastic shells, and moreover the procedures of three laboratories (A, B and C) were designed to eliminate errors arising from any mismatching (Ne78c). A more probable source of error was the use of an inappropriate lung

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substitute in all of the Alderson phantoms. At 17.4 keV, the mass attenuation coefficient for Alderson lung material is reported to be only 65% of that for the human tissue (Wh74). If a similar discrepancy is assumed to exist at 20 keV, one may predict, by a method described elsewhere (Ne78b), that calibration with such a phantom would overestimate, by ~30%, the detection efficiency for ^{103}Pd uniformly distributed in the lungs. This would lead to $P \sim 0.77$, if it were the only source of error. The other factors, (ii) and (iii) above, may explain why the actual values obtained with Alderson phantoms were generally lower than 0.77.

Of those laboratories employing phantoms, only lab N produced values of P acceptably close to unity in all three subjects. This laboratory used ^{103}Pd-loaded Temex* foamed lungs inside its purpose-built phantom. Temex lung is more similar to soft tissue in its mass attenuation properties at 17 keV than were the substitutes employed in other phantoms (Wh76), lending support to our view that this factor may have contributed to the errors in the estimates of labs A, B, C, E, F and J. However, a curious paradox appears in the data supplied by lab N. If the values in Table 7 were taken to imply that its phantom was realistic in its geometrical arrangement and attenuation properties for 20 keV radiation, then one would expect its accuracy at 17 keV to be comparable. It was therefore surprising that, from its measurements of ^{239}Pu and ^{103}Pd in the phantom, lab. N quoted calibration factors E_{Pu} and E_{Pd} which were numerically similar when expressed as counts per photon emitted in the lungs. From the data of Table 1 one would anticipate, in a typical subject of 2.5-cm chest wall thickness, more than a factor of two difference in the transmission through the chest wall of X-rays from the two nuclides, and one would expect to find E_{Pd} further enhanced relative to E_{Pu} because of differential self-absorption within the lungs. It may be noted that unresolved contradictions of this nature have emerged in previous work (Bu69; De70; Bu70).

*Manufactured by James Girdler & Co. Ltd., London.

Calibration factors for plutonium

Participants were invited to suggest methods by which calibration factors E_{Pu} for plutonium *in vivo* might be derived from their observed values of E_{Pd} (Tables 4–6). Only a minority did so. One laboratory (An76) made adjustments for the differences in attenuation of the two spectra within the chest wall, and the resulting values of E_{Pu} were some 30% lower than had been deduced from measurements of plutonium in its Alderson phantom; differential attenuation within the lungs, had it been possible to assess this reliably, would presumably have led to still lower values. A similar approach suggested by lab B leads to conclusions which are much the same when applied to its data. Three advocated use of the concept of "effective tissue thickness" (Ru69), or approaches which were fundamentally similar, and one of these laboratories (To76) has reported essential consistency between the values of E_{Pu} thus derived and calibration factors deduced from a simple phantom containing plutonium. One laboratory developed a theoretical model (Co73) to estimate the relative attenuation, both in the lungs and in the chest wall, of radiation from the two nuclides, and this led to estimates of E_{Pu} which were lower by a factor of 3 than those suggested by its phantom. Its predictions were broadly consistent with those of another model developed for the same purpose at Harwell. A description of the latter, and some estimates of E_{Pu} derived on this basis, may be found elsewhere (Ne78b).

DISCUSSION

The evidence of Tables 4–6 suggests that in most situations relatively small phoswich detectors are more useful in the assessment of low-energy X-ray emitters than are large-area gas counters. In addition to this advantage of sensitivity, the phoswich has the merits of being more robust and, in our experience, less prone to occasional instability in operation. Our survey has failed to identify or suggest a detection procedure of greater sensitivity than the most common arrangement of two 12-cm diameter phoswiches viewing the anterior surfaces of the up-

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per thorax. We can see no advantage in the addition of further detectors viewing other thoracic regions, or in the substitution of larger detectors, except that the response of the latter may be less strongly influenced by small changes in position or orientation and that larger units are more versatile in their application. They would be more useful, for example, than 12-cm diameter counters in the detection of 60-keV radiation from ^{241}Am which is systemic, i.e. not confined to the relatively small volume of the lungs.

The calibration techniques employed by participants (Table 7) showed a marked tendency to underestimate the subjects' ^{103}Pd contents, in some instances by more than a factor of 3 on average, rather than to overestimate them. Laboratories were reluctant to attach error estimates to their assessments. Of the three (A, C and H) which did, two apparently underestimated the true error by a substantial margin. Estimates of the various uncertainties complicating the determination of plutonium in lungs have been given recently (Sw76), and the propagated errors from all sources affecting calibration amounted to ±80%, for 95% confidence limits. In several cases (Table 7), this also would appear to understate the actual error.

It should however be emphasized that, while values of P in the region of unity would provide an encouraging (although not wholly incontrovertible) indication to a laboratory that its calibration methods were soundly based, a value appreciably different from this ideal would not imply that a laboratory's estimates for plutonium in lungs were necessarily incorrect to the same extent. Some of the factors determining the efficiency of detection will be energy-dependent (ED) and others will be relatively independent of photon energy (EI). The effects of attenuation within the lungs and in the soft tissues of the chest wall are strongly ED. Factors whose influence is largely EI include the proportion of activity concealed from the detectors by bone, or by organs such as the heart which are so thick that virtually no photons, whatever their energy in this spectral region, can penetrate them. If a laboratory's procedures reproduce the effects of the EI factors correctly, but not those of ED factors, and consequently lead to estimates of ^{103}Pd in lungs which are in error (i.e. $P \neq 1$) then the errors in its estimates of plutonium in lungs, by measurements of X-rays which are more severely attenuated, would presumably be greater. On the other hand, if a laboratory's value of P reflects the combined or net effect of incorrect assumptions regarding both ED and EI factors, the error in its estimates of plutonium in lungs could be either larger or smaller, depending on the relative sizes of the two types of error and on whether they acted in the same or in opposite directions. Thus, it is conceivable, if somewhat unlikely, that some of the laboratories in Table 7 would produce values of P close to 1 if the experiment could be repeated using plutonium, through a fortuitous combination of errors which were in balance for the X-rays from ^{239}Pu but not for the emissions of ^{103}Pd. Conversely, values of P close to 1 for ^{103}Pd do not guarantee absolutely that a laboratory's calibration methods are valid at lower energies.

Thus, the data of Table 7 are of limited relevance in the evaluation of calibration methods for plutonium in lungs, and it would obviously be useful to perform an exercise of this kind employing an X-ray emitter more similar to ^{239}Pu in its energy spectrum, if a suitable nuclide were available. Irrespective of these reservations however, our survey has shown that laboratories can differ by an order of magnitude in their assessments of lung burdens, and this in itself must be a cause for concern.

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REFERENCES

An74 Anderson A. L., 1974, Lawrence Livermore Laboratory, Personal communication.

An76 Anderson A. L. and Campbell G. W., 1976, LLL Plutonium Lung Counter Calibration and Discussion of Errors, Battelle Northwest Laboratory Report, BNWL-2088, pp. 133–149.

Ba71 Bates D. V., Macklem P. T. and Christie R. V., 1971, *Respiratory Function in Disease*, p. 93 (Philadelphia: Saunders).

Boo67 Booker D. V., Chamberlain A. C., Rundo J., Muir D. C. F. and Thomson M. L., 1967, "Elimination of 5-μ Particles from the Human Lung", *Nature* **215**, 30–33.

Bou67 Boulay P., Chivot J. J., Jeanmaire L. and Lansiart A., 1967, "Le protactinium-233, simulateur de plutonium, pour la mesure directe d'une contamination pulmonaire", *C. R. Lebd. Séanc. Acad. Sci., Paris* **265D**, 1571–1574.

Bou68 Boulay P., 1968, Mise au point d'une méthode directe de détermination de la charge corporelle en plutonium-239 chez l'homme par détection X de l'uranium-235, Rapport du Commissariat à l'Energie Atomique, CEA-R-3559.

Bu69 Bukovitz A. G., Sayeg J. A., Spritzer A. A. and Brodsky A., 1969, "Effective Transmission of the Human Thorax for Photons from ^{239}Pu, ^{241}Am and Other Low-Energy Emitters", *Health Phys.* **17**, 71–75.

Bu70 Bukovitz A. G. and Brodsky A., 1970, Reply to De70, *Health Phys.* **19**, 585.

Cl73a Clemente G. F., 1973, "*In Vivo* Measurement of ^{239}Pu in Man", *Proc. 2nd European Conf. Radiation Protection*, pp. 503–507 (Budapest: Akademiai Kiado).

Cl73b Clemente G. F., 1973, "A Comparison of Calibration Methods for the Assessment of ^{239}Pu Lung Burdens", *Proc. Regional Conf. Radiation Protection*, pp. 805–819 (Yavne: IAEC).

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

De70 Dean P. N., Ramsden D. and Rundo J., 1970, Comment on Bu69, *Health Phys.* **19**, 312.

De73 Dean P. N., 1973, "Estimation of Chest Wall Thickness in Lung Counting for Plutonium", *Health Phys.* **24**, 439.

Eh64 Ehret R., Kiefer H., Maushart R. and Möhrle G., 1964, "Performance of an Arrangement of Several Large-Area Proportional Counters for the Assessment of ^{239}Pu Lung Burdens", in: *Assessment of Radioactivity in Man*, Vol. 1, pp. 141–152 (Vienna: IAEA).

Ga77a Garg S. P. and Newton D., 1977, "Reduction of the Low-Energy Background of a Phoswich Detector", *Health Phys.* **32**, 113.

Ga77b Garg S. P., 1977, "Ultrasonic Measurements of Chest Wall Thickness", *Health Phys.* **32**, 34–39.

Ho71 Hounam R. F., Black A. and Walsh M., 1971, "The Deposition of Aerosol Particles in the Nasopharyngeal Region of the Human Respiratory Tract", *Aerosol Sci.* **2**, 47–61.

In70 International Atomic Energy Agency, 1970, *Directory of Whole-Body Radioactivity Monitors* (Vienna: IAEA).

Int64 International Commission on Radiological Units and Measurements, 1964, *Nat. Bur. Stand. Hand.* **85** p. 4.

Is69 Ishihara T., Iinuma T. A., Tanaka E. and Yashiro S., 1969, "Plutonium Lung Monitor Using a Thin NaI(Tl) Crystal of Large Area", *Health Phys.* **17**, 669–678.

Lan64 Lansiart A. and Morucci J. P., 1964, "Nouveau compteur proportionnel destiné à la détection *in vivo* de traces de plutonium dans les poumons", in: *Assessment of Radioactivity in Man*, Vol. 1, pp. 131–140 (Vienna: IAEA).

Lau68 Laurer G. R. and Eisenbud M., 1968, "*In Vivo* Measurements of Nuclides Emitting Soft Penetrating Radiations", in: *Diagnosis and Treatment of Deposited Radionuclides*, pp. 189–207 (Excerpta Medica Foundation).

Lo51 Loevinger R. and Berman M., 1951, "Efficiency Criteria in Radioactivity Counting", *Nucleonics* **9**, 26.

Ma76 Macias E. S., Phelps M. E., Sarantites D. G. and Meyer R. A., 1976, "Decay of 39-Day ^{103}Ru and 17-Day ^{103}Pd to the Levels of ^{103}Rh", *Phys. Rev.* **C14**, 639–644.

Ne71 Newton D., Rundo J. and Taylor B. T., 1971, "Progress in Instrumentation and Calibration Techniques for the Assessment of Lung Burdens of ^{239}Pu", in: *Radiation Protection Problems Relating to Transuranium Elements*, EUR 4612-d-f-e, pp. 469–482 (Luxembourg: CID).

Ne72 Newton D., Fry F. A., Taylor B. T. and Eagle M. C., 1972, "Factors Affecting the Assessment of ^{239}Pu *In Vivo* by External Count-

1106865

ing Methods", in: *Assessment of Radioactive Contamination in Man*, pp. 83–96 (Vienna: IAEA).

Ne76 Newton D., Fry F. A., Taylor B. T., Eagle M. C. and Sharma R. C., 1976, Interlaboratory Comparison of Techniques for Measuring Lung Burdens of Low-Energy X-Ray Emitters (preliminary report), International Atomic Energy Agency Report, IAEA-R-1260F.

Ne78a References cited by Newton D., Toureau A. E. R., Anderson A. L. and Meyer R. A., 1978, "Relative High-Energy Gamma- and X-Ray Emissions Following the Decay of ^{103}Pd", *Int. J. appl. Radiat. Isotopes*, **29**, 188.

Ne78b Newton D., Taylor B. T. and Anderson A. L., 1978, "X-Ray Counting Efficiencies for Plutonium in Lungs Derived From Studies with Inhaled ^{103}Pd", *Health Phys.* **34**, 573.

Ne78c 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 (final report), International Atomic Energy Agency Report, IAEA-R-1260F (Appendix).

Ra67 Ransden D., Peabody C. O. and Speight R. G., 1967, "The Use of Ultrasonics to Investigate Soft Tissue Thicknesses on the Human Chest", UK Atomic Energy Authority Report, AEEW-R493.

Ra69 Ramsden D., 1969, "The Measurement of ^{239}Pu *In Vivo*", *Health Phys.* **16**, 145–153.

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).

Ru68 Rundo J., Taylor B. T., Booker D. V., Newton D. and Scargill D., 1968, "Attenuation in the Chest Wall of 20-keV X-Rays from an Inhaled Radioactive Aerosol", *Nature* **217**, 642.

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.

Ru73 Rundo J., Eckerman K. F., Toohey R. E. and Essling M. A., 1973, The Measurement of Mock Plutonium *In Vivo*, Argonne National Laboratory Report, ANL-8060, Part II, pp. 228–241.

Sa72 Salmon L. and Booker D. V., 1972, FATAL—a General Purpose Computer Program for Fitting Experimental Data to Any Required Function, UK Atomic Energy Authority Report, AERE-R7129.

Sh72 Sharma R. C., Nilsson I. and Lindgren L., 1972, A Twin Large-Area Proportional Flow Counter for the Assay of Plutonium in Human Lungs, Aktiebolaget Atomenergi Report, AE 463.

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

Sn74 Snyder W. S., Ford M. R., Warner C. G. and Watson S. B., 1974, A Tabulation of Dose Equivalent per Microcurie-Day for Source and Target Organs of an Adult for Various Radionuclides, Oak Ridge National Laboratory Report, ORNL-5000.

Sn75 Snyder W. S., Ford M. R., Warner C. G. and Watson S. B., 1975, A Tabulation of Dose Equivalent per Microcurie-Day for Source and Target Organs of an Adult for Various Radionuclides, Oak Ridge National Laboratory Report, ORNL-5000, Part 2.

Sp64 Speight R. G., Peabody C. O. and Ramsden D., 1964, "An Improved Chest Phantom for Studies of Plutonium and Americium in Human Lungs", in: *Assessment of Radioactivity in Man*, Vol. 1, pp. 115–130 (Vienna: IAEA).

Sw70a Swinth K. L., 1970, Tabulation of Transuranic Nuclide Photon Intensities, Battelle Northwest Laboratory Report, BNWL-SA-1914.

Sw70b Swinth K. L. and Griffin B. I., 1970, "A Developmental Scintillation Counter for Detection of Plutonium *In Vivo*", *Health Phys.* **19**, 543–550.

Sw76 Swinth K. L., editor, 1976, Proceedings of the Workshop on Measurement of Heavy Elements *in vivo*", Battelle Northwest Laboratory Report, BNWL-2088, p. 87.

Ta65 Taylor B. T. and Rundo J., 1965, "A Progress Report on the Measurement of Plutonium *In Vivo*", in: *Radioactivity in Man* (second symposium), pp. 52–64 (Springfield: Thomas).

Ta69 Taylor B. T., 1969, "A Proportional Counter for Low-Level Measurement of ^{239}Pu in Lungs", *Health Phys.* **17**, 59–69.

To70 Tomitani T. and Tanaka E., 1970, "Large Area Proportional Counter for Assessment of Plutonium Lung Burden", *Health Phys.* **18**, 195–206.

To75 Toohey R. E., 1975, Measurements of Attenuation Coefficients for Uranium L X-Rays in Tissue-Equivalent Absorber, Argonne National Laboratory Report, ANL-75-60, Part II, pp. 131–133.

To76 Toohey R. E., 1976, The Use of ^{103}Pd as a

1106866

Calibration Source for the Determination of ^{239}Pu *in vivo*, Battelle Northwest Laboratory Report, BNWL-2088, pp. 53–62.

Wa70 Waite D. A., Anderson B. V. and Bramson P. E., 1970, "A Projection Chest Phantom For *In Vivo* Dosimetry", *Am. ind. Hyg. Ass. J.* **31**, 322–326.

Wh74 White D. R., 1974, The Formulation of Substitute Materials with Predetermined Characteristics of Radiation Absorbtion and Scattering, Ph.D. Thesis, University of London.

Wh76 White D. R., 1976, St. Bartholomew's Hospital, London, Private communication.

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