X-RAY COUNTING EFFICIENCIES FOR PLUTONIUM IN LUNGS, DERIVED FROM STUDIES WITH INHALED PALLADIUM-103

D. NEWTON and B. T. TAYLOR
Environmental and Medical Sciences Division, Atomic Energy Research Establishment, Harwell, Oxon, U.K.

and

A. L. ANDERSON
Lawrence Livermore Laboratory, Livermore, CA 94550

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Abstract—A semi-empirical model of the human thorax has been developed to predict the influence of photon energy on the efficiency of external counting of low-energy X-ray emitters in lungs. The calculations allow for differential attenuation of X-rays according to photon energy both within the lungs and in the chest wall. The model is specifically employed to derive calibration data for estimating accidentally inhaled $^{239}$Pu from known X-ray detection efficiencies for $^{103}$Pd inhaled by volunteers; however, it is also potentially useful in the assessment of other low-energy photon emitters. The influence of anatomical and other factors on the relative detection efficiency, $R$, for $^{103}$Pd and $^{239}$Pu is investigated. The dominant factor in the model is the thickness of the chest wall in the regions viewed by the detectors, but the unknown pattern of distribution of activity within the lungs would be the largest single source of uncertainty in $R$. In general, a uniform distribution is assumed, leading to an error which would probably not exceed 20% unless the actual distributions were grossly uneven. Experimental evidence of the validity of the model in this application is provided, and calibration factors are derived for assessing $^{239}$Pu in lungs with phoswich detectors of 12 cm diameter.

INTRODUCTION

LUNG BURDENS of certain toxic alpha-emitters, such as $^{239}$Pu, can only be assessed directly through the detection of low-energy X-rays. This is most commonly done with phoswich detectors* (Fig. 1) or proportional counters viewing the anterior surfaces of the thorax. Because these X-rays are severely attenuated in escaping from the body, the efficiency of detection, or calibration factor, depends critically on a variety of anatomical and physiological factors, but it is frequently deduced from measurements of phantoms containing the relevant activity which are presumed to possess appropriate X-ray attenuation properties.

An alternative approach, which has been proposed or practised by several laboratories (Bo67, Run68, Ne71, Ne72, Ra72, Sha76), is to administer known amounts of short-lived X-ray-emitting aerosols to volunteers, in order to establish correlations between detection efficiency and parameters such as weight, height and chest wall thickness. Most investigations of this nature (Run68, Ne72, Sha76) have employed the nuclide $^{103}$Pd as a simulator for $^{239}$Pu. The relevant decay properties of the two nuclides

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*Dual NaI(Tl)/CsI scintillators (La68).
are summarized in Table 1. It is immediately obvious that $^{103}\text{Pd}$ is not a perfect simulator for $^{239}\text{Pu}$. While a 20.2 keV component is common to the X-ray spectra of both nuclides, the lower energy lines in the $^{239}\text{Pu}$ spectrum (uranium L X-rays) are not represented in the spectrum from $^{103}\text{Pd}$ (rhodium K X-rays), which also includes a Kβ component at 22.8 keV. Attenuation coefficients in tissue vary rapidly with energy in this range (Table 2); allowance must be made for this variation if measured X-ray detection efficiencies for $^{103}\text{Pd}$ in volunteers are to be used in calculating calibration data for other nuclides emitting only low-energy photons. Specifically, we may require estimates of the ratio $R$, where

$$R = \frac{\text{counts per Rh K X-ray emitted by } ^{103}\text{Pd in lungs}}{\text{counts per U L X-ray emitted by } ^{239}\text{Pu in lungs}}.$$  

The factors that might be expected to influence $R$ include the size and density of the lungs, the pattern of radionuclide distribution within the lungs, and the degree of attenuation in the tissues of the chest wall, consisting of muscle, adipose tissue and bone. Most of this paper is concerned with (i) the evaluation of $R$ through calculations based on a simple semi-empirical model, (ii) the dependence of $R$ on certain anatomical and physiological factors, and (iii) the use of $R$ to derive calibration factors for estimating plutonium in lungs, given data acquired after the inhalation of $^{103}\text{Pd}$ by volunteers. We shall also deal briefly with potential applications of our data and model, in the assessment of low-energy X-ray emitters other than $^{239}\text{Pu}$.
Table 2. Linear attenuation coefficients (cm$^{-1}$) for relevant materials

<table>
<thead>
<tr>
<th>Material</th>
<th>Energy (keV)</th>
<th>Photoelectric $\mu_p$</th>
<th>Compton $\mu_C$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Muscle (Int64)</td>
<td>13.6</td>
<td>1.81</td>
<td>0.17</td>
</tr>
<tr>
<td></td>
<td>17.2</td>
<td>0.85</td>
<td>0.17</td>
</tr>
<tr>
<td></td>
<td>20.2</td>
<td>0.31</td>
<td>0.18</td>
</tr>
<tr>
<td>Adipose tissue (Int75)</td>
<td>22.8</td>
<td>0.35</td>
<td>0.18</td>
</tr>
<tr>
<td></td>
<td>17.2</td>
<td>0.63</td>
<td>0.17</td>
</tr>
<tr>
<td></td>
<td>20.2</td>
<td>0.26</td>
<td>0.17</td>
</tr>
<tr>
<td></td>
<td>22.8</td>
<td>0.17</td>
<td>0.17</td>
</tr>
<tr>
<td>Temex (St61)</td>
<td>13.6</td>
<td>1.58</td>
<td>0.17</td>
</tr>
<tr>
<td></td>
<td>17.2</td>
<td>0.78</td>
<td>0.18</td>
</tr>
<tr>
<td></td>
<td>20.2</td>
<td>0.48</td>
<td>0.18</td>
</tr>
<tr>
<td></td>
<td>22.8</td>
<td>0.33</td>
<td>0.18</td>
</tr>
<tr>
<td>Alderson lung.</td>
<td>13.6</td>
<td>2.57</td>
<td>0.043</td>
</tr>
<tr>
<td>e.g. = 0.27</td>
<td>17.2</td>
<td>0.121</td>
<td>0.045</td>
</tr>
<tr>
<td>(Aid70)</td>
<td>20.2</td>
<td>0.073</td>
<td>0.046</td>
</tr>
<tr>
<td></td>
<td>22.8</td>
<td>0.049</td>
<td>0.047</td>
</tr>
<tr>
<td>Compact bone</td>
<td>13.6</td>
<td>2.10</td>
<td>0.26</td>
</tr>
<tr>
<td>(Wo62)</td>
<td>17.2</td>
<td>10.4</td>
<td>0.28</td>
</tr>
<tr>
<td></td>
<td>20.2</td>
<td>6.39</td>
<td>0.29</td>
</tr>
<tr>
<td></td>
<td>22.8</td>
<td>4.43</td>
<td>0.29</td>
</tr>
</tbody>
</table>

*Based on calculations by D. R. White using published methods (Wh74). Elemental compositions from the references given for each material were assumed.

the unshaded regions represented inactive unit-density material. An additional feature was incorporated in some instances. Within the chest wall, a series of more heavily absorbing regions of variable thickness was assumed to be present, reproducing crudely the configuration of the sternum and ribs.

In most instances the detector was assumed to be mounted in a horizontal plane above the supine thorax (Fig. 2), and a uniform distribution of activity within the lungs was envisaged. The sensitive area of the detector window was divided into a number of regions, typically 20 for a counter of 12 cm diameter, and a three-dimensional array of approximately 2000 points, separated by $\sim$2.5 cm horizontally and $\sim$0.2 cm vertically, was set up inside each lung. From each of energy $E_n$. The detector response $C(E_n)$ for a given chest/lung/detector configuration was calculated in arbitrary units by summing the transmission fraction for each path, weighted inversely according to the square of the total path length as a geometry factor, i.e., we effectively performed a double integration of the response over the detector area and the lung volume. However, in studying the most common systems, in which two detectors are employed (one viewing each lung), the response of each counter to activity in the opposite lung was neglected. Measurements of $^{239}$Pu in a phantom, which were made using the equipment shown in Fig. 1, suggested that the activity in each lung contributes only 5% of the total response recorded by the opposite detector.

The intrinsic efficiency of scintillation counters was assumed to be independent of the energy of the quantum and of its angle of incidence on the window. For phoswich detectors fitted with thin beryllium windows, this was a legitimate assumption; however, in applying the method to proportional counters, it would be necessary to take into account path lengths and energy-dependent absorption in the filling gas.

**Calculation of R**

Given values of $C(E_n)$ calculated in this way for a particular arrangement of lungs, chest, and detector and for the relevant energies (Table 1), and weighting according to the relative intensities in Table 1, we have

$$R = \frac{4.6}{80} \times \frac{67C(20.2) + 13C(22.8)}{1.67C(13.6) + 2.38C(17.2) + 0.55C(20.2)}$$  \hspace{1cm} (2)

these points, straight lines were constructed to the centre of each element of window area, and for each combination the total path length and also the path lengths within unit-density soft tissue, low-density lung tissue and (where applicable) bone were calculated. Simple exponential attenuation was assumed, with linear attenuation coefficients $\mu_p(E_n)$ for unit-density soft tissue, $\mu_L(E_n)$ for lung tissue, and $\mu_b(E_n)$ for bone at a particular

**Selection of attenuation coefficients**

In the model we assume simple exponential attenuation of X-rays in passing through all tissues. For bone, this is entirely legitimate, since throughout the 13–23 keV range, Compton scattering is an unimportant process (Table 2). For soft tissues, photoelectric absorption is still the dominant process at energies below 25 keV, but Compton scattering increases in relative importance as the photon
energy is raised from 13.6 to 22.8 keV. Some of the scattered quanta will escape from the body and contribute to the response of the detector. However, this is not a major complication. Calculations by Monte Carlo methods (Rue75) have predicted that, for a uniform distribution of activity in the lungs and a wide range of chest wall thicknesses, the ratio of total to uncollided X-ray flux emerging from the body should be very similar for 103Pd and 239Pu. Hence, in deriving values of $C(E_n)$ to evaluate $R$ [equation (2)], use of the total attenuation coefficient $\mu_p + \mu_C$ (for photoelectric and Compton interactions respectively), would appear to be justified.

**Experimental verification**

It was necessary to confirm that our approach to the evaluation of $R$ allowed for the differences in (i) self-absorption in the lungs for X-rays of 103Pd and 239Pu, (ii) their penetration through bone, and (iii) their transmission through the soft tissues of the chest wall. We were able to do this using data obtained with the equipment shown in Fig. 1, in support of the monitoring program at Lawrence Livermore Laboratory. In this program, calibration for the assessment of low-energy photon emitters involves the use of an Alderson Remab phantom thorax, which incorporates a human rib cage and lungs made from a low-density, nominally tissue-equivalent material (Table 2). This is uniformly impregnated with a known amount of the nuclide concerned (An76). From data obtained in the course of these calibration procedures, which have been followed using both 103Pd- and 239Pu-impregnated lungs, we have deduced a value for $R$, according to equation (1), for the Alderson lungs encased in the rib cage but with no overlying material. This value is 1.31. Calculations based on our model, employing appropriate values for lung dimensions, rib thicknesses, and attenuation coefficients $\mu_p + \mu_C$ in the relevant materials (Table 2), led to a value $R = 1.39$ from equation (2). This acceptable agreement with observation would appear to confirm that the combined effects of (i) and (ii) above can be predicted satisfactorily for a known pattern of deposition. Similar calculations, made with attenuation in the rib cage neglected, gave $R = 1.28$; i.e., of the two effects, (i) has the dominant influence.

The success of the model in predicting (iii), the relative transmission through the chest wall, may be deduced from the attenuation curves shown in Fig. 3, which were obtained with 103Pd and 239Pu, each uniformly distributed in a pair of Alderson "lungs." For each pair of lungs, the response was measured at a distance of 5 cm using the detectors shown in Fig. 1, first without overlying material and then with increasing thicknesses of absorber interposed between the lungs and the detectors. For the 239Pu, curves for both Temex unit-density tissue substitute and beefsteak were obtained, with virtually identical results (Fig. 3); with the 103Pd-impregnated lungs, only beefsteak was used. To simulate this experimental arrangement more closely, the model (Fig. 2) was modified so that attenuation of quanta escaping from the lungs was assumed to occur not in a curved shell embodying "bone" to represent the chest wall, but in a homogeneous overlying slab. Otherwise the calculations were performed as has been described above, employing appropriate values for the relevant physical dimensions and using the assumed coefficients for Alderson lung material and for Temex given in Table 2. From the results, attenuation curves were predicted and these are included in Fig. 3. The model, employing appropriate values for lung dimensions, rib thicknesses, and attenuation coefficients $\mu_p + \mu_C$ in the relevant materials (Table 2), led to a value $R = 1.39$ from equation (2). This acceptable agreement with observation would appear to confirm that the combined effects of (i) and (ii) above can be predicted satisfactorily for a known pattern of deposition. Similar calculations, made with attenuation in the rib cage neglected, gave $R = 1.28$; i.e., of the two effects, (i) has the dominant influence.

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Response as a function of attenuation coefficient for the chest wall, $\mu_w$

For a given lung/chest/detector configuration, the calculated detector response $C$ will be a function of the selected coefficients $\mu_w$, $\mu_L$, and $\mu_B$, for the three types of tissue envisaged in the model. To facilitate presentation of the results, we shall assume initially that the chest wall consists of homogeneous soft tissue (without a bone structure), whose elemental composition is identical with that of the lungs; i.e., $\mu_L = 0.3\mu_w$, where 0.3 is the assumed specific gravity $\rho$ of lung tissue. These simplifications enable us to express the response $C$ in terms of the single attenuation coefficient $\mu_w$. This has been done in Fig. 4 for eight hypothetical subjects characterized by the stated values of overall chest thickness (i.e., $2Z_c$ in Fig. 2), chest width ($2Y_c$), and chest wall thickness ($t$). The detectors were assumed to be scintillation counters of 12 cm diameter, which were displaced to the left and right of the midline of the chest to simulate approximately the arrangement shown in Fig. 1. The results should be more generally applicable, however: curves calculated with other assumptions (larger detectors of up to 20 cm diameter and/or detectors with modified orientation relative to the thorax) were similar in shape to those in Fig. 4.

Convenient functions of the form

$$C = \sum_{i=1}^{4} a_i \exp (-b_i \mu_w)$$

(3)

were found empirically to represent each of the curves in Fig. 4, with an accuracy of better than 0.5% in the range $0.3 \leq \mu_w \leq 2.0$. They may be evaluated from the coefficients $a_i$ and $b_i$ given in Table 4.

From these relations, the values of $C(En)$ appropriate to the values of $\mu_w^P(En) + \mu_w^C(En)$ for muscle (Table 2) were derived, and using equation (2), the estimates of $R(t)$, shown by the solid curve in Fig. 5, were obtained. We also show (Table 5) the relative contributions from the three major components in the spectrum of uranium L X-rays transmitted through the chest wall: i.e., the individual terms $1.67C(13.6)$, $2.38C(17.2)$, and $0.55C(20.2)$ in equation (2), expressed as a percentage of their sum. Even small values of $t$ attenuate the 13.6 keV pho-

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**Table 3. Calculated and measured values of the relative transmission of X-rays from distributed sources of $^{103Pd}$ and $^{239Pu}$ through layers of absorber**

<table>
<thead>
<tr>
<th>Thickness of overlying absorber (cm)</th>
<th>Relative transmission $^{103Pd}/^{239Pu}$</th>
<th>Calculated</th>
<th>Observed</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>(1.00)</td>
<td>(1.00)</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>1.66</td>
<td>1.62</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>2.36</td>
<td>2.32</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>3.69</td>
<td>3.66</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>3.89</td>
<td>3.87</td>
<td></td>
</tr>
</tbody>
</table>

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**Table 3. Calculated and measured values of the relative transmission of X-rays from distributed sources of $^{103Pd}$ and $^{239Pu}$ through layers of absorber**

**Fig. 3.** Observed and calculated transmission of X-rays from distributed sources of $^{239Pu}$ and $^{103Pd}$ through Temex and beefsteak. ○, observed in beefsteak; ×, observed in Temex; −−−−−, calculated using $\mu_w^P(En)$ only; −−−−, calculated using $\mu_w^P(En) + \mu_w^C(En)$.

between the calculated curve using values of $\mu_w^P(En)$ only and that derived using $\mu_w^P(En) + \mu_w^C(En)$, although they are noticeably closer to the latter. However, the curves based on either set of coefficients lead to virtually identical predictions of the ratios of X-ray transmissions for the two nuclides, and the agreement with observation, illustrated in Table 3 for the calculations employing $\mu_w^P + \mu_w^C$, is very close in the relevant region $t \leq 4$ cm.
Effect of variables other than $t$

The use of Fig. 5 (solid curve) to predict the value of $R$ in a particular situation involves assumptions about the influence of other variables, and it is necessary to examine the validity of these assumptions more closely.

(i) Lung density. To investigate how critical factors governing the ratio $R$ is the relative transmission for photons of 17.2 and 20.2 keV.

![Table 4. Values of the coefficients $a_i$ and $b_i$ in equation (3)](image)

<table>
<thead>
<tr>
<th>Chest wall thickness (cm)</th>
<th>$a_1$</th>
<th>$b_1$</th>
<th>$a_2$</th>
<th>$b_2$</th>
<th>$a_3$</th>
<th>$b_3$</th>
<th>$a_4$</th>
<th>$b_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>82.89</td>
<td>3.113</td>
<td>95.93</td>
<td>1.275</td>
<td>1.997</td>
<td>0.108</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>1.0</td>
<td>76.18</td>
<td>3.777</td>
<td>70.83</td>
<td>1.982</td>
<td>2.009</td>
<td>0.754</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>1.5</td>
<td>70.58</td>
<td>4.430</td>
<td>49.42</td>
<td>2.500</td>
<td>4.639</td>
<td>1.644</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>2.0</td>
<td>56.38</td>
<td>4.993</td>
<td>34.29</td>
<td>3.307</td>
<td>10.94</td>
<td>2.990</td>
<td>4.705</td>
<td>2.173</td>
</tr>
<tr>
<td>2.5</td>
<td>45.51</td>
<td>6.209</td>
<td>20.52</td>
<td>4.213</td>
<td>19.99</td>
<td>2.773</td>
<td>6.176</td>
<td>2.817</td>
</tr>
<tr>
<td>3.0</td>
<td>41.06</td>
<td>6.975</td>
<td>32.62</td>
<td>4.364</td>
<td>6.247</td>
<td>3.381</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>3.5</td>
<td>32.81</td>
<td>7.615</td>
<td>27.80</td>
<td>5.247</td>
<td>6.540</td>
<td>3.970</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>4.0</td>
<td>26.31</td>
<td>8.754</td>
<td>26.72</td>
<td>6.021</td>
<td>6.801</td>
<td>4.561</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>
The thickness of human rib cortex is estimated to be \( \sim 1 \text{ mm} \) (In75), so that the thickness of the trabeculae is neglected, the total thickness of compact bone in the path of mental compositions of lung and muscle tissues are believed to be very similar (In75), this approach should be valid for a chest wall composed largely or entirely of muscle tissue. In practice, adipose tissue will, of course, also be present. To investigate the effect on \( R \), the calculations were repeated with values of \( \mu_L(En) \) unchanged but with \( \mu_W(En) \) set equal to various weighted means of the appropriate coefficients in Table 2, simulating a chest wall composed of muscle and adipose tissue in the proportions (by volume) 90/10, 78/22 and 66/34. These values represented the smallest, mean and largest ratios found in a study of 18 male cadavers (Do73). The resulting estimates of \( R \) are presented in Fig. 6 as fractions of \( R \) for the same chest wall thickness composed entirely of muscle. If, in the absence of specific information on the proportions of these tissues, a correction based on the middle curve in Fig. 6 were applied, the result would rarely be in error by as much as 5%.

(ii) Thickness of active lung viewed by detector. For any given value of \( t \) in the range 1–4 cm, and for \( \rho = 0.3 \), doubling the uniform depth of active lung from 5 to 10 cm increased \( R \) by only 4%. A further doubling to 20 cm produced an additional increase of only 2%. This is an important result since the thickness of lung viewed by a detector may vary considerably according to the position of the detector on the thorax.

(iii) Cross-section of the chest. In calculating the results shown in Fig. 4, it was assumed that the thorax could be represented by a cylinder of elliptical cross-section with eccentricity, \( e = 1.5 \). A series of calculations in which \( Y_c \) and \( t \) (Fig. 2) were fixed at 12 and 2.5 cm, respectively, with \( Y_c \) varying from 15 to 21 cm, showed that the effect on \( R \) was negligible: \( R \) increased from 4.27 for \( e = 1.25 \), to 4.32 for \( e = 1.5 \), and further to 4.36 for \( e = 1.75 \)—a total change of only 2%.

(iv) Volume of unit-density tissue separating the lungs. The predictions of Figs. 4 and 5 were obtained for \( W_L = W_R = 2 \text{ cm} \). The calculations for \( t = 2.5 \text{ cm} \) were repeated employing values in the range \( 0 \leq W_L = W_R \leq 8 \text{ cm} \) and produced a maximum change in \( R \) of 2% between these limits.

(v) Composition of soft tissues in the chest wall. The curves in Figs. 4 and 5 are based on a fixed (i.e. energy-independent) relationship between \( \mu_L \) and \( \mu_W \). Given that the elementary compositions of lung and muscle tissues are believed to be very similar (In75), the calculations were repeated with the alternative assumptions \( \mu_L = 0.2 \mu_W \) and \( \mu_L = 0.4 \mu_W \). This led to a series of curves relating \( C \) and \( \mu \) which were displaced in magnitude from those of Fig. 4, but which were similar in shape, so that the relationship between \( R \) and \( t \) was not materially affected (Fig. 5).

Table 5. Spectral composition of transmitted X-rays from uniformly distributed \(^{239}\text{Pu} \) in lungs.

<table>
<thead>
<tr>
<th>Chest wall thickness, ( t ) (cm)</th>
<th>% total transmitted</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>13.6 keV</td>
</tr>
<tr>
<td>0*</td>
<td>36*</td>
</tr>
<tr>
<td>1.0</td>
<td>7</td>
</tr>
<tr>
<td>2.5</td>
<td>1</td>
</tr>
<tr>
<td>4.0</td>
<td>&lt;0.2</td>
</tr>
</tbody>
</table>

*These are the values for a bare source, from the intensities given in Table 1.
Application to the assessment of plutonium in lungs

In Table 7 we show the observed efficiencies, $E_{\text{pd}}$, for detecting known amounts of $^{103}\text{Pd}$ in the lungs of three volunteers whose X-ray emissions were recorded with the 12 cm diameter detectors at Livermore (Fig. 1). Values of $E_{\text{pd}}$ in these subjects, recorded at Harwell with an otherwise similar arrangement of 19 cm diameter detectors, were on average ~50% higher. Estimates of the corresponding efficiencies of the smaller detectors for $^{239}\text{Pu}$, $E_{\text{pu}}$, have been derived from the tabulated values of $E_{\text{pd}}$ and $R$ and are also given in Table 7.

Calibration factors obtained by these methods were employed to assess $^{238}\text{Pu}$ in vivo after a case of accidental inhalation. The X-ray emission from the subject’s chest was investigated with (i) phoswich detectors of 12 cm diameter, as shown in Fig. 1, (ii) a similar arrangement of detectors of 19 cm diameter and (iii) a single detector of 19 cm diameter in a central position above the sternum of the supine subject. To convert the X-ray count-rates into estimates of lung burden, values of $E_{\text{pu}}$ appropriate to each arrangement were derived, as in Table 7, from our investigations of $^{103}\text{Pd}$ in subject JR, whose chest wall thickness was very similar to that of the contaminated man. An adjustment was made to allow for the X-ray abundance of $^{239}\text{Pu}$ (11.4%), which is larger than that of $^{238}\text{Pu}$ (4.6%). The contamination contained a small proportion of $^{241}\text{Am}$ (0.69% by activity, assessed from early fecal excretion) which could be estimated in vivo through measurement of its 60-keV gamma radiation. From our estimate of the $^{241}\text{Am}$ initially deposited in the nonciliated regions (derived by extrapolation of serial estimates
Fig. 7. Variation of the ratio $R$ [equations (1) and (2)] with thickness of compact bone covering 45% of the frontal area of the thorax, for chest wall thickness $t = 2.5$ cm. The results are shown both as calculated (scale on left) and as a fraction of the value for zero bone thickness (scale on right).

commencing after 7 days) and the known composition of the mixture, we deduced a corresponding estimate for $^{238}$Pu in the lungs which was independent of the values based on its 17 keV emission. Expressed as percentages of this independently known value, the three direct estimates of $^{238}$Pu by X-ray counting were (i) 110 ± 28%, (ii) 96 ± 24%, and (iii) 116 ± 29%, with the quoted errors largely reflecting uncertainties in the subject's estimated $^{241}$Am content. It was shown earlier that our calculations successfully predicted the relative transmission of X-rays from $^{103}$Pd and $^{239}$Pu for experimental situations. These additional data suggest that the method can be used satisfactorily in operational assessments of plutonium in vivo.

**DISCUSSION**

**Errors in $R$ and in derived calibration factors for plutonium in lungs**

We do not envisage administering $^{103}$Pd to a contaminated subject in order to improve the assessment of his plutonium content. In practice, a calibration factor would be

<table>
<thead>
<tr>
<th>Subject</th>
<th>Chest wall thickness, $t$ (cm)</th>
<th>$R$</th>
<th>$E_{Pd}$ observed (counts/phon)</th>
<th>$E_{Pd}$ derived (cpm/$\mu$Ci $^{239}$Pu)</th>
<th>$E_{Pd}$ lung burden equivalent to $E_{Pd}$ (cpm/$\mu$Ci $^{239}$Pu)</th>
</tr>
</thead>
<tbody>
<tr>
<td>DN</td>
<td>1.7</td>
<td>3.4</td>
<td>$3.10 \times 10^{-3}$</td>
<td>84</td>
<td>0.004</td>
</tr>
<tr>
<td>JR</td>
<td>2.9</td>
<td>4.7</td>
<td>$1.74 \times 10^{-3}$</td>
<td>40</td>
<td>0.009</td>
</tr>
<tr>
<td>KFB</td>
<td>2.9</td>
<td>4.7</td>
<td>$1.47 \times 10^{-3}$</td>
<td>29</td>
<td>0.012</td>
</tr>
</tbody>
</table>

*Measured at Lawrence Livermore Laboratory by published methods (Ca76).
†Calculated from Figs. 5, 6, and 7 for (i) the measured values of $t$, (ii) a chest wall composed of 22% adipose tissue and 78% muscle, and (iii) an assumed cortical rib bone thickness of 2 mm.
‡Pulled recorded X-ray peak area.
§Counting rate in the range 13–24 keV, including ~ 90% of the full X-ray peak area.

$E_{Pd}$ = standard deviation of the background counting rate, measured for 1 hr. This background is $7.6 \text{min}^{-1}$ for 13–24 keV, including an assumed contribution of 4.0 $\text{min}^{-1}$ from the subject's $^{40}$K and 1976 levels of fall-out $^{137}$Cs.

†Details, may be found elsewhere (An76).
derived indirectly, e.g., by reference to a correlation between chest wall thickness, or chest circumference, and detection efficiencies for $^{103}$Pd administered to volunteers. The estimated value of $R$ would then be applied to the predicted efficiency, $E_{pd}$, to give the calibration factor for plutonium, $E_{pu}$. Such correlations show an appreciable scatter of observed values of $E_{pd}$ about the best-fitting function; e.g., the results of Rundo et al. (Run68) suggest a scatter of $\pm 30\%$ ($2\sigma$). In assessing $E_{pu}$ one would also need to bear in mind the possibility of additional error from the uncertainties in predicting $R$.

Of the anatomical variables we have examined, the chest wall thickness, $t$, in the regions viewed by the detectors is evidently the most important factor affecting $R$. From experience at our two laboratories and elsewhere (Tom76), we estimate uncertainties ($2\sigma$) in determining $t$ by ultrasonic methods to be in the range of 5-10%. The higher value would imply typically a 6% uncertainty in $R$, determined from Fig. 5. If the intermediate ($22\%$) curve of Fig. 6 were used to correct for the presence of adipose tissue, errors would occur in its application to leaner or more obese subjects, leading to an additional uncertainty in $R$ of perhaps 5%. The effects of the other anatomical variables we have studied, including the thickness of bone in the rib cage (Fig. 7), are, taken together, probably similar, suggesting a combined uncertainty approaching 10%. Anatomical factors are not the only important variables, however, and in making this assessment, we assume that the distribution of activity within the lungs is uniform, or essentially uniform, both in the volunteers for whom the data on $E_{pd}$ were obtained and in the subject containing plutonium. In fact, next to the chest wall thickness, the distribution pattern appears to be the most important factor governing $R$ (Table 6), and because it is unknown, its influence would appear to constitute a major source of additional error.

When $^{103}$Pd has been used in calibration studies, the activity has been incorporated into polystyrene particles of size 1-5 $\mu$m and has in general been administered to subjects possessing normal respiratory function and breathing with a tidal volume in the range 0.5-0.81 (Run68, Ne72, Sha76). From studies with gamma-cameras (Sho75) in normal subjects who had inhaled $^{99m}$Tc-labeled 5-$\mu$m polystyrene microspheres under these conditions, it appears that there is a well-dispersed distribution of particles remaining after early rapid clearance from the ciliated regions is essentially complete. One cannot reliably deduce from these observations that the distribution is close to being uniform, but any non-uniformity is certainly not as extreme as in most of the situations considered in Table 6. Similar inferences may be made from reports dealing with the inhalation of other materials, e.g., 2-$\mu$m iron oxide particles (Lo71). Concerning plutonium, findings at autopsy (Vo76) encourage similar conclusions for activity measured by detectors viewing the upper thorax at relatively short times ($\sim 1$ month) after intake. It is impossible to predict how the value of $R$ for typical, or "normal", deposition patterns will differ from the value of $R$ for a hypothetical uniform distribution. However, on the basis of Table 6 and the evidence discussed above, an uncertainty of 10 or perhaps 15% in $R$, as calculated by our methods, would appear to be possible. Combining this assessment with our estimate of $\sim 10\%$ from the anatomical factors would suggest an overall error in $R$ of 15-20%.

It is not clear how such uncertainty in $R$ should be compounded with the comparable estimate of 30% uncertainty in $E_{pd}$ to assess the error in $E_{pu}$, since to some extent $R$ and $E_{pd}$ will be affected by common factors, but an estimate in the range of 35-50% is indicated. For patterns of pulmonary deposition of plutonium which differed markedly from that of the $^{103}$Pd-labeled aerosol in the experimental subjects, larger errors in derived values of $E_{pu}$ would occur. The subject's respiratory pattern during intake of the activity can certainly influence the detection efficiency, and there is some evidence of modified calibration factors in habitual smokers (Ne72); other respiratory abnormalities may well produce similar effects.
Furthermore, it appears that the distribution of plutonium in supposedly normal lungs several years after intake may show peripheral concentrations (Vo76).

**Calibration methods employing phantoms**

The potential errors in estimates of $E_{pu}$ deduced from observations of $E_{pd}$ are evidently substantial. We nevertheless believe that the methods we have described offer a more reliable approach to calibration than is generally achieved by measuring plutonium in currently available phantoms. Of the various factors governing detection efficiency, some are strongly energy dependent (ED) in their effect, while the influence of others is relatively independent of photon energy (EI). For example, the importance of attenuation within the lungs and soft tissues of the chest wall is markedly ED, whereas the effects of screening by bone or by large organs, such as the heart, are essentially EI in the spectral region of interest. By administering $^{103}$Pd to volunteers one can reproduce the EI factors with greater confidence than one can with phantoms, and the remaining problem, with which we have been concerned in this account, is essentially allowing for the effect of the differential attenuation for X-rays of two energies, 17.2 and 20.2 keV. (To judge by the estimates in Table 5, the 13.6 keV component of the U L X-ray spectrum may be neglected.) By contrast, in designing a phantom to predict $E_{pu}$, one must attempt to reproduce correctly the much larger attenuation factors for the emissions of $^{239}$Pu, as well as the EI parameters. Recent indications (Ne78) are that the result may frequently be in error by a factor of three or more.

Minimum detectable activities

Employing the derived values of $E_{pu}$ in Table 7, we have calculated the lung burdens of $^{239}$Pu in each subject that would correspond to the statistical standard error $\sigma$ of the background counting-rate, determined from a measurement lasting 1 hr. These estimates have been included in the table. If correct, they would imply that, for matched subject and background measurements of this duration, the minimum significant measured activity of $^{239}$Pu (equivalent to 2.33$\sigma$, for 95% confidence limits) would exceed the maximum permissible lung burden (0.016 $\mu$Ci) except for the smallest of these subjects.

Other models

Other models have been developed for the specific purpose of deriving calibration factors for plutonium in lungs from observed detection efficiencies for $^{103}$Pd. Cohen, Guilmette and Wrenn (Co73), employing a simpler representation of the lungs and chest that made fewer demands on computer time and storage than ours, have predicted similar values of $R$. An alternative method employs the concept of an “effective tissue thickness” (Run69; To076). When applied to data recorded at Harwell for the three subjects of Table 7, this approach led to values of $R$ between 20 and 38% larger than those predicted by our model.

Extended applications

We found that the estimates of $R(t)$ (Fig. 5) were not materially affected by substantial changes in the assumed size and orientation of the detectors. Thus those given in Table 7 could reasonably be used to calculate esti-
mates of $E_{Pu}$ from values of $E_{Pd}$ recorded for other phoswich arrangements. Values of $E_{Pd}$ have been provided by nine laboratories which investigated the X-ray emissions from the subjects of Table 7, using one or more phoswich detectors of 10-19 cm diameter in various configurations relative to the thorax. These may be found elsewhere (Ne78).

Equation (2) (with suitably modified X-ray abundances) and equation (3) (with the coefficients given in Table 4 and appropriate values of $\mu_w$) may be used to estimate calibration factors, relative to $E_{Pd}$, for other nuclides emitting X-rays comparable in energy to those from $^{239}Pu$, e.g., isotopes of curium (14-21 keV). However, the experimental justification for our approach derives specifically from investigations within the range of 13-23 keV. We would not advocate its uncritical use for extrapolation between situations in which Compton scattering is an unimportant process (e.g., in soft tissue for 15-keV photons) and those in which it is a dominant mechanism (energy $>$ 30 keV). A further caution is necessary. While the shapes of the curves relating $C$ and $\mu_w$ for fixed values of $t$ [equation (3) and Fig. 4] did not depend at all critically on the assumed size and orientation of the detector, their separation was to some extent affected by changes in these assumptions, i.e., the variation of $C$ with $t$ for a given value of $\mu_w$ was somewhat dependent on geometry. Consequently, extrapolation of data on $E_{Pd}$ (Ne78), or of derived values of $E_{Pu}$ (Table 7), to values of $t$ substantially outside the range of observation (1.7-2.9 cm in Table 7) could not reliably be based on equation (3) or Fig. 4. For such extrapolations, it would be better to study experimentally the variation of $E$ with $t$ for the geometry employed, using an anatomically realistic simulation of active "lungs" and overlying "chest wall" composed of approximately tissue-equivalent materials.

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