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STATUS AND TRENDS IN THE EXTERNAL COUNTING OF INHALED HEAVY ELEMENTS DEPOSITED *IN VIVO*

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Abstract—External counting has been routinely used for estimation of plutonium *in vivo* for approx. 10 yr. However, this method is fraught with inherent uncertainties resulting from the few radiations and the severe attenuation of the radiations. Present counting capability allows detection of from 1/2 to 1 Maximum Permissible Lung Burden (16 nCi). Current efforts in the development of counters and in intercalibration may lead to small improvements in detection limits and accuracy, but substantial improvements are not expected. Two troublesome areas in the *in vivo* counting area are the non-uniform distribution of material within the lungs and the influence of material translocated from the primary deposition site. New concepts such as induced nuclear fission of the deposited material can possibly lead to improvements in accuracy and in the detection limit.

INTRODUCTION

TECHNIQUES permitting accurate and rapid quantitation of internal deposits of heavy elements such as plutonium are essential in order to protect the health and safety of personnel working with these materials. Direct measurement of heavy elements in excreta has been used as an index of exposure since the days of the Manhattan project; however, the interpretation of bioassay (urine) results is an indirect technique requiring assumptions regarding the metabolism and subsequent excretion rate of deposited material. Results from the Transuranium Registry have indicated that analysis of excreta (urine) does not accurately predict the deposition of inhaled insoluble plutonium (No74). Roesch and Baum (Ro59), first demonstrated the feasibility of direct measurement of plutonium *in vivo* by counting the *L* X-rays from its uranium daughter in an experiment with an anesthetized pig. Direct measurements such as this are generally favored over indirect techniques because of the improved accuracy. This report describes the state-of-the-art of assay of inhaled heavy elements *in vivo* by photon counting, as well as methods of correction for tissue absorption, background, calibration, isotopic composition and tissue distribution. Recent efforts in the field are also described to illustrate trends that may influence the development of such measurements.

The photon radiation from the long-lived α-particle emitting isotopes of plutonium is of very low intensity; the most significant radiations are the 13.6, 17.0 and 20.2 keV *L* X-rays (average energy 17 keV) emitted by

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Table 1. Data for some heavy isotopes

Isotope	Specific activity (Ci/g)	Maximum permissible body burden (μCi)	(μg)	X-rays per alpha
^{238}Pu	16.8	0.04	0.0023	0.115
^{239}Pu	6.16×10^{-2}	0.04	0.65	0.0465
^{240}Pu	0.227	0.04	0.18	0.108
^{241}Pu	113	0.90	0.0083	—
^{242}Pu	3.91×10^{-3}	0.05	13	0.100
^{241}Am	3.22	0.05	0.016	0.376
^{242}Cm	3.21×10^{3}	0.05	1.55×10^{-5}	0.106
^{244}Cm	81.1	0.1	0.00123	0.0923
^{237}Np	7.06×10^{-4}	0.06	85	0.57

the uranium daughters. These X-rays result from internal conversion when excited states of the uranium daughter decay to the ground state; although often referred to as plutonium *L* X-rays, it is important to remember that, in fact, they are intensities (Table 1). Note that even with similar photon intensities, the dominant nuclide by mass in a mixture may not be the dominant nuclide by activity. Most nuclear weapon or reactor grades of plutonium in frequent use are mixtures of various isotopes of plutonium and contain a small but significant amount of ^{241}Am from the beta decay of ^{241}Pu. Americium-241 emits a 60-keV gamma that can be used as a tracer when measuring plutonium. To do this requires both a knowledge of the isotopic composition of the material (ppm of ^{241}Am) and the assumption that americium and plutonium are similarly metabolized by the body.

STATUS OF OPERATIONAL COUNTING

The following sections provide a summary of the present state of operational counting of inhaled heavy elements *in vivo*. The hardware, calibration methods and influencing factors are described.

Counting hardware

Currently, the "phoswich" is the most common type of detector used for assessing lung depositions of low-energy photon emitters. This consists of a thin (e.g. 1–3 mm) crystal of NaI (Tl) backed by, and optically coupled to, a thick (e.g. 50 mm) crystal of CsI (Tl). The combination is commonly viewed by a single photomultiplier tube. The difference in decay times of the light from the two crystals permits the exclusive detection of events which occur in the thin crystal (i.e. low energy quanta such as uranium *L* X-rays) while reducing the response to scattered higher energy photons. This results in very low background at low energies while retaining high detection efficiency. These detectors, with their associated instrumentation, have proved reliable for routine monitoring. Most phoswich detector systems are capable of detecting about one-half the maximum permissible lung burden (MPLB) of ^{238}Pu or a little more than 1 MPLB of ^{239}Pu in an average individual with a uniform distribution of plutonium in the lungs. Reference (Sw76a) describes several phoswich counting systems and discusses their limitations.

Many of the large-area NaI (Tl) detection systems have been replaced by phoswich detectors. The NaI detector is basically similar to the phoswich, but has a higher background and consequently a lower sensitivity at energies (<70 keV) used for assessing inhalation exposures to heavy elements. Table 2 compares some important characteristics of various counting systems.

Some facilities use proportional counters for assessing lung burdens (Ne75; La64; Ra69; Ta69; Tor70). Most of the counters use anti-coincidence techniques to reduce background counts and improve sensitivity. The counting efficiency of proportional counters is lower than that of most other types of detectors and decreases rapidly as photon energy increases. This precludes the use of ^{241}Am (60 keV γ) to assess lung depositions of ^{239}Pu. The detectable limit of proportional counters is generally higher than that of phoswich detectors. One significant advantage of the proportional counter over the scintillation counter is its improved energy resolution, which permits one to distinguish between external and internal contamination.

Semiconductor detectors are only practical when used in arrays and, with the current high cost of intrinsic germanium, they are the most expensive detection system available.

Table 2. Comparison of properties of detectors

Detector	Energy resolution (keV, FWHM at 17 keV)	Background (cpm/cm^2, 12–24 keV)	Relative efficiency*
Large-area scintillator	8	0.125	1.1
Phoswich	8	0.05	1
Proportional counter	2.6	0.0091	0.45
Solid-state detector	0.7	0.02	—

*100 mm from point source of ^{238}Pu.

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These detectors have excellent energy resolution. Only one such system is currently constructed specifically for external counting of inhaled heavy elements (Ty76). This system was installed in 1976 and consists of two arrays of four intrinsic germanium detectors, each of which has an area of 10 cm^2. Preliminary results indicate that backgrounds per unit area are very low and resolution (<700 eV at 60 keV) is far superior to that of other types of detectors. Absolute detection efficiency, as with the scintillation detectors, should be about 100% for photons below 60 keV.

Tissue absorption

Photons from heavy elements (*L* X-rays) are severely attenuated by small thicknesses of tissue. Small changes in the chest wall thickness will cause large changes in the transmitted photon intensity. It is thus imperative that an estimate be made of the average chest wall thickness of each individual counted in order that the proper calibration factor be used.

Ultrasonic chest wall measurements. There has been considerable effort in recent years to develop ultrasonic methods to measure chest wall thickness (De73; Ra67; De70). In addition to the wide range of average chest wall thickness among individuals, the thickness may vary in a given individual from 20 to 50 mm. Also, the left chest wall is typically thicker than the right. The most common procedure used to allow for this is to make at least six measurements over the inter-costal spaces within the area covered by each detector. For example, in using two 125-mm diameter phoswich detectors, a total of 12 measurements would be made within two 125-mm diameter circles, one on each side of the chest. The effective thickness for both sides of the chest is then calculated. Since it is the average transmission that is required rather than the average thickness, $e^{-\mu T}$ is averaged, where T is the measured thickness in mm and μ is the linear attenuation coefficient (mm^{-1}).

Another ultrasonic method of measuring average chest wall thickness is the compound arm "B" scan system which uses an ultrasonic probe mounted on a flexible arm (Ca76). Transducers on the arm not only indicate chest wall thickness but also measure the position of the probe on the subject's chest. As the probe is moved across the chest, a two-dimensional display is generated which yields contours of a cross section of the chest and of the lung. Several profiles are made, typically four, across the area covered by the detectors. Thickness measurements can then be made at many points and exponentially averaged. The principal advantage of this system is that the complete chest profile is seen, making it somewhat easier to identify the lung surface. However, this advantage is mediated by the high cost and complexity of the mechanical and electronic assemblies and by the longer period of time required to make the measurement.

Height-weight predictors. Studies to develop methods of predicting the average chest wall thickness (De73; Ra67) and "effective" tissue thickness (Ru69) from easily measured body parameters such as weight and height have yielded three predictive equations.

$$T_M = 0.71 + 0.512\ W/H. \qquad (1)$$

This equation was derived from ultrasonic measurements on 393 subjects (De73). The standard error of an estimate for T_M with equation (1) was 2 mm.

$$T_M = 1.53\ W/H - 10.0\ C - 35.5. \qquad (2)$$

The standard error of an estimate using equation (2), based on 19 subjects (Ra67), was 1 mm with a 95% confidence limit of 2 mm. In both equations (1) and (2), T_M is the mean thickness in mm, W the weight in kg, H, the height in m and C the chest circumference in m.

The "effective" tissue thickness was determined by measuring the relative transmission through the chest wall of 20-keV X-rays and 323-keV gamma-rays emitted by a known mixture of ^{103}Pd and ^{51}Cr inhaled by human subjects. The data from this experiment yielded a third equation (Ru69).

$$T_{eff} = 30 + 0.9\ T_M. \qquad (3)$$

The coefficients of this equation have estimated standard deviations of less than 10%. The constant term (30 mm) represents absorption in the lung itself and it shows that

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the lung is approximately an infinitely thick source of low energy X-rays.

As an example of the use of these equations, a direct ultrasonic measurement of one subject with a weight of 74.2 kg, height of 1.71 m and chest circumference of 1.0 m yielded an average chest wall thickness of 23 mm. Equation (1) predicts 22 mm, equation (2) predicts 21 mm and equation (3) predicts an "effective" thickness of 49 mm. In this example equations (1) and (2) predicted the actual thickness very well, but the equations are based on averages and will not necessarily do as well for the nonaverage subject. The best and preferred procedure is to ultrasonically measure the chest wall thickness of each individual at the time of each count.

Subject background prediction

Some laboratories determine total background (subject plus detector) by averaging counts from two or more individuals of similar physique who have never worked with radioactive materials. This background is then subtracted from the gross count of a possibly contaminated subject to give a net count which is the result of the inhaled heavy element. This method has an estimated accuracy of $\pm 15\%$ (2σ) for determining correct background; it does not account for variations which occur from day to day or month to month, such as those in room background or in a person's diet. Therefore, it is used most successfully by laboratories in which background count rates are not significantly affected by atmospheric changes, nearby reactors, plant effluents, or diet uptakes of ^{137}Cs. Where the room background is not particularly stable, the current value must be subtracted along with the net counts from the previously counted background individuals.

At other laboratories, a person's background is derived from a correlation of the subject's count in a higher energy band (e.g. 75–100 keV) with that in lower energy bands of interest (Um76). The accuracy of this technique is believed to be ± 10 to $\pm 12\%$ (2σ).

The effect on an estimated lung burden of any error in estimating a subject's background is a function of the amount of inhaled heavy elements. When the lung content approaches the minimum detectable limit a $\pm 10\%$ error in background estimate can affect the final assessment by as much as $\pm 50\%$. In cases where the content is well above the detectable limit (gross count much greater than background), the error in an assessment resulting from an incorrect background ($\pm 10\%$) is less than $\pm 10\%$.

Calibration techniques

Detector calibration, used to convert net counting rate to lung burden of an isotope, is a critical part of any counting system. It can be accomplished with the aid of one of several different methods of varying complexity. Four parameters must be considered in an instrument calibration.

- Effect of chest wall thickness.
- Interference from high-energy gamma-rays.
- Correction for natural subject background.
- Distribution of material in the lung.

Two methods are currently used to determine the calibration factor as a function of chest wall thickness. Most investigators use an anthropomorphic phantom with a known average chest wall thickness to establish one point on the calibration curve. A transmission curve measured in anthropomorphically correct geometry (mock lung source with detector, absorber and source in constant contact) is then used to generate the complete calibration curve (De70). However, this method is dependent on the similarity between the attenuation properties of the phantom materials and those of human tissues. The second method is to use human subjects of different chest dimensions who have inhaled particles labelled with radioactive isotopes (Ne72; Ne75; Ra72; Ru68). For example, a mixture of ^{103}Pd and ^{51}Cr in known proportions has been used. The ^{103}Pd with a 17-day half-life emits 20.2-keV X-rays and was selected to simulate ^{239}Pu. The ^{51}Cr with a half-life of 27 days emits 323 keV gamma-rays and is used to determine the retention of the particles. From the known composition of the inhaled aerosol, the lung content of ^{103}Pd can be calculated. A calibration point is thus derived for each person who inhales the material. This method is more direct but much harder to use and thus seldom used.

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Other *in vivo* methods (e.g. injected albumin microspheres labelled with ^{99m}Tc) are currently under investigation and may prove to be useful.

If more than one isotope is present, for example, ^{239}Pu and ^{241}Am, detector response in the X-ray region must be corrected for the effect of scattered gamma-rays (60 keV for ^{241}Am). This correction will depend on absorber thickness and must be determined in the same way as is the primary calibration.

Intercalibration for operational counting

Three interlaboratory calibration experiments pertinent to operational counting programs have been performed. In the first experiment, "point" sources of ^{238}Pu and ^{241}Am, with nominal activities of 5, 50 and 500 nCi, were measured by ten laboratories (Sw73). This study was designed to test the performance of counting instruments in a fixed geometry and with a wide range of count rates, as well as to compare source standards. It showed that there is good instrumental control at the participating laboratories; the major discrepancies appeared to be in the assays of the activities of the individual laboratory standard sources. All ten participating laboratories have now been provided with standard sources (National Bureau of Standards) so that all future intercalibration efforts will have a common basis for comparison.

An intercalibration experiment using a person accidentally exposed to ^{238}Pu has also been performed. An individual with an estimated 10–35 nCi of ^{238}Pu was counted at six laboratories. The estimates of the lung burden had a mean and standard deviation of 17.7 ± 5.6 nCi. The average chest wall thickness was measured (5 laboratories) at 23.7 ± 1.5 mm (standard deviation). One difficulty with this kind of intercalibrations is that the measurements are only relative; the exact lung burden is never known. These results are, however, encouraging and this type of intercalibration experiment should be repeated as often as possible.

Another intercalibration experiment, sponsored by the International Atomic Energy Agency, has been performed using the ^{103}Pd-^{51}Cr mixture technique (Ne75). Fifteen laboratories participated, including six in Europe. Each laboratory counted three individuals for both isotopes. With the known Pd/Cr ratio the ^{51}Cr content, easily measured by conventional whole-body counting techniques, could be used to estimate the ^{103}Pd burden and thereby provide a calibration point. Although there was good agreement among the laboratories on the lung burdens of ^{51}Cr, there were large discrepancies in the estimates of ^{103}Pd lung burdens and in the plutonium calibration factors derived therefrom. These differences are attributed to differences in calibration techniques. A preliminary report on this experiment has been presented (Ne75), and a final report was recently published (Ne78).

Isotopic composition

When counting plutonium *in vivo* one counts either the *L* X-rays from uranium daughters or the 60-keV gamma-rays emitted by the ^{241}Am in the material. When attempts are made to correlate the amount of material with the X-ray or 60-keV gamma-ray intensity, isotopic composition of the material is a major concern. Material that is nominally ^{239}Pu will contain small percentages by weight of ^{238}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{241}Am, the relative amounts depending on the irradiation history of the plutonium stock material and the time since separation. Since ^{239}Pu has a low specific activity and a low X-ray yield, other isotopes with higher specific activities and/or higher X-ray yields can influence the X-ray yield of the material when present in amounts normally considered insignificant on a weight basis (Sw70; Sw72b).

Americium-241, which results from the decay of ^{241}Pu, may be a major factor in increasing the number of X-rays (Np *L* X-rays) emitted on a per gram basis; it also emits gamma-rays at 26.4 and 59.6 keV. With its high specific activity and 60-keV gamma-ray, ^{241}Am is often used as a tracer for the plutonium. Although the penetrating power of the 60-keV gamma-ray makes this a useful method, many people prefer to rely on measurement of the X-rays because of the possibility of different metabolisms of plutonium and americium, thus increasing the possibility of an error. Soon after a suspected

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exposure, the effects of differential metabolism will not have had time to occur, and ^{241}Am can be used as an indicator of exposure. If the X-ray counting rate is used to estimate the lung content, the 60-keV gamma-ray response is used to correct for the ^{241}Am contribution at X-ray energies. Correction of counts for ^{241}Am content will leave one with X-ray counts from all the isotopes of plutonium present. For a typical plutonium isotopic composition the number of X-rays emitted per alpha particle can easily be 30% greater than for pure ^{239}Pu (see Table 1).

As more plutonium produced in power reactors enters our working environment, the percentage of ^{239}Pu in a plutonium sample will decrease and the number of X-rays emitted per alpha particle will increase, due to the increase in plutonium isotopes with higher X-ray yields than ^{239}Pu. ^{241}Pu (the parent of ^{241}Am) decays by beta emission 99 + % of the time and does not contribute directly to the X-ray emissions from a plutonium sample. Until ^{241}Am grows into a plutonium sample, the X-ray emission rate per maximum permissible lung burden will decrease for high burnup plutonium. The growth of ^{241}Am will greatly increase the X-ray intensity of aged high burnup material. For reactor material containing about 70% ^{239}Pu by weight, the number of X-rays per alpha particle will nearly triple in the first 5 yr following separation because of the growth of ^{241}Am from the ^{241}Pu. For weapons grade plutonium from 3 to 10 yr post separation, one can use 0.085 as a rough estimate of the X-ray to alpha particle ratio and be accurate to ± 25%.

Lack of knowledge of isotopic composition will generally cause errors smaller than those arising from other sources, such as prediction of subject background and tissue absorption, except where ^{241}Am is used as a tracer for the plutonium. In this case it is important that the content of americium in the plutonium be accurately estimated. Estimates taken from bioassay samples have generally proven to be unreliable in the past and an estimate must be made from the contaminating material. To estimate the lung content one should know the approximate isotopic compositions used at the site and for the most reliable estimate of the lung content, one should obtain an isotopic analysis of the contaminating material.

Tissue distribution

Lung counting procedures are influenced by both interorgan and intraorgan distributions. The influence of intraorgan distribution (translocation to lymph nodes, liver, etc.) is widely recognized and calibrations have been made for such distributions (Sw68b; Sw70; Do74). Although nonuniform distribution within the lungs has been suspected, present calibrations of lung counting systems assume a uniform distribution of plutonium.

Recent evidence corroborates nonuniform distribution of material within the lungs. Autopsy data at both PNL (Nel72) and at Los Alamos (Mc76; Vo76) indicate a definite long-term trend of accumulation of plutonium in the outer areas of the lung. Recent data from Los Alamos (Mc76) have also shown a difference in concentration from the apex to the base of the upper lobe.

The distribution of material within the lungs of an individual will influence the estimate of the lung content. Information presented at a recent workshop on lung counting showed that a nonuniform distribution can exist (Sw76b) and indicated that an increased concentration of activity in the periphery of the lungs could cause a substantial increase in detected X-ray intensity when compared to a uniform distribution. Results, both experimental (Go76; Um76) and theoretical (Sw76b), indicate that a factor of 2.5–3 is a reasonable estimate of the maximum increase. An informal discussion at the workshop suggested that an error of about ±70% for the effect of nonuniform distribution was reasonable (Sw76a). The errors caused by redistribution will affect the intensities of the X-ray emission more than the intensity of the 60 keV gamma-ray emission. This will lead to an erroneous correction for the X-ray contribution due to ^{241}Am, which will add to the error in an estimate of the plutonium lung burden.

Several years after exposure the influence of intraorgan and interorgan distribution will complicate the lung counting geometry. Concentration in the lung periphery will form a "shell" of increased activity which will

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yield increased counting efficiency. Translocation to the lymph nodes (interorgan distribution) will form points of activity near the center of the shell (lung) which will reduce counting efficiency. It is difficult to predict whether the net effect of long-term redistribution (translocation) will be positive or negative or to estimate its magnitude. Even if one could estimate an average effect, biological variation would put wide ranges on both the intraorgan and interorgan distribution of plutonium. Factors such as particle size, respiratory infections, smoking habits, breathing pattern, etc. will also influence the deposition pattern of inhaled materials and, thus, the early clearance of material (Ta64). The counting efficiency and calibration of a lung counter may be modified by these variables.

Surface contamination

Since photon emissions from surface-deposited plutonium will not be attenuated by overlying tissues, surface activity in excess of 0.01% of the internal activity can lead to overestimation of the internal deposition (Sw68b) when a small-area detector (a few cm^2) is directly over the surface activity. This effect will not be as serious for larger-area detectors at such a low level of surface activity. Precount showers and alpha surveys are used to control surface contamination. When surface contamination is suspected, a number of techniques can be used to determine if such contamination is a problem. If the intensity of the *L* X-rays compared to the 60-keV gamma-ray of ^{241}Am is higher than expected, surface contamination is suggested; radically different count rates over the chest and over the back also suggest surface contamination. The appearance of the 13.6-keV line in a proportional counter spectrum is proof of surface contamination.

Precision and accuracy in lung counting

The possibility of an error in the estimated lung content is large when the estimate approaches the detectable limit of the lung counter. Errors result from various assumptions used in developing the method of calibration. For example: the lungs contain a uniform distribution of activity; estimates of chest-wall thickness are accurate; the subject's physical characteristics are closely approximated by the phantom used for calibration; the subject's background can be predicted with accuracy; and the isotopic composition of the contaminant is known.

Error analysis of the known factors is probably handled differently at each laboratory since there are at least as many specific methods of calibration as there are laboratories. Discussion of the errors involved in the calibration was a significant part of the recent workshop (Sw76a) on lung counting. The final session was followed by a general discussion on the effect of errors on the assessment of a maximum permissible lung burden (16 nCi) of ^{239}Pu. Major sources of error are listed below with their estimated effects for a typical phoswich detector system, and for a subject of average build.

- Subject background ±50% (2σ).
- Chestwall thickness ±40% (2σ).
- Activity distribution in lung ±70% Systematic error.
- Counting statistics ±50% (2σ).

Other sources of error are small. As already mentioned, the third item is commonly ignored and the resultant combined error (items 1, 2 and 4) amounts to ±81% (±13 nCi for 2σ). Another indicator of precision in the assessment of inhaled plutonium is suggested by the results of interlaboratory measurements on an individual in 1974. In that case the subject of an accidental exposure was counted at six different laboratories within 2 weeks and results varied from 10 to 23 nCi of ^{238}Pu (one laboratory's result of 54.8 nCi is excluded by Chauvenet's criterion). The mean of the six results was 17.7 nCi or about twice the average laboratory's detectable limit. This is merely an indication of precision and not necessarily of accuracy.

Precision is expected to improve significantly by 1980 as a result of a single intercalibration phantom that will be used by most U.S. lung counting laboratories during the next 2 yr. This should improve precision to the point that true accuracy will be the challenge. However, accuracy will probably develop slowly because of unknown factors such as distribution of lung depositions of heavy elements.

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TRENDS IN OPERATIONAL COUNTING RESEARCH

The measurement of heavy elements *in vivo* involves many variables and the utmost sensitivity in a counter. Research efforts are continually exploring new concepts which may increase counting sensitivity or may lead to improved accuracy. Trends in research will foretell the future of operational counting equipment and procedures. This section surveys a number of recent concepts in lung counting and evaluates their performance.

New counters

Interest in the possible use of semiconductor detectors for assessment of lung content has recently been aroused. To approach a detection limit comparable to that of existing phoswich detectors, it is necessary to use arrays of semiconductor detectors to achieve a reasonable sensitive area. Single semiconductor detectors are not currently available with areas larger than $\approx 1200\ mm^2$ (see Table 2).

The only detector array currently being investigated for lung counting uses intrinsic germanium, with two arrays, each with four $1000\ mm^2$ detectors, costing a total of about $90,000 (Ty76). Reported experience with this system is limited. Others (Be76) have investigated single germanium detectors and predicted the response of an array from the single detector response in order to compare it to a phoswich detector. Results look somewhat promising for solid-state detector arrays, but a conclusive report has yet to show actual improvements in detectable limits compared to those for a phoswich counting system.

Arrays of silicon detectors have not yet been adequately considered. It is possible that silicon surface barrier or lithium-drifted silicon detectors would work as well as germanium and could possibly be operated on a thermoelectric cooling device without liquid nitrogen. The performance of silicon detectors (1–2 cm depleted depth) should compare to that of germanium for photons up to 60 keV in energy, and they should cost considerably less.

Cadmium telluride, gallium arsenide and mercuric iodide are other semiconductor detectors with potential for counting heavy elements in the chest if large active areas can be obtained. Recent developments in proportional-scintillation counters (Pa76) have indicated a potential for detecting heavy elements in the chest. Preliminary results indicate that resolutions are superior to conventional proportional counters (Pa75).

Fluorescent K X-rays

The use of characteristic *K* X-rays of plutonium as a method for its assay has been investigated at Argonne National Laboratory (Ru78). The *K* X-rays have energies of 99.5 keV ($K\alpha_2$) and 103.7 keV ($K\alpha_1$); the K_β lines have even higher energies but are less intense. Assaying plutonium by measuring these photons rather than uranium *L* X-rays has the advantage of less attenuation in the lung and chest wall, and less variable transmission with chest wall thickness. The ANL investigation showed that the *K* X-rays of plutonium (absorption edge at 121.72 keV) could be excited by the γ-rays from ^{57}Co (energy of most intense line 121.97 keV). It was calculated that, even if the rate of the absorbed dose at the skin were as high as 350 mrad/hr from the ^{57}Co, the counting rate in the $K\alpha_1$ peak in the spectrum of a solid-state detector (40 mm diameter) would be much lower than the counting rate of a phoswich detector from the uranium *L* X-rays from the same amount of plutonium in the lungs. Furthermore, scattered radiation from the 136-keV γ-rays from ^{57}Co (intensity 11%) would increase the background of the fluorescent X-ray system. The method is thus quite impractical.

Methods using induced fission

Rundo *et al.* (Ru78) have suggested that induced fission of some of the plutonium in the lung by neutrons from an external source might provide possibilities for reducing the limit of detection. Three methods were considered: collection and assay of exhaled fission product gases; measurement of prompt fission γ-rays; and measurement of delayed neutrons. The first and third of these offer a high degree of specificity; the second

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deserves investigation as it may be the most sensitive of the three techniques.

Collection and measurement of exhaled 17.5-min ^{138}Xe (fission yield 0.06) would be a highly specific method for detecting and quantifying fissile material in the lung. With some reasonable assumptions, it was calculated that 2000 induced fissions would be detectable.

In principle it should be possible to detect as few as 115 fissions by assay of the abundant (about seven per fission) prompt fission γ-rays, with a large (e.g. 200 mm diameter, 100 mm thick) NaI (Tl) detector; but interference from γ-rays produced by radiative capture in the abundant elements (especially hydrogen) in tissues of the lung and chest wall would probably be severe. Spectrometric selection of the most intense (11.3%) single line at 139 keV would offer no real advantage because the reduction in signal strength would far outweigh the reduction in background. There might be an advantage in measuring the (30 ± 3)-keV X-rays (38% abundant) with a phoswich detector.

Measurement of fissions via the delayed neutrons from ^{127}I and ^{87}Br would be less sensitive because of the low yields of the emitters (2.4% and 0.7% respectively) and the low intensities of their neutron emissions (6% and 2% respectively). It would be difficult to obtain a low-background neutron detector of the size necessary to achieve 10% overall efficiency. Some of the neutrons emitted would be moderated and captured *in vivo*, primarily by hydrogen, which might be turned to advantage. That is, the neutron detector could be replaced by a large NaI(Tl) detector, which would be used to measure the 2.23-MeV capture γ-rays from hydrogen. This method has been proposed (Ne72a) for the determination of spontaneously fissioning ^{252}Cf *in vivo*.

The practicality of any of these methods depends on it being possible to induce a sufficient number of fissions in 0.256 μg of ^{239}Pu (which equals 16 nCi, the maximum permissible lung burden) with an acceptable radiation dose. Rundo *et al.* (Ru78) estimated that a 1-rem dose equivalent from about 10 keV neutrons would be necessary to induce about 650 fissions in 1 MPLB. Much experimental work would be required before there could be any question of facing the legal and administrative problems involved in deliberately exposing personnel to the required external radiation.

Calibration and intercalibration

Investigators in the lung counting field are continually working to improve the calibration of their counters and the accuracy of their results. Intercalibration experiments allow a comparison of laboratory capabilities and point out deficiencies in individual capabilities. Direct calibration techniques, if successful, will permit the exposed individual to act as his own calibration, thus reducing individual variations.

Direct calibration. A direct human calibration experiment using ^{99m}Tc is planned. In this experiment albumin microspheres (15–30 μm diameter), labelled with about 20 μCi of ^{99m}Tc (18.3 keV *K* X-rays, half-life 0.25 days) are to be introduced into the blood stream of the lung by intravenous injection. The microspheres are of such a size that they become trapped in the lung capillaries, thereby providing a direct calibration for a uniform distribution of an X-ray emitter in the lung volume. The procedure is used for lung scanning in nuclear medicine and could in principle be used for direct calibration for any subject with a detectable deposition of plutonium in the lungs. The predicted radiation dose to the lungs is less than 1 mrem/μCi injected.

Another nuclear medicine technique being studied for direct counter calibration is the use of radioactive noble gases (0176). In nuclear medicine the inhalation of a radioactive noble gas is used to determine the perfusion of the lung. For *in vivo* counting the subject would inhale a bolus of radioactive noble gas (^{85m}Kr, ^{133m}Xe) and hold his breath for the duration of a short count. Comparison of the count rate *in vivo* with the count rate from the same activity of gas outside the chest will indicate the absorption in the lungs and chest wall.

Intercalibration phantom. Due to the difficulty in performing intercalibration

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experiments with humans, a major effort is being directed toward developing an anthropomorphic phantom designed specifically for use in X-ray and low-energy photon emitters (De76a; De76b). The phantom will have a variety of chest wall thickness profiles corresponding to average profiles of plutonium process workers to permit the generation of complete calibration curves. Its easily removable organs will permit calibration for different isotopes in the lungs, liver and lymph nodes, for both uniform and nonuniform distributions. Construction of this phantom was completed in 1978.

Human intercalibration. It is anticipated that another ^{103}Pd–^{51}Cr intercalibration experiment will be performed during 1979–1980. The proposed experiment is strongly endorsed by the U.S. Intercalibration Committee, which would assist in data collection, data analysis and coordination of activities within the United States. This experiment will parallel the 1972 experiment (Ne78) and should reduce or explain the large discrepancies in estimation of ^{103}Pd lung burdens. These experiments provide a realistic basis for comparison of counting facilities and are extremely valuable in improving the state-of-the-art.

Intraesophageal probe. Plutonium originally deposited in the lungs will translocate to other portions of the body including the thoracic lymph nodes. Early experiments with dogs indicated that the translocated material in the lymph nodes may reach concentrations 50–100 times greater than those in the lung volume, about 10 yr after exposure (Pa72). Results obtained in autopsy sampling programs have shown similar results for humans (Ca74; Nel72). Biological evidence has not yet indicated that the concentration of material in the lymph nodes is detrimental to the well-being of the organism, but this translocation will affect the dosimetric evaluation of depositions and may affect interpretation of external lung counts since counting efficiency varies with plutonium distribution.

Small detectors (4.7 mm diameter, 25 mm long NaI(Tl) crystals) have been developed to detect material deposited in lymph nodes (Sw72b; Sw76c). In one experiment (Sw76c) two human subjects were examined with an intraesophageal probe containing one of these detectors. Results from one subject clearly demonstrated the deposition of material in the tracheobronchial lymph nodes. The ^{241}Am gamma-rays were used to estimate the plutonium lung content. The prototype probes have a minimum detectable amount of approx. 3 nCi* (2σ = 1.2 nCi) of weapons grade plutonium deposited in the tracheobronchial lymph nodes. This could readily be reduced by a factor of two by selection of low background materials and redesign of the probe housing.

The intraesophageal probe is not widely accepted as a routine counting technique partly because of the need to have a physician in attendance during its insertion. The probe was not intended to replace conventional chest counting techniques so much as to aid in data interpretation. The probe could be used to provide information on the translocation rate of the material and thus allow for some correction for its distribution. It offers an alternative technique for assessment of lung content of heavy elements. Estimates indicate that a probe approx. 120 mm long and 10 mm diameter in the esophagus would be equivalent to an external phoswich detector system. Alternatively, with the present probe in a major bronchus it is estimated that lung contents as low as 3–5 nCi* of ^{239}Pu could be detected.

Spatial resolution counters

Because of the uncertain distribution in the lung of inhaled radionuclides, it would be useful to have a detector that could measure not only the total activity but also its distribution in the lung.

Current calibration techniques are based on a uniform distribution of activity in the lung. Efforts are now underway, particularly in the design of the intercalibration phantom, to permit calibration with nonuniformly distributed sources. It will still be necessary to develop a method to determine the dis-

*Calculated at 90% confidence level with a 20-min counting period (i.e. MDA = 2.56/S B/20, S = cpm/nCi, B = Background rate).

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tribution in the lung, preferably for each subject. Two detector systems have been proposed for this purpose. One proposed detector is the xenon gas-filled multiwire proportional counter, using resistive wire elements such that localization in two directions is possible with one chamber. This detector also has good energy resolution, low background and can be made quite large. The advent of solid-state detectors has made it possible to construct counter arrays with very good energy resolution and low background. However, to duplicate the area of two of the 127 mm diameter phoswich detectors would require 25 of the largest solid-state detectors. An array of solid-state detectors could provide information while counting plutonium deposited in the lungs.

The major difficulty in using these devices has to do with the expected count rates. A maximum permissible lung burden of ^{239}Pu (16 nCi) produces a photon rate at the surface of the average chest of about 0.016 cpm/cm^2. Each element of a solid-state array of 25 detectors would then produce 0.16 cpm, for a total of approx. 120 counts in the typical counting time of 30 min. Once the subject's natural background is added to this, it appears highly unlikely that either of the proposed detector systems will be able to measure activity distributions at the maximum permissible level, certainly not at lower levels. However, if a large fraction of the lung content were concentrated in a small region of the lung, these detectors might be of value. The major potential of these instruments lies in their use in high-level accidental exposures, and in human calibration experiments using large quantities of short-lived isotopes.

Tissue absorption estimation

Detectors with good energy resolution such as proportional counters and solid-state detectors allow direct observation of the plutonium daughter *L* X-ray groups at 13.6, 17.2 and 20.2 keV. At these energies the absorption coefficient of tissue varies rapidly with energy, and the measured differential attenuation of the X-rays provides an indication of the amount of interposed tissue. The 13.6-keV X-rays will not be observed for lung-deposited plutonium because of their rapid attenuation. Detection of these X-rays indicates surface contamination.

Comparison of the relative X-ray intensities has often been suggested for estimating depth of deposition in wounds and for chest-wall absorption for lung counts (Pa68). The technique is effective for point sources of activity; however, for distributed sources, the X-ray ratios will indicate a depth or a chest wall absorption representative of the exponential average of the distribution (Sw68a), which may or may not be representative of the absorption path of interest. More importantly, in lung counting one is dealing with net counts having poor statistical precision, and separating the net counts (17.2 and 20.2 keV) will result in two numbers with poor statistical precision. Any ratios formed from such numbers will be of little use because of the large statistical error. Furthermore, scattered photons from the 20.2 keV line make a significant contribution to the counts observed in the 17.2 keV peak. Consequently the ratio of the two becomes nearly constant for absorber thicknesses greater than 20 mm.

X-ray intensities

The X-ray intensities for the common isotopes of plutonium, americium and curium are well known, but the *L* X-ray intensities of some uranium isotopes (i.e. ^{232}U, ^{233}U) have not been accurately measured. The advent of solid-state detectors has increased interest in the measurement of the relative intensities of major *L* X-ray groups (L_l, L_α, L_β, L_γ). The differential attenuation of the three major *L* X-ray groups (α, β, γ) could result in errors if the partitioning of the X-rays among the groups is different in the exposure material from that in the calibration material. Preliminary measurements indicate that a serious discrepancy does not exist for the isotopes of plutonium (Be76; Too76).

Organ distribution

In principle it should be possible to determine the heavy element contents of lung, liver and skeleton from the results of

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measurements at three appropriate locations, provided the levels of activity are sufficiently high. What little information is in the published literature on this topic (Do74) does not give sufficient data to assess reliably the sensitivity of the technique for determining liver and skeleton burdens. For the latter a crude estimate can be calculated. A detector 127–200 mm in diameter will view about 0.1–0.2% of the total surface area of the skeleton when placed over the calvaria, where absorption in overlying tissue is quite small (about 25%). Then it is estimated that 40 nCi in the skeleton (the maximum permissible) would give 0.01–0.02 counts/sec, which should be detectable if the contribution from body "background" can be predicted accurately. The limited experimental data available are in approximate agreement with these estimated counting rates (within a factor of two to three) (Too75; Too76).

The situation for the liver is not unlike that of the lung, except that absorption in the organ (which is much smaller) is greater because of the higher density; like the lung, the liver is covered by a variable thickness of tissue. The calibration data (Do74) show that the counting efficiency for plutonium in a phantom liver is about half that for plutonium in the lung, when the chest-wall thickness is the same as the abdominal wall thickness. Thus, the limit of detection for plutonium in the liver must be roughly twice that for plutonium in the lung. Interference ("cross-talk") from activity in the lung, when the liver is being measured, can be allowed for by making a "background" measurement on the left side of the abdomen. The possibility of detecting a contribution from plutonium in the spleen in such a measurement should also be considered.

INFLUENCING TRENDS

Development of lung counting capabilities will be influenced by trends in the nuclear field. Changes in our assessment of radiological risk, in the production rate and composition of transuranic materials or in our knowledge of metabolism of transuranic materials will influence the requirements for lung counting. This section indicates some of the trends recognized at the present time.

Lowering maximum permissible lung burdens

Requests for substantial reductions in the permissible level of plutonium deposited in the lungs are based on the nonuniform distribution of the dose from discrete particles. This "hot particle" problem led to a reassessment of current radiological data (Ba74) and to new experiments. To date there is no indication that a nonhomogeneous dose distribution will result in a demonstrably greater risk than that assumed for a uniform dose distribution. Present biological evidence does not necessitate a substantial reduction in present maximum permissible lung burdens. Small reductions may occur, but will certainly be less than a factor of ten.

Future production of transuranium elements

Plutonium from power production will account for the major mass of transuranium material in the working environment in future years. If current forecasts are correct, the annual production rate for plutonium in the U.S. will increase from 4400 kg/yr in 1975 to 40,000 kg/yr in 1985 (Fa74). In the same period of time the weight percentage of ^{239}Pu in the material will decrease from 64 to 54% and the weight percentage of ^{241}Pu will increase from 10 to 12% (Fa74). The isotopic composition and masses of the plutonium material will be changed slightly when breeder reactors begin producing plutonium. Delays and cancellations in power reactors could reduce the projected production rates and change the average isotopic composition of available material.

In addition to the plutonium from light-water moderated power reactors discussed in the previous paragraph, a large inventory of weapons grade plutonium will continue to exist. This is plutonium containing approx. 93.6% by weight of ^{239}Pu.

Production of ^{238}Pu will continue to be of concern since ^{238}Pu enters the working environment in power sources for space and biological (heart pacers) applications. Production of ^{238}Pu has been forecast at 10–20 kg/yr in 1970–1980 and 100 kg/yr in 1980–1990 (La72). It has a specific activity 275 times as great as ^{239}Pu, so the alpha activities of these quantities of ^{238}Pu will be similar to that of ^{239}Pu from light-water reactors.

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Quantities of other transuranic isotopes such as ^{241}Am, ^{244}Cm and ^{252}Cf will also enter the working environment. Production of ^{244}Cm with a specific activity about 1350 times that of ^{239}Pu is expected to increase from 40 kg/yr in 1970–1980 to 180 kg/yr in 1980–1990 (La72). Californium-252, with a specific activity about 6900 times greater than ^{239}Pu, will be produced in quantities of about 0.1 kg/yr in 1970–1980 and 0.8 kg/yr in 1980–1990 (La72). These transuranic elements (^{244}Cm ^{238}Pu, ^{252}Cf) have *L* X-ray intensities associated with their decay of around 0.1 X-ray/alpha and present counting difficulties similar to those of ^{239}Pu (Sw72a). With minimum effort the expertise developed for counting ^{239}Pu can be extended to the other transuranic elements, although ^{252}Cf can be assayed as a result of its spontaneous fission (Ne72a).

Trends in biohazards

In the early 1960s the primary transuranic element of concern was plutonium, especially PuO_2, since it was the likely form to be encountered following an accidental release. Animal experiments demonstrated that inhaled PuO_2 was largely retained in the lungs and in the thoracic lymph nodes. With the growing availability of other transuranics for research and with the emphasis on nuclear power development, americium and curium, as well as other isotopes and forms of plutonium, are of increasing concern as potential health problems.

Recent animal experiments (Ba75) have shown that the biological behavior of inhaled transuranics other than $^{239}PuO_2$ is quite different than that of $^{239}PuO_2$. Americium and curium, regardless of the chemical form in which they are inhaled, are largely translocated to liver and skeleton and they accumulate to a lesser degree in thoracic lymph nodes than $^{239}PuO_2$. Evidence indicates that $^{238}PuO_2$ translocates to liver and skeleton more than $^{239}PuO_2$, but in lesser amounts than Am and Cm. Further, experimental data suggest that particle size characteristics and the method of preparing the aerosol, as well as the magnitude of the amount initially deposited in the lungs, may alter pulmonary retention and translocation rates. Exposure to mixtures of transuranics such as mixed reactor fuels has not been studied and knowledge of their biological behavior in an accidental release is essential.

For accidental inhalation exposures, counting of materials other than $^{239}PuO_2$ in the lungs will remain critical during the initial evaluation. However, for assessing old exposures and to follow the long-term translocation of many transuranics, techniques to determine depositions in the liver, the skeleton and other soft tissues must be developed.

IDEALIZED COUNTING CAPABILITY

The previous sections have suggested the present counting capabilities, the research trends, and developing trends outside the field which could influence future counting requirements. It is valuable to project the capabilities one would consider ideal and thus provide the specific goal. This goal may not be realistic in terms of known potentials, but it will point out areas needing most attention.

Parametric information

An ideal counting capability would provide several specific pieces of information from which the lung content would be estimated. An accurate assay will involve the following parameters:

1. The specific isotopic composition of the material.
2. The background contribution from the individual.
3. The photon absorption characteristics of the chest wall.
4. The distribution of material within the lungs.
5. The presence or absence of surface contamination.
6. The influence of ribs and posture on counting efficiency.
7. The organ distribution.
8. The room background.
9. The radiologic properties of the calibration phantom versus those of the subject.
10. The chemical form of the material.

Knowledge of these parameters and their influence on counting makes possible an accurate estimate *in vivo* of the lung content. Furthermore, this information will allow a

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reasonable estimate of the long-term effects of an accidental inhalation.

Many of the parameters are routinely used in an assessment. However, counting data are not usually compensated for effects due to distribution between organs and within the lungs. The accuracy with which we know the influence of the various parameters on the assessment should be improved.

Counting limits

Most gamma emitters routinely assayed by whole-body counting techniques can be counted at levels well below one-tenth of a maximum permissible body burden with good accuracy. This gives the health physicist good control of his assessments and provides confidence in the measurement. While the body burden of most gamma emitters can be accurately counted, the accurate measurement of lung burdens of heavy elements is still far from ideal. The present capability is from 1/2 to 1 Maximum Permissible Lung Burden.

Although the standard bioassay procedure of urine analysis can provide an estimate of the internal (systemic) body burden of heavy elements, external counting is currently the only method available to estimate lung burdens. However, until the detection limit is reduced to about one-tenth of a Maximum Permissible Lung Burden, exposures near MPLB levels cannot be accurately evaluated and reliance must be placed on chemical bioassay techniques to detect exposures to both soluble and insoluble forms of the materials.

If the MPLB were to be substantially reduced (a factor of 10), the feasibility of measuring lung burdens by conventional techniques at levels useful for routine health physics would disappear.

Precision and accuracy

A source intercalibration experiment demonstrated reasonable lung counting precision (Sw73). The laboratories were generally within ±8% (1σ) in their determinations of source activity. The precision will be verified by counting the anthropomorphic phantom under development at Lawrence Livermore Laboratory. The use of this phantom will also improve our knowledge of how variables affect accuracy.

The accuracy of lung counting procedures will have to be estimated by post-mortem studies on contaminated subjects. Comparison of tissue analysis with bioassay data will provide an estimate of the accuracy. Such procedures have been used successfully with dogs and have indicated agreement between tissue analysis and external counting procedures (Sw67). Counting on humans will never produce accuracies comparable to animal experiments, which generally involve higher levels of activity and small tissue masses. Information is not available to estimate the accuracy for human counting. Ideally, the accuracy of a determination would be the same as the counting precision.

SUMMARY AND CONCLUSIONS

This report describes the status of instrumentation methods used for measurements of heavy elements *in vivo* by photon counting as well as methods to compensate for uncertainties resulting from the many variables in lung counting. External counting has been routinely used for estimation of plutonium *in vivo* for approx. 10 yr. However, this method is fraught with inherent uncertainties, most of which derive from the fact that the low-energy photons (17 keV average) resulting from plutonium decay (uranium *L* X-rays) are few and are severely attenuated by the body tissues; for instance, measurements for material in the lungs have indicated that typically about 1% of the photons will escape from the thorax (Ru69). The scarcity of the radiations requires an accurate estimate for background counts caused primarily by scattered radiation from other photon emitters within the body. Corrections must also be made for the variation of photon emission rate with isotopic composition, including corrections for the contribution from ^{241}Am, a daughter of ^{241}Pu. The rapid attenuation of the radiation requires that determinations *in vivo* include corrections for chest wall thickness, distribution of plutonium, and surface contamination. Once the data are corrected for

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these various effects, the result is interpreted from a calibration which is usually performed with a phantom. Whether the phantom and resulting calibration are truly representative of the exposed subject is a final uncertainty in the counting procedure.

Present counting capability allows detection of from 1/2 to 1 Maximum Permissible Lung Burden (1 MPLB = 16 nCi) of plutonium depending on isotopic composition. Variables seriously limit the accuracy of any estimate *in vivo*, and in the case of 16 nCi of pure ^{239}Pu, the error limits are $\pm 107\%$ at 2σ (Sw76a). The complexity of determining other transuranic elements with similar photon energies is comparable. Accurate and rapid assessment of depositions *in vivo* requires that continual emphasis be placed on development of counters for low-energy photons and on improved techniques for dealing with the many variables in determining transuranic elements *in vivo*. We do not anticipate a dramatic reduction in detection limits in the near future, although the development of new counters such as solid-state detector arrays and scintillation proportional counters may lead to small improvements in detection limits and accuracy. Their progress should thus be monitored.

Efforts at intercalibration will lead to improved accuracy and better understanding of counting capabilities and they should be encouraged. Intercalibration efforts have shown laboratories where weaknesses exist in their individual capabilities. A repeat of the experiment in which a ^{103}Pd-^{51}Cr aerosol was inhaled by volunteers (Ne75) is needed to resolve discrepancies in the estimates of ^{103}Pd lung burdens and in turn, the Pu calibration factors based on ^{103}Pd lung burdens.

One particularly troublesome area in lung counting is the inability to determine the distribution of inhaled material within the lungs. A direct estimation of the distribution *in vivo* does not appear feasible for lung burdens near the MPLB and efforts must concentrate on improvements in our understanding of the influence of nonuniform distribution on counting efficiency. Animal experiments, medical studies and autopsy studies would help in this area. Calibration phantoms can be used to estimate the magnitude of the effect of nonuniform distribution on *in vivo* counting.

In the past, counting of inhaled materials *in vivo* has emphasized materials which translocate from the lung (solubilize) slowly. Animal experiments indicate that transuranic materials other than $^{239}PuO_2$ may translocate to other organs more rapidly and that bone may become the critical organ for these materials. Thus, procedures should be developed for estimating the quantity of material translocated to the other organs (e.g. in lymph nodes, liver, bone, etc.)

The possible application of new concepts such as induced nuclear fission of the deposited materials should be investigated, and preliminary studies to provide information necessary to justify large-scale experiments should be encouraged. A substantial reduction in the detection limit for transuranic elements can be achieved only through research, development and application of new concepts for the measurements of the material *in vivo*.

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