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A. L. Anderson
G. W. Campbell

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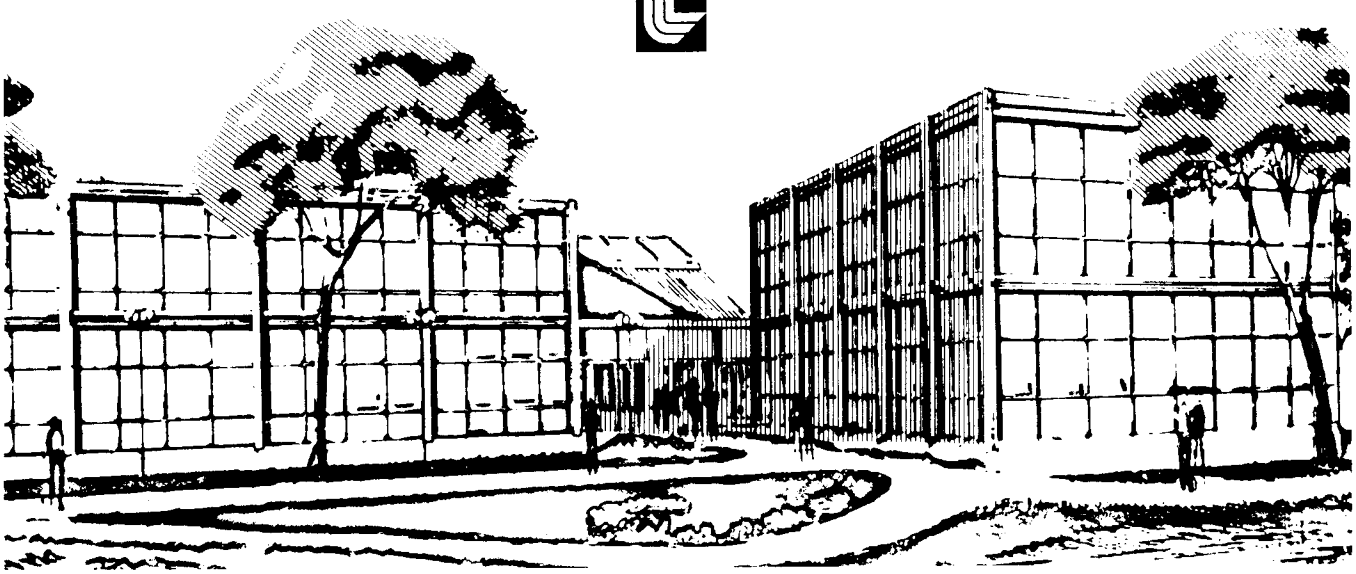

LLL PLUTONIUM LUNG COUNTER CALIBRATION AND DISCUSSION OF ERRORS*

A. L. Anderson and G. W. Campbell
Lawrence Livermore Laboratory
Livermore, California

INTRODUCTION

The Whole Body Counter at Lawrence Livermore Laboratory is located in a shielded room 6.1 metres underground, and is equipped with a filtered air supply to remove approximately 99% of the radioactivity present from randon-thoron daughters in the air. The details of the room construction have been described in previous reports.(1)(2)

Two Harshaw Phoswich detectors 120 mm in diameter are presently used for lung counting. These are placed high on the chest tangent to the sternum and clavicle, as shown in Figure 1. The front face of the detector consists of a 1.6-mm-thick NaI(Tl) scintillation crystal with a 0.25-mm beryllium window, and is used as the x-ray detector. This is backed by a thicker 38.1-mm CsI(Na) crystal coupled to the same photomultiplier tube. This crystal is used for compton background suppression, and also to provide spectral information over the energy range from 100 keV to 2.5 MeV. The detector outputs, using risetime discrimination electronics, can be summed or accumulated separately in a pulse height analyzer. The complete design and operation of the system has been described elsewhere.(3)

An Alderson Remab phantom is used for calibration. This is a take-apart phantom, shown in Figure 2, which has a human skeleton and fillable compartments to simulate the body organs. The lung cavities which are shown in the figure can be filled with lung-equivalent material containing

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radioactive standards for calibration. Several uniformly loaded sets of these lungs have been made incorporating various isotopically pure radionuclides including ^{238}Pu, ^{239}Pu, and ^{241}Am. The phantom has a somewhat "average man" external shape, but contains internally a small Asiatic skeleton. This causes the lungs to be smaller than those of the average American male, and also results in an abnormally large chest-wall thickness for the phantom, which is approximately 45 to 50 mm thick compared with an average American with a 25-28 mm chest wall in the region viewed by the detectors. The lungs themselves are also shaped differently compared with normal human anatomy so that the phantom, although entirely adequate for many purposes for which it was originally intended, is somewhat deficient as a calibration medium for counting plutonium in lungs. It is, however, one of the only nearly realistic phantoms available at the present time. Since the water-filled phantom contains plastic and perhaps other materials which are not truly representative of human tissue, it becomes necessary to determine the equivalent tissue thickness of the material overlying the phantom lungs and then provide a means of extrapolation to humans of differing chest-wall thickness. This is done at Livermore in several steps as follows, and provides the basis for construction of individual lung counter calibration curves for each nuclide of interest.

CALIBRATION PROCEDURE

First, standard lungs are made up containing the desired x-ray emitter and inserted into the water-filled phantom with the rib cage removed. The detectors are then lowered into position over the lungs at contact with the chest as shown in Figure 3, and a count is made. Figure 4 shows an end view of the phantom with the lower section removed. Then in Figure 5 the entire phantom is removed leaving the lungs in open air in the same geometry as if in the phantom, and a second count is made. From these two measurements, the x-ray fraction transmitted through the phantom chest wall can be calculated. This region is equivalent to that measured ultrasonically for the subject whose chest-wall thickness is determined using a "Compound B" scanning technique which has been described previously.[4]

To determine what this corresponds to in terms of human chest-wall thickness, a series of transmission measurements were made using beefsteak and tissue-equivalent temex. This was done by placing the beefsteak or temex absorber directly over the phantom lungs and heart as in Figure 6, for various thicknesses from 0 up to and including 5 cm and with the detector fixed at a constant 5-cm position. The resulting transmission data are plotted in Figure 7 for ^{241}Am, ^{103}Pd, and ^{238}Pu showing the fraction transmitted versus absorber thickness. If the curves are developed using a two-dimensional source or disc source, the transmission curves drop much lower as indicated in the lower curve. The difference between the two curves is presumed to be due to a hardening effect of the x-ray beam as it passes through additional material contained in the heart and lungs of the phantom. This results in a greater transmission of the x rays through the beefsteak layers because of the higher initial effective energy of the x-ray beam. Some slight buildup is also presumed to have occurred in the lungs. The equivalent chest-wall thickness of the phantom is determined by selecting the same fractional transmission as determined previously in the phantom measurement and then reading over on the curve to find the chest-wall thickness in centimetres.

The calibration is completed by replacing the rib cage in the phantom and counting the lungs to get the total sensitivity in counts per minute per microcurie for this particular thickness. This sensitivity and the transmission data obtained earlier for beefsteak, with suitable geometry corrections to place the detectors at contact with the chest wall at each calibration point, are then used to construct the total calibration curves shown in Figures 8 and 9 for plutonium and americium, respectively.

Another method of obtaining the same or similar curves involves the use of transmission curve measurements through the beefsteak where the detectors are always placed in contact with the absorber at each calibration point as shown in Figure 10, rather than at a fixed distance as described previously. The overall calibration curve which results is automatically corrected for the geometry of varying chest-wall thickness, and is perhaps the best approach to calibration since any given subject will always be counted with the detectors placed over his chest at contact with the body surface.

METHOD

Each person is counted at LLL for a total of 4000 seconds with the detectors in full contact with the chest, or as nearly so as possible. A 4000-second detector background, obtained by placing a sack of sugar under each detector, is subtracted from the subject spectra to obtain the net count. A typical background counting rate for two detectors summed in the plutonium band (13 to 24 keV) is 0.06 counts per second. Until recently, the person's normal background count-rate in the 17-keV band and 60-keV band was estimated from background data on "clean" subjects of a similar size, weight and shape, and an approximate "clean person" match was subtracted from the subject's net spectra. If there was a residual present, the plutonium burden was calculated using the calibration curve shown in Figure 8. The "clean person" background is now estimated by integrating a portion of the subject's own spectra in a higher energy band (80-100 keV) and then using this value to calculate a normal "clean person" background in the plutonium band, according to the relationship established in Figure 11 for 62 "clean" individuals of varying body size. If the isotopic composition of the material is different from that used in the calibration, the calculation is modified accordingly.

If ^{241}Am is present in the spectrum, the americium burden is calculated first by subtracting a normal clean person background from the 60-keV region according to Figure 12 for the same 62 "clean" individuals discussed earlier to derive Figure 11 and then by using the 60-keV gamma peak and calibration factor obtained from Figure 9 to assess the americium content.

A fraction of the net count rate at 60 keV is subtracted from the plutonium band as determined from the 17-keV to 60-keV ratio curve shown in Figure 13 which shows the ratio of 17-keV to 60-keV count rates versus chest-wall thickness for a pure ^{241}Am x-ray emitter. This curve was also derived by counting varying thicknesses of beefsteak overlying the Remab lungs. After this subtraction, if there is still a net count rate in the plutonium band, the plutonium burden is calculated from the calibration curve of Figure 8 directly.

All of the above calculations have been incorporated into a BASIC computer program which allows quick and efficient analysis of the data.

ERRORS AND MINIMUM DETECTABLE ACTIVITY

The total error on the count is calculated at the 95% confidence level by taking the square root of the sum of the squares of essentially four major individual errors. These include the statistical error of the person's own count and the "clean person" background estimate for that count, the calibration error associated with the particular calibration curve in use, and the estimated error in terms of percent effect due to uncertainties in the subject's chest-wall thickness, which is approximately ±2 mm. Uniform deposition is assumed in the lungs, and no attempt is normally made during routine counting to correct for differing lung size, rib spacing, tissue composition overlying the lungs, or other parameters which may be at variance with the calibration phantom, although some of these corrections can and should be applied (if known) when lung positives are encountered.

Minimum detectable activities are computed at the 95% confidence level from a modified formula of Altschuler and Pasternak[5] as shown in Equation (1) where the background count rate is not well known.

$$MDA = \frac{2k\left(2\frac{B}{T}\right)^{1/2} + \frac{k^2}{T}}{S} \qquad (1)$$

where

> k = constant determined by confidence level (1.645 for 95%)
> B = background count rate (cpm)
> T = counting time (min)
> S = sensitivity (cpm/μCi)

In this case, the background is assumed to be the detector background plus the estimated "clean person" background for the energy band of interest. For example, in a 4000-second count, the minimum detectable activity of ^{239}Pu present in a typical subject having a 2.5-cm average chest-wall thickness would be 14 nanocuries, assuming a detector background of 3.6 cpm, a normal "clean person" background count rate in the plutonium band of 4 cpm, and a corresponding calibration factor of 116 cpm/μCi. In a similar manner, the MDA for ^{238}Pu would be 7 nCi using the same background but with a calibration factor equal to 250 cpm/μCi. For ^{241}Am, the MDA would be approximately 0.1 nCi, assuming a detector background of 7.6 cpm in the energy band of interest, a net "clean person" background of 25 cpm, and a calibration factor of 41 cpm/nCi.

A series of 34 chest-wall thickness measurements recently made at LLL using Compound B scanning techniques gave an average of 2.8 cm for the 34 individuals involved in the study. This is somewhat thicker than the average chest-wall thickness generally accepted by most laboratories, and perhaps indicates the need for further investigation. However, if the chest-wall thickness of the hypothetical subject in the example given above were just 3 mm thicker, or about 2.8 cm, the minimum detectable activity for isotopically pure ^{239}Pu in the chest would be 19 nCi, or slightly above the 16-nCi maximum permissible lung burden which has been presently established for humans.

Fortunately, ^{239}Pu is usually composed of a mixture of several plutonium isotopes, including ^{241}Pu which decays by beta emission to ^{241}Am. The ^{241}Am emits a 60-keV gamma ray, which is rather easily detected. Thus, if the isotopic composition of the material involved is known or can be measured, the ^{241}Am may be used as a tag to quantitate the amount of plutonium present. This procedure is valid under certain conditions for new exposures where the material has not had sufficient time to translocate within the body. Since the MDA for ^{241}Am in the average person is about 0.1 nCi, this makes the practical MDA for ^{239}Pu in the lung about 2 nCi when a typical weapons-grade mixture of plutonium isotopes containing approximately 1200 ppm americium is involved. It is highly unlikely, however, that this procedure could be used with confidence when dealing with old exposures because of the apparent rapid translocation of americium to other body sites as compared to ^{239}Pu.

DISCUSSION

One disturbing aspect of the Livermore calibration technique is that it does not appear to match the results observed through human experience. In late 1972, 15 laboratories in the United States and other countries participated in an in-vivo calibration experiment under joint IAEA and AERE, Harwell sponsorship involving three volunteer subjects who inhaled ^{103}Pd as a plutonium simulant.(6) The ^{103}Pd decays by electron capture, emitting 20.2-keV rhodium K x rays which are very close in energy to those from ^{239}Pu. The material was administered using a normal breathing pattern as an aerosol containing ^{51}Cr as a tag in a known ratio to the palladium. By counting the 320-keV gamma rays from ^{51}Cr which are more easily detected in the body, it was possible to quantitate the amount of palladium present more accurately than would be possible through normal x-ray counting of the palladium alone. However, the palladium could also be counted directly as a comparison.

In 1972, the Livermore lung counter was calibrated for ^{103}Pd in exactly the same manner as previously described except that the phantom lungs were loaded with palladium instead of plutonium. The resulting calibration is shown by the upper curve in Figure 14. The lower curve is the adjusted calibration curve based on the observed palladium count rate for the three individuals in the study and the known amount of palladium present in the lungs of these individuals at the time of measurement. The difference between the two calibration curves is about 40%. Although the exact reason for the discrepancy is not known, there are four clear possibilities among several which might be considered.

First, the chest-wall thickness for the subjects may be incorrect, requiring an increase of about 3 mm to bring the curve more into agreement.

Second, the palladium count-rates during the in-vivo experiment were shown to be highly geometry-dependent. Simply moving the detectors slightly from one position to another sometimes produced large differences in detector response, and thus may account for some of the observed discrepancy.

Third, there may be some type of x-ray absorption taking place in the body which is not being properly accounted for by the phantom such as an increase in blood volume for the subject in the supine counting position.

Fourth, there may be a non-uniform distribution of activity within the lung which causes the material to be viewed with lower effective efficiency by the detectors. In fact, the observed ^{51}Cr counting data on two of the three individuals in the experiment (the third was not counted posterially) tend to indicate that the deposition of material may have been more toward the back of the body. Previous ^{103}Pd in-vivo studies conducted at Harwell, England[7] have shown at least a 70% effect apparently due entirely to distribution factors within the lung, although in that study an increase in efficiency was observed when comparing the effects of a normal and abnormal breathing pattern.

If we are to assume, however, that the adjusted calibration in Figure 14 is the more correct one for ^{103}Pd and represents the normal situation with humans, then the phantom calibration underestimates the radioactivity content for humans, and appropriate modification of the plutonium calibration curves is necessary. This is inferred in Figure 15 by constructing a new calibration curve, based on the ^{103}Pd in-vivo data. This calibration is approximately 30% different than the previous Remab calibration curve for ^{239}Pu in Figure 8, and being the most conservative is the one presently used for plutonium assay at Livermore. The use of this calibration implies an increased Minimum Detectable Activity limit of several nanocuries in proportion to the adjustment in calibration factor, and is equal to about 19 nCi for ^{239}Pu at 2.5-cm chest-wall thickness, or 26 nCi at 2.8-cm chest-wall thickness. It should be noted, however, that these values refer only to what we have considered to be an average man in terms of human chest-wall thickness, when in fact humans vary widely over a normal range of thicknesses from approximately 1 cm or less to 4.5 cm. Thus, the question of Minimum Detectable Activity of isotopically pure plutonium in the lung is not one which can be answered simply by using a single number. Rather, to put the problem in proper perspective when discussing counter capabilities, MDA's should perhaps be expressed as a range of values, depending upon the person's chest-wall thickness and other factors such as activity distribution which may, but probably will not, be known for any given inhalation exposure history.

In terms of chest-wall thickness alone, these values for the Livermore calibration from 1 to 4.5 cm are from 4 to 106 nanocuries, respectively, with the large majority of people falling in the 2- to 3.5-cm bracket. This latter group would give an equivalent range of MDA's at the 95% confidence level of from 12 to 48 nCi of ^{239}Pu.

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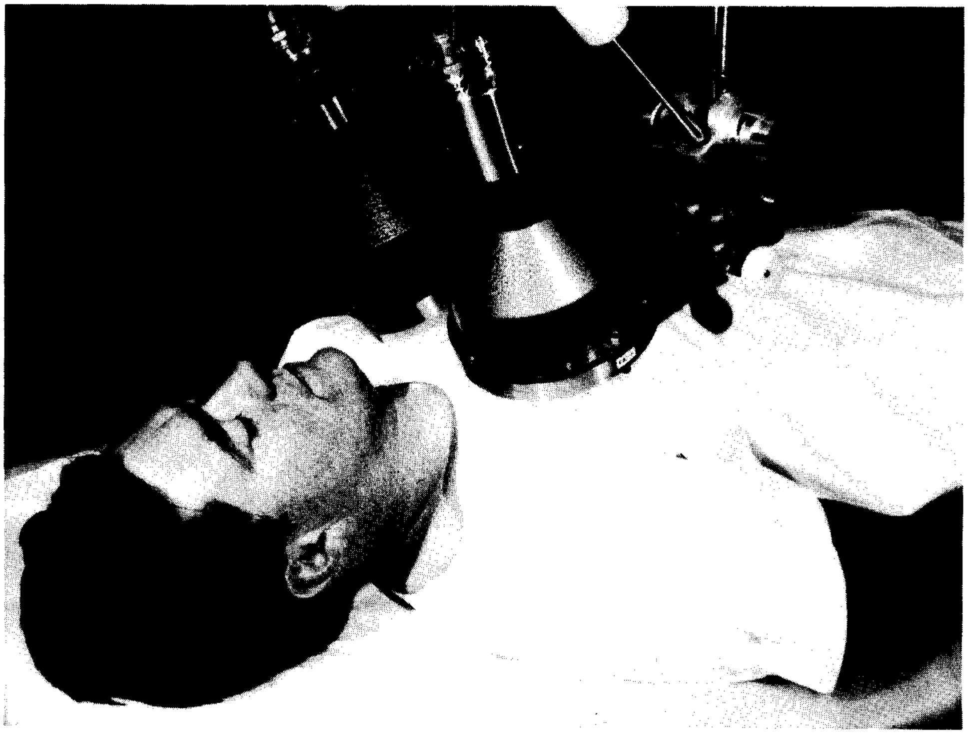

Figure 1. Plutonium lung counter counting arrangement at Lawrence Livermore Laboratory.

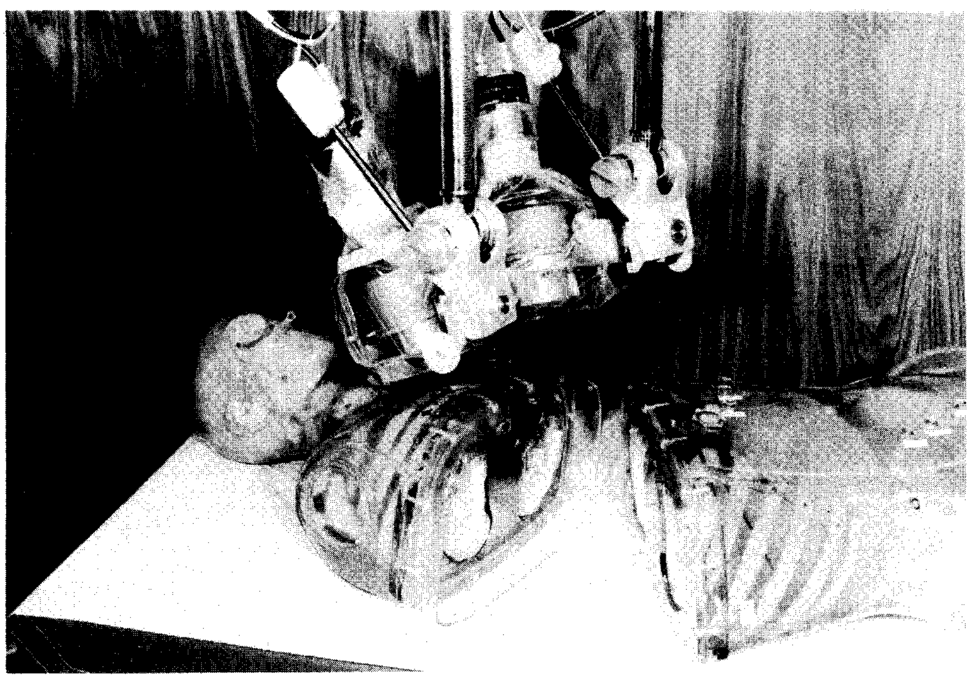

Figure 2. Alderson Remab calibration phantom showing plutonium loaded lungs.

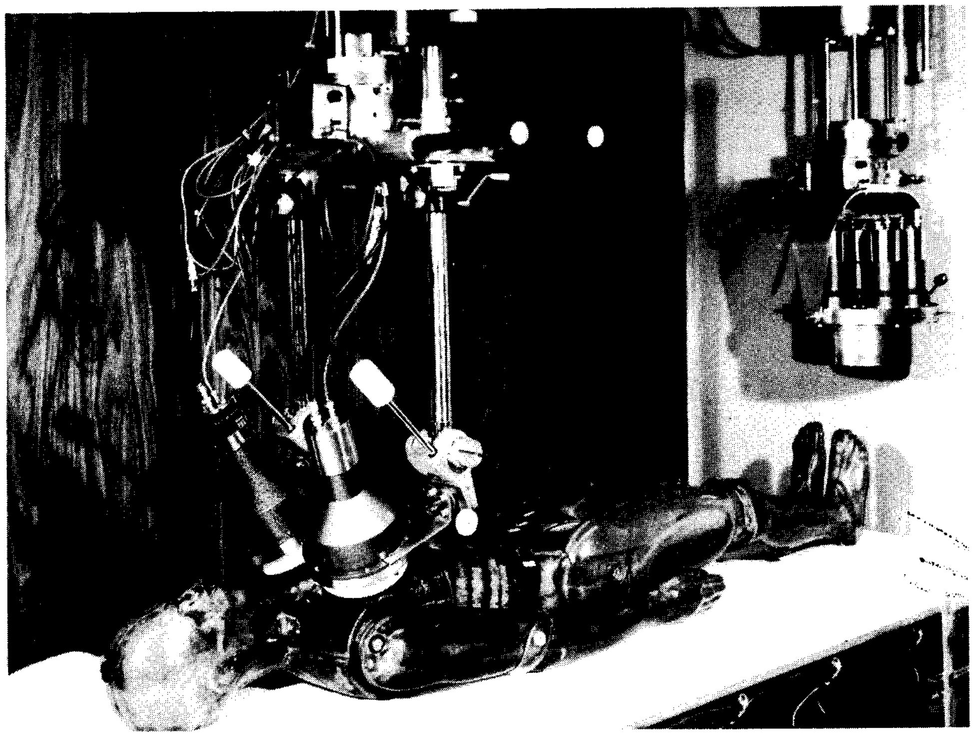

Figure 3. Remab calibration phantom without ribs and with Phoswich detectors in position for counting.

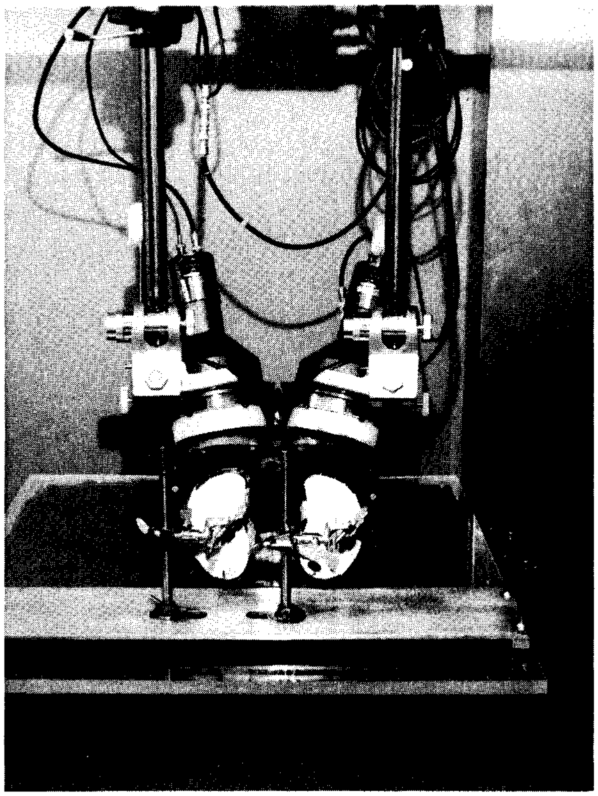

Figure 4. End view of the Remab phantom with the lower section removed.

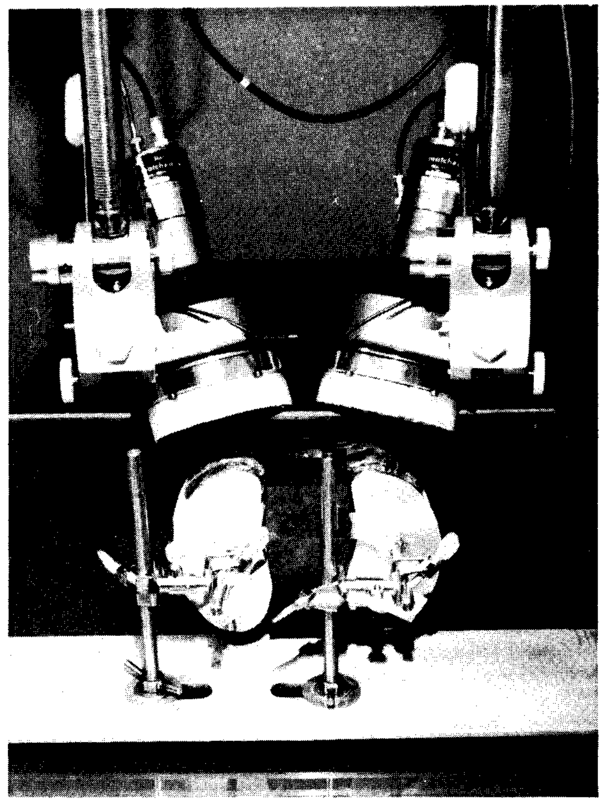

Figure 5. View of Remab lungs in air in same geometry as if in the phantom. Heart in place over left lung with Temex filling between lungs.

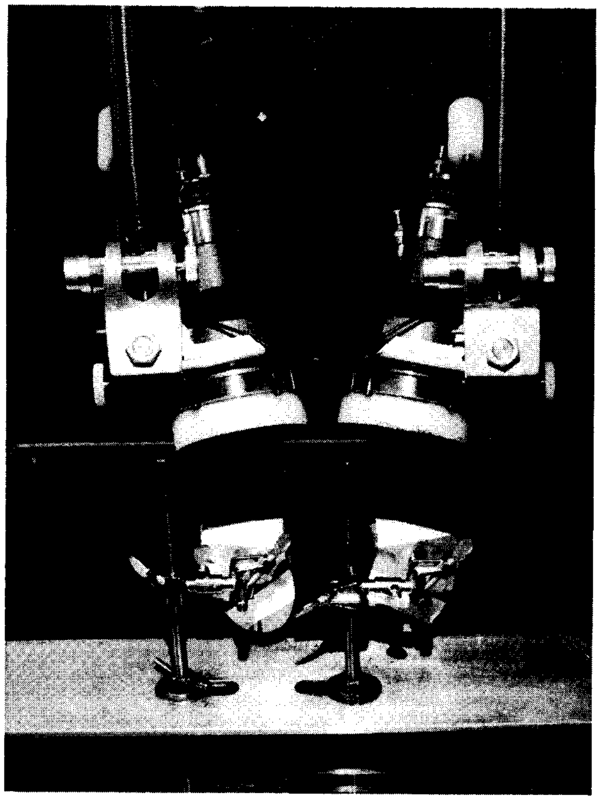

Figure 6. Remab lungs in air as in Fig. 5 with Temex absorber interposed between detectors and lungs.

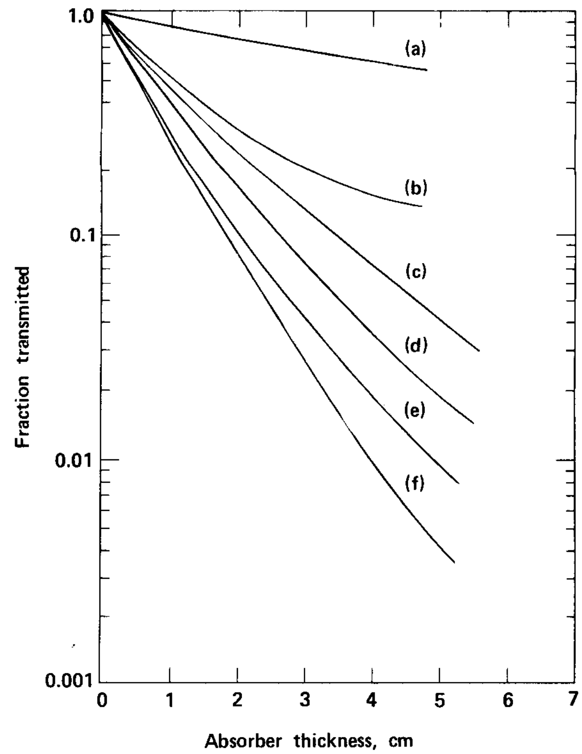

Figure 7. X-ray transmission curves showing X-ray fraction transmitted versus absorber thickness.

(a) ^{241}Am Remab lung plus beefsteak (60 keV)

(b) ^{241}Am Remab lung plus beefsteak (17 keV)

(c) ^{103}Pd Remab lung plus beefsteak

(d) ^{238}Pu Lucite

(e) ^{238}Pu Remab lung plus beefsteak.

(f) ^{238}Pu disc source plus beefsteak.

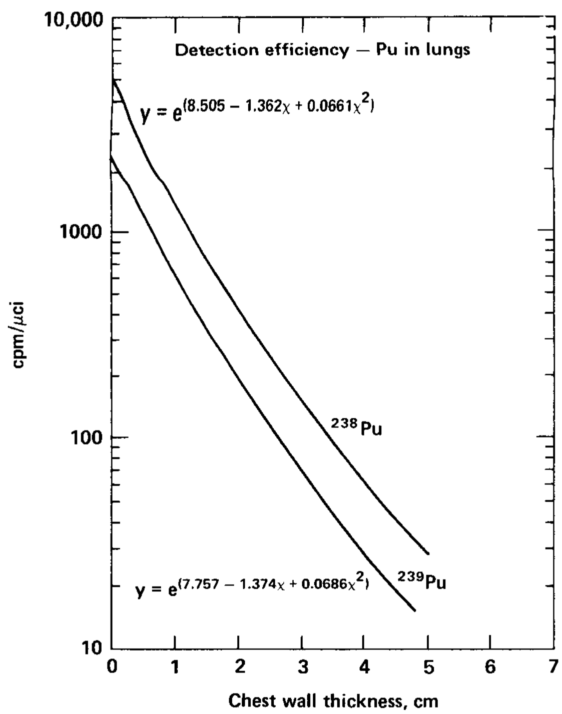

Figure 8. Detection efficiency for ^{238}Pu and ^{239}Pu in lungs versus chest wall thickness.

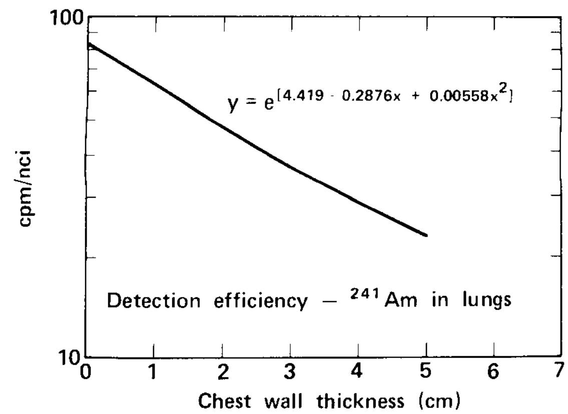

Figure 9. Detection efficiency for ^{241}Am in lungs versus chest wall thickness.

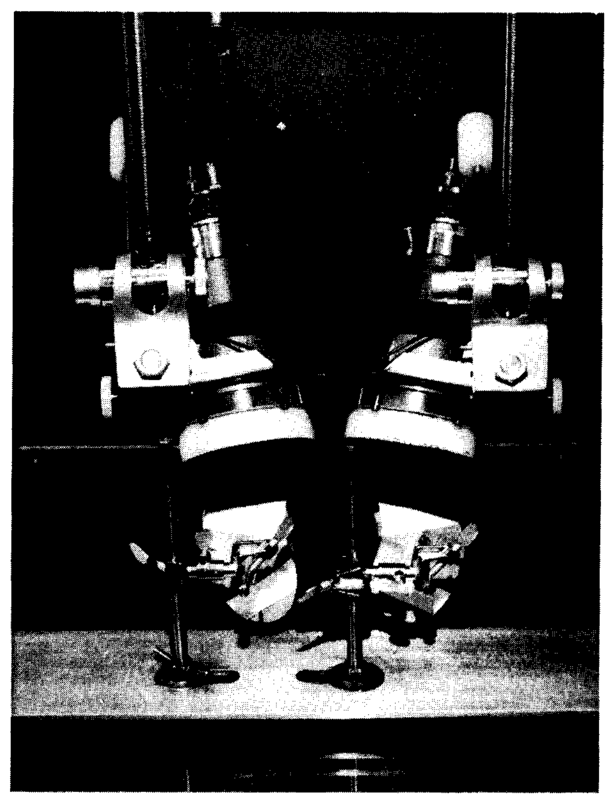

Figure 10. View of Remab lungs with detectors at contact with overlain Temex absorber.

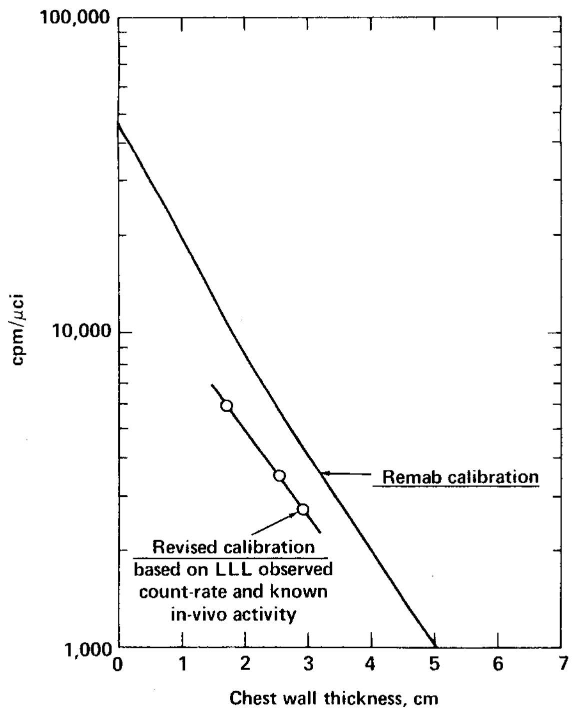

Figure 14. Detection efficiency for ^{103}Pd in lungs derived from Remab phantom calibration and observed in-vivo activity versus chest wall thickness.

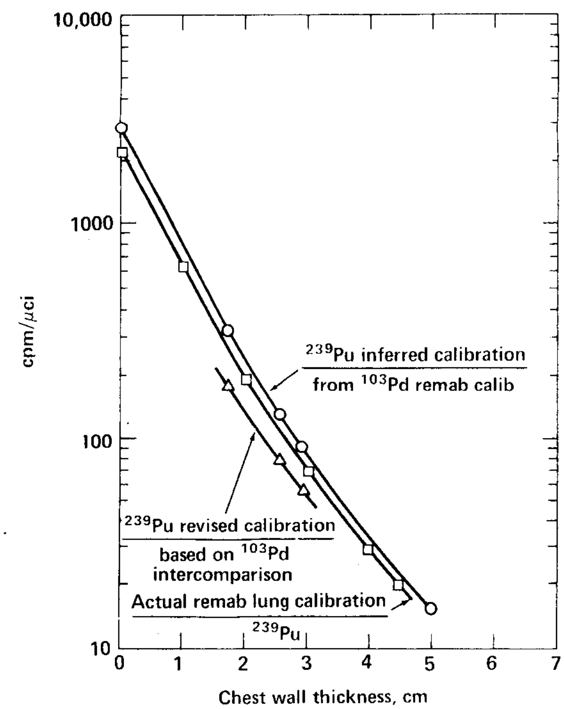

Figure 15. Detection efficiency for ^{239}Pu in lungs derived from ^{239}Pu and ^{103}Pd Remab calibrations and from observed ^{103}Pd in-vivo data.

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