

INTERNAL DOSIMETRY MONITORING EQUIPMENT: PRESENT AND FUTURE

J. Selby†, M. M. Lardy‡, E. H. Carbaugh§, T. P. Lynch§ and D. J. Strom§

†M. H. Chew & Associates, Inc., 1933 Jadwin,
Suite 135, Richland, WA 99352, USA

‡IT Corporation, 2800 George Washington Way,
Richland, WA 99352, USA

§Pacific Northwest Laboratory, PO Box 999,
Richland, WA 99352, USA

INVITED PAPER

Abstract—Internal dosimetry monitoring equipment used in the successful measurement and assessment of internal exposures is highly dependent on effective capabilities in the measurement of radioactive materials directly and/or indirectly in the body. This capability must be supplemented by comprehensive field measurements to quantify materials (i.e. surface and airborne contamination) in the workplace which become the source of exposure to the workers. Because of current limitations on minimum detectable activities or amounts for *in vivo* or *in vitro* measurements for some radionuclides (i.e. poorly transported alpha emitters), a routine bioassay programme which would maintain the potential minimum detectable dose to a few millisieverts committed effective dose equivalent ($H_{E,50}$) would be prohibitively expensive to operate and unacceptable to the worker. Thus the operation of a strong field measurement programme to provide warning of possible intakes is extremely important to permit timely bioassay measurements to be performed within the limits of detection. The present and future capabilities of the three measurement programmes will be discussed together with the need to balance research and development costs for new or improved measurement techniques with the costs associated with improving knowledge as to the relationship(s) between measurement and the amount present in various organs.

INTRODUCTION

The successful measurement and assessment of internal exposures is highly dependent on effective capabilities of measuring radioactive materials directly and/or indirectly in the body. This capability must be supplemented by comprehensive field measurements to quantify materials (i.e. surface and airborne contamination) in the workplace which may become the source of exposure to the workers.

In the United States (US), the Department of Energy (DOE) established a performance goal for the internal exposure monitoring programme for each radionuclide or mixture of radionuclides⁽¹⁾. Such programmes should be capable of identifying: (1) the occurrence of exposures during a year that could result in a committed effective dose equivalent in excess of the limiting value of 1 mSv (100 mrem); or (2) internal exposures where air monitoring results project an intake of greater than 0.02 of the annual limit of intake (ALI) shown in ICRP 30⁽²⁾.

Because of current limitations on minimum detectable activities or amounts for *in vivo* or *in vitro* measurements for some radionuclides (e.g. poorly transported alpha emitters), a routine bioassay programme which would maintain the potential minimum detectable dose to a few millisieverts committed effective dose

equivalent ($H_{E,50}$) would be prohibitively expensive to operate and unacceptable to the worker. Thus the operation of a strong field measurement programme to provide warning of possible intakes is extremely important to permit timely special bioassay measurements to be performed.

Assessing absorbed dose from irradiation by internally retained radioactive materials depends on knowing, inferring, or assuming the quantity of activity retained in each organ or tissue of interest T as a function of time $q_T(t)$. In its most basic form, the dose to a target tissue, D_T , for nuclides emitting non-penetrating radiation is

$$D_T \propto \frac{\int q_T(t) dt}{m_T}$$

where m_T is the mass of the tissue. More complex relationships exist for radionuclides that emit penetrating radiations, due to the fact that activity in one organ may emit radiation that is absorbed in a different organ.

Three categories of measurements are made to help address the three questions of how much activity is where for how long:

- (1) *in vivo* (direct) counting of radionuclides with photon energies that can be detected outside the body;

- (2) *in vitro* (indirect) assay of radionuclides in excreta samples such as urine or faeces; and
- (3) use of concentration measurements of airborne radioactive materials, respiratory protection factors, aerosol chemical and physical properties and occupancy or stay time.

Each of these three categories of measurements leads down different inferential pathways to address the questions of how much, where and how long. Each *in vivo* measurement leads to an estimate of retained quantities at one or more locations in the body at a moment in time. Each *in vitro* measurement leads to estimates of excretion rates, that is, the time derivative of the whole-body biological retention function. The air concentration approach provides a means of estimating an initial intake via inhalation. All methods require inferential leaps of faith, with the air concentration approach requiring the most, and the *in vivo* approach requiring the least. *In vivo* measurements can give a fairly direct assessment of $q_T(t)$, especially for a radionuclide like ^{60}Co in the respiratory tract. Seldom is that measurement used to calculate dose directly. Rather some calculational method might use such information to assess the value of an intake that would lead to the observed value(s) of $q_T(t)$. That intake is then compared with the ALI or multiplied by an appropriate factor to give dose.

In vitro measurements give the rate at which body content is changing due to excretion by one or more routes. If the mode, time and duration of intake are known (or assumed), then *in vitro* measurements can be used to estimate an intake that is most consistent with observed measurements.

The use of air concentration measurements (which are generally average values over some sampling period) requires assumed or measured values of particle size distribution, chemical solubility class⁽²⁾ (D, W, Y), breathing rate, time during which the aerosol was inhaled, respiratory protection factor, and a set of biokinetic models and model parameters such as those of the ICRP. Intake is calculated for the pertinent time period and compared with the ALI or converted to dose by appropriate dose conversion factors. In some regulatory applications the actual conversion to intake is bypassed and results are reported in terms of DAC-hours exposure.

BIOASSAY CAPABILITY GOALS

The US DOE has specified in its Radiological Control Manual⁽¹⁾ that workers must participate in a bioassay programme (i.e. *in vivo* and/or *in vitro*) when they are likely to receive intakes resulting in a committed effective dose equivalent ($H_{E,50}$) or 1 mSv or more. They must also participate in follow-up bioassay monitoring if routine bioassay monitoring indicates an intake in the current year with a $H_{E,50}$ of 1 mSv or more. No specific statement of required minimum detectable dose

for periodic bioassay has been specified; however, the goal is somewhat controversially argued to be 1 mSv $H_{E,50}$. While readily achievable through *in vitro* and *in vitro* measurement techniques for common fission and activation products, this goal is not currently attainable for most plutonium and many uranium mixtures encountered at US DOE facilities. In either case it is also important that workplace indicators be adequate to identify the need for special bioassay measurements so that worker doses can be effectively controlled.

It is appropriate to consider as a starting point the type and magnitude of intakes which could result in a 1 mSv $H_{E,50}$ for these challenging mixtures. Two specific mixtures are considered: a class Y plutonium-amer-icium mixture characteristic of aged weapons-grade plutonium and a class Y natural uranium (U-nat) mixture. The magnitude of intake for each significant nuclide of each mixture is shown in Table 1. Other mixtures, such as mixed oxides or enriched uranium, tend to be less challenging for bioassay because of the significantly higher quantities of detectable material present.

Using the Table 1 intakes, the retained lung activity, urine excretion, or faecal excretion can be estimated using the method of Lessard *et al.*⁽³⁾ or a computer code such as CINDY⁽⁴⁾. The retention or excretion at various times after acute intake can be taken as approximations of the minimum detectable amount (MDA) required for an appropriate bioassay measurement. The US DOE has not specified that these amounts must be detectable; however, it has been suggested by auditors and operational health physicists that these should be the design goals. The results of these calculations are shown in Table 2 for the Pu mixture and Table 3 for U nat.

The plutonium bioassay goals of Table 2 show that significant technology improvement is needed if routine bioassay measurement programmes are to be capable of meeting the 1 mSv goal. Currently the only way of confidently meeting the goal for Pu mixtures is to obtain faecal samples within the first ten days following an

Table 1. Class Y inhalation mixtures to result in 1 mSv $H_{E,50}$.

Nuclide	Weapons grade Pu		Natural U	
	(Bq)	(ng)	(Bq)	(mg)
$^{239+240}\text{Pu}$	6.5	16 (15 ^{239}Pu) (1 ^{240}Pu)	—	—
^{238}Pu	0.5	0.0008	—	—
^{241}Pu	50	0.01	—	—
^{241}Am	0.6	0.005	—	—
^{238}U	—	—	13	1.1
^{235}U	—	—	0.6	0.008
^{234}U	—	—	13	0.0001
Total mass	—	16	—	1.1

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intake and to interpret these in light of urine samples and longer term faecal samples. This implies that criteria for initiating special bioassay must be quite conservative. The present capability of high purity planar germanium detectors for *in vivo* lung measurements remains 50 to 100 times short of what is potentially required if the goal is to be met. Excreta analyses require comparable improvements. Such improvements will not come easily or cheaply.

Uranium bioassay presents a different set of problems. Table 3 shows that the technology is currently available to meet the bioassay goals; however, the issue is how to recognise excretion from occupational intakes on top of a natural uranium background which is likely to be as large or much larger than the incremental occupational contribution. This is rendered even more complex by the highly variable nature of the background excretion depending on such things as individual work-

Table 2. Bioassay measurement goals for class Y inhalation of weapons grade plutonium (1 µm AMAD^(a)) resulting in 1 mSv H_{E,50}.

Time after intake (d)	Bioassay measurement				
	Urine		Faeces		Lung burden ²⁴¹ Am (Bq)
	²³⁹ Pu + ²⁴⁰ Pu (Bq.d ⁻¹)	²³⁹ Pu (ng.d ⁻¹)	²³⁹ Pu + ²⁴⁰ Pu (Bq.d ⁻¹)	²³⁹ Pu (ng.d ⁻¹)	
1	1.01E-4	2.33E-4	3.39E-1	7.81E-1	1.30E-1
7	1.16E-5	2.67E-5	5.28E-2	1.22E-1	9.00E-2
30	5.73E-6	1.32E-5	8.71E-4	2.01E-3	8.80E-2
90	4.54E-6	1.05E-5	8.00E-4	1.85E-3	8.50E-2
300	5.07E-6	1.17E-5	5.99E-4	1.38E-3	7.30E-2
500	5.94E-6	1.27E-5	4.55E-4	1.05E-3	6.40E-2
Current MDA ^(b)	3.00E-4 ^(c)	5.00E-5 ^(d)	3.00E-3 ^(c)	N/A	5.00E 0

Footnotes: E-2 = × 10⁻⁴

^(a)Activity median aerodynamic diameter.

^(b)Minimum detectable amount.

^(c)Radiochemistry and alpha spectrometry analyses.

^(d)Inductively coupled plasma-mass spectrometry analyses.

Table 3. Bioassay measurement goals for a class Y natural uranium inhalation intake (1 µm AMAD) resulting in 1 mSv H_{E,50}.

Days after intake (d)	Bioassay measurement				
	Urine excretion		Faecal excretion		Lung burden ²³⁵ U(Bq)
	²³⁸ U (Bq.d ⁻¹)	U (µg.d ⁻¹)	²³⁸ U(Bq.d ⁻¹)	U (µg.d ⁻¹)	
1	2.90E-2	2.45E 0	6.76E-1	5.72E 1	1.28E-1
7	1.39E-3	1.18E-1	1.05E-1	8.92E 0	8.94E-2
30	4.25E-4	3.60E-2	1.73E-3	1.46E-1	8.70E-2
90	2.48E-4	2.10E-2	1.59E-3	1.34E-1	8.16E-2
300	2.38E-4	2.01E-2	1.19E-3	1.00E-1	6.60E-2
500	2.35E-4	1.99E-2	9.00E-3	7.61E-2	5.44E-2
Natural Background (ICRP-23)	6.00E-4	5.00E-4	1.70E-2	1.40E 0	Negligible
Current MDA**	3.00E-4 Bq	6.00E-2 µg	5.00E-3 Bq	3.00E-3 µg	5.00E 0 Bq

*Active median aerodynamic diameter.

**Minimum detectable amount.

ers' geographic location, diet and water source. Again, the conclusion is that workplace monitoring must be sufficiently robust to identify relatively small magnitude intakes so that special bioassay can be initiated immediately after the intake. In addition, routine monitoring programmes for uranium must be carefully designed with environmental baselines and the expected 'normal' variation carefully developed.

Similar challenges, such as identifying small magnitude intakes, face bioassay for ^{238}Pu oxide and thorium oxides. In addition, the future decommissioning of former graphite-moderated reactors raises the possibility of exposure to relatively insoluble forms of ^{14}C . Bioassay for such material has not been seriously investigated.

INDIRECT RADIOBIOASSAY — PRESENT AND FUTURE

In vitro bioassay, or indirect radiobioassay as it will be referred to here, utilises radioanalytical techniques to isolate, identify and quantify the amount of radioactive materials in excreta and biological substances. The results from these measurements provide the internal dosimetrist with information which may be used to assess radiological exposure to the worker. Ideal indirect radiobioassay produces unbiased and precise results, with detection limits below action levels, expeditiously and at a reasonable price.

INDIRECT RADIOBIOASSAY — PRESENT

A range of minimum detectable concentrations (MDCs) achieved with current measurement and analytical techniques for commonly expected radionuclide of interest is presented in Table 4 by radionuclide of interest for the two most common matrices (urine and faeces). Also shown in the table is the general analytical technique and the measurement instrument(s) (including count time) normally employed for the radionuclide. A brief description of each measurement system follows the table. The MDCs for other body fluids would be of approximately the same magnitude. The term minimum detectable concentration is used here to signify that in most cases chemical separations are an essential factor in the determination of the radionuclide of interest. The exceptions are direct counting by liquid scintillation or gamma spectrometry. A laboratory may determine its ability to achieve the MDC goal using the equations in the draft American National Standards Institute (ANSI) standard 'Performance Criteria for Radiobioassay', N 13.30⁽⁵⁾.

From Table 4, the lower MDC value for ^{239}Pu in urine is 0.3 mBq. This value is nearly 70 times higher than the measurement goal of $4.54 \text{ E-}6 \text{ Bq.d}^{-1}$ for 90 days after intake as shown in Table 2. Method improvement for the same radionuclide of interest in the faecal matrix may reduce the MDC from 3 to 0.3 mBq. This is still a factor of 3 higher than the goal MDC for the faecal

matrix at 90 days after intake as shown in Table 2. It will be time consuming and difficult to improve the MDC for routine operations, even for faecal, without extensive effort and expense. Thus, it is extremely important that the field of measurement programme provide early warning that special bioassay analyses are requested rather than establishing a more frequent routine bioassay programme.

Radioanalytical techniques range from direct counts of the radioactive materials using gamma ray spectrometry to the use of complex chemical separation techniques and sophisticated measurements systems. Chemical separation techniques which are frequently used include ion exchange, solvent extraction and precipitation (individually or sequentially). Measurement systems commonly used are: liquid scintillation spectrometry, alpha spectrometry, gas proportional counting, gamma spectrometry, fluorimetry, kinetic phosphorimetry (KPA) and alpha scintillation counting. Extraction chromatography⁽⁶⁾ using new resins promises that additional radionuclides of interest can be separated with a single separation.

The capability of an analytical method to achieve the desired bias and precision and MDC requirements depends on several parameters: counting efficiency, count time, chemical recovery, background of the detector, background contributed by the analytical process and the degree to which the chemical procedure removes chemical and radiochemical interferences (matrix interference).

The counting and measurement instruments described above, with the exception of the KPA⁽⁷⁾, have been in use for several years. With improvement in computer technology and associated software many of these instruments are available with advanced data management techniques including data collection and analysis and calculation of the final result. Detector systems have also been improved. Examples are the production of low background ($\sim 3 \text{ mBq.s}^{-1}$) external gas proportional detectors, high purity germanium detectors with improved resolution and the passive implanted planar (PIP) detectors used for alpha spectrometry. The PIP show a marked increase in counting efficiency over the surface barrier detector.

A laser fluorimeter for the determination of uranium was introduced about 15 years ago, followed by the KPA which was commercially produced nearly 5 years ago. These instruments enhanced the determination of uranium by providing more sensitive and precise measurements.

INDIRECT RADIOBIOASSAY — FUTURE

A relatively new technique that holds much promise for indirect radiobioassay is the inductively coupled plasma-mass spectrometer (ICP-MS)⁽⁸⁾. The ICP-MS uses an inductively coupled plasma as an ion source for a mass spectrometer. The basic units of an ICP-MS sys-

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tem include a sample introduction device, plasma, plasma-mass spectrometer interface, ion focusing-ion filtering system, detector and the data acquisition-data handling system. This system has been used for the determination of ⁹⁹Tc, ¹²⁹I, ²³⁷Np, ²³⁹Pu and ²⁴⁰Pu in a variety of environmental matrices, uranium in urine and thorium and uranium in various biological matrices. Radiochemical separations are often required and depending on the radionuclide of interest, an electrothermal vaporisation unit is essential to achieve low sensitivities. This system is attractive because the measurement process is much faster than, for example, the long counts necessary for alpha spectrometry. In addition it can separate ²³⁹Pu and ²⁴⁰Pu. However, for those radionuclides with half-lives less than 1×10^4 years, radiological hazards will become a concern because of the

number of atoms necessary for the mass measurement. Minimum detectable amounts which appear achievable for ²³⁹Pu range from 5–50fg. Testa *et al*⁽⁶⁾ quote a research article which indicates that a MDC of approximately 0.02 mBq (9 fg) is achievable for ²³⁹Pu in aqueous samples with ICP-MS. This is an approximate order of magnitude better than alpha spectrometry, but is still an order of magnitude higher than the bioassay measurement goal. Much effort is needed to even achieve 0.02 mBq in routine operations.

There is continual effort to improve analytical laboratory procedures. It is hoped that some of the new developments discussed below may hold promise for future improvement in bioassay measurements.

The *Analytical Chemical Application Reviews* for 1993^(9,10) and other recent journals include references to

Table 4. *In vitro* bioassay analysis capabilities.

Radionuclide of interest	MDC RANGE		Analytical techniques	Measurement systems*	Count times (s)
	Urine (l ⁻¹)	Faecal (per sample)			
³ H	37-570 Bq	Not Available	Direct and/or distillation	LS	6E2-1E4
¹⁴ C	37-370 Bq	3.3-33 Bq	Direct and/or chemical separation	LS or GPC	1E2-8E3
⁶⁰ Co	0.25-2.5 Bq	0.25-2.5 Bq	Direct count	GS	5E2-6E4
¹³⁷ Cs	0.25-2.5 Bq	0.25-2.5 Bq	Direct count	GS	5E2-6E4
^{89,90} Sr	0.5-5 Bq	0.74-7.4 Bq	Chemical separation	GPC	8E2-1E3
²²⁶ Ra	3.7-37 mBq	25-150 mBq	Chemical separation	ASC or GS	1E2-2E3
²¹⁰ Po	3.7-37 mBq	0.2-6 Bq	Spontaneous deposition	AS	1.2E4-1.5E5
^{228,230,232} Th	1.5-3.7 mBq	37-180 mBq	Chemical separation	AS	1.2E4-1.5E5
^{234,235,238} U	0.3-2.2 mBq	5-37 mBq	Chemical separation	AS	1.2E4-1.5E5
²³⁷ Np	0.3-2.2 mBq	1.7-37 mBq	Chemical separation	AS	1.2E4-1.5E5
²³⁸ Pu and ^{239,240} Pu	0.3-2.2 mBq	3-37 mBq	Chemical separation	AS	1.2E4-1.5E5
²⁴¹ Am	0.3-2.2 mBq	13-37 mBq	Chemical separation	AS	1.2E4-1.5E5
U-mass	0.025-5 µg	0.3-8 µg	Chemical separation	KPA, F	

ASC: alpha scintillation counter. This counter consists of a scintillation counting chamber (phosphor coated on the inside) with a clear silicon window, photomultiplier tube, preamplifier, high voltage supply and scaler.

AS: alpha spectrometer. An alpha spectrometer consists of a low background (~2 counts per 60,000 seconds), high resolution (~24-35 keV depending on the detector size), silicon surface barrier or passive implanted planar detector, vacuum system, bias supply, preamplifier, shaping and stretching amplifier, biased amplifier, discriminator, analogue-to-digital converter (ADC) and multichannel analyser (MCA).

F: Fluorimeter. This instrument measures the fluorescence of a fused disc of sodium fluoride, lithium fluoride, and uranium compound exposed to ultraviolet light. The intensity of the fluorescence is proportional to the uranium concentration.

GS: gamma spectrometer. This system includes a lithium-drifted germanium (GeLi) or high-purity germanium (HPGe) detector (p- or n-type), generally with a full width at one-half the peak maximum (FWHM) Less than 2.2 keV at 1332 keV, lead shielding, high voltage power/bias supply, preamplifier, ADC and amplifier with a MCA.

GPC: Gas proportional counter. The gas proportional counter may utilize an end-window Geiger-Mueller tube or an internal or external gas-flow chamber for the detector which is constructed from material free from detectable radioactivity. The detector is shielded and is associated with a power supply, amplifier and scaler. The system may be equipped with a cosmic guard to further reduce backgrounds.

KPA: Kinetic phosphorescence analyser (Pulsed laser phosphorimetry). This instrument analyses the phosphorescence signal produced from laser excitation of complexed uranium(VI). The signal is proportional to the quantity of uranium present.

LS: Liquid scintillation spectrometer. The sample is mixed with a liquid scintillator and the radiation energy is expended in the ionisation and excitation of the solvent. The energy is transferred to the solute and re-emitted as photons, which are detected with one or more phototubes. The subsequent signal may be routed through one or more amplifiers and recorded with a counter such as a single channel or multichannel analyser.

N.B. 6E2-1E4 means $6 \times 10^2 - 10^4$.

measurement methods such as accelerator mass spectrometry⁽¹¹⁾, thermal ionisation mass spectrometry⁽¹²⁾, beta spectrometry⁽¹³⁾, neutron activation⁽¹⁴⁾, fission track analysis⁽¹⁵⁾, and photon electron rejecting alpha liquid scintillation spectrometry^(16,17). Applications are shown for radionuclide determinations but not necessarily in excreta or other biological matrices. It remains to be seen whether these methods will become universally available and cost effective.

The measurement system and the required MDC dictate the degree of sample preparation. As shown in Table 4, most determinations require that the radionuclide of interest be chemically separated from the sample matrix before counting. Most initial sample preparation methods still rely on classical muffle furnace, wet ash and precipitation techniques. Although total sample dissolution utilising microwave energy is being widely used for the analysis of environmental matrices, the sample size most microwave digestion systems are capable of handling is still too small for most bioassay needs of today. However, new or improved detection systems should allow the use of a smaller sample size and thus less effort⁽¹⁸⁾.

For existing measurement systems such as those described above, separation methods may be necessary for eliminating chemical and radiochemical interferences (depending on the final measurement technique) and reducing a large sample to a reasonable size while still achieving a low MDC. Many indirect radiobioassay chemical separation methods such as ion exchange, solvent extraction and precipitation, are labour intensive and time consuming. There is continual effort to improve the existing methods and/or develop alternatives that are more efficient and robust. A robust radioanalytical method would be simple to use, capable of removing chemical and radiochemical interferences, have few chemical steps, not be sensitive to small changes in reagent concentrations, not use hazardous chemicals and have very little chemical and/or radiochemical waste. New separation methods, such as those using crown ethers⁽¹⁸⁾, have been developed and are now seeing limited use.

DIRECT BIOASSAY MONITORING EQUIPMENT — PRESENT AND FUTURE

The radiation detection systems used to perform *in vivo* measurement of radioactive material in humans are designed to detect photons emanating from the body. The different systems can be broadly classified according to the type of detectors used, whether they detect high or low energy photons and how the detector(s) are configured. These systems provide a direct indication of the presence of radioactive material in the body. The data interpretation of *in vivo* measurement results requires fewer assumptions compared to excreta or workplace estimates for assessing doses. The MDA for each of the following techniques is presented in Table 5.

HIGH ENERGY PHOTON DETECTION

NaI and intrinsic germanium detectors are the two most commonly employed detectors for the quantitative measurement of radioactive materials that emit high ($E > 200$ keV) energy photons.

The NaI detectors are physically large; typical crystal dimensions for a cylindrical detector being 23.8 cm diameter by 10 cm thick and for a rectangular detector 10 cm by 10 cm by 40.6 cm long. They offer high detection efficiency for high energy photons and are extremely durable. The resolution available is much less than the resolution achieved with solid state detectors.

Intrinsic germanium detectors have essentially replaced lithium-drifted germanium detectors in the past five years. The intrinsic germanium detector can be warmed to ambient temperature then brought back to liquid nitrogen temperatures for operation. These detection systems typically have full width at half maximum (FWHM) resolution of less than a few keV at 663 keV. Crystal sizes are small compared to NaI but the technology has progressed to the point where detection efficiencies for the larger crystals equal or exceed that of a 7.5 cm by 7.5 cm NaI detector. The detectors require a liquid nitrogen filling system that can reliably fill the detectors at set frequencies.

In general, the sensitivities for the measurement of radioactive materials that emit high energy radiations

Table 5. Typical minimum detectable activities (MDA) for DOE Hanford Site *in vivo* measurement systems.

Nuclide	Whole-body count		Lung count Ge Detectors ^(c) MDA (Bq)
	NaI detectors ^(a) MDA (Bq)	Ge detectors ^(b) MDA (Bq)	
⁴⁰ K	400	300	NA ^(d)
⁵⁴ Mn	200	40	NA
⁵⁹ Fe	300	70	NA
⁶⁰ Co	200	40	NA
¹³¹ I	100	60	NA
¹³⁷ Cs	100	40	NA
¹⁵⁴ Eu	400	100	NA
²⁰⁸ Tl	100	30	NA
²¹⁴ Bi	400	100	NA
²³⁴ Th	NA	NA	100
²³⁵ U	NA	NA	7
²⁴¹ Am	NA	NA	10

^(a)200 s measurement using vertical array of five cylindrical detectors (9.375 × 4, 11.5 × 4, 9.375 × 4, 9.375 × 4, 9.375 × 4 in²).

^(b)1200 s measurement using four (59.4, 66.3, 69.0, 77.1 per cent) coaxial detectors (as related to a 3 × 3 in² NaI crystal using ⁶⁰Co 25 cm from the end cap of the detector) positioned below a supine subject.

^(c)1200 s measurement using six planar Ge (each 51 mm diameter × 13 mm thick) detectors placed over the lungs.

^(d)Not applicable.

are very good and the detection capabilities represent a small fraction of the annual limits on intake.

LOW ENERGY PHOTON DETECTION

The detection of low energy photons ($E < 200$ keV) is commonly done with detectors containing thin scintillation crystals to optimise the detection efficiency. Phoswich detectors offer excellent background rejection capabilities utilising the different decay times of the CsI and NaI(Tl) phosphors. NaI detectors have high backgrounds but require less complex electronics since gating is not required as with the phoswich to separate the CsI and NaI pulses.

Contemporary solid state detectors typically contain small crystals of germanium or silicon for low energy applications. A 2000 mm² area detector 20 mm thick is a typical crystal size used in detectors for measuring actinides in the lungs. Crystals up to 4000 mm² in area and approximately 30 mm thick will soon be commercially available in the US. Testing of prototypes of the larger detectors have shown that lower detectable activities can be expected compared to the 2000 mm² area detectors currently being used. Thin intrinsic germanium crystals offer excellent resolution and relatively low background count rates over a wide range of energies. Lithium drifted silicon detectors have excellent resolution for low energy X rays but must be kept at liquid nitrogen temperatures at all times to prevent diffusion of the lithium. Both the silicon and germanium detectors require a reliable supply of liquid nitrogen to be delivered for operation.

The detector sensitivities for actinides that emit low energy photons (e.g. ²⁴¹Am and ²³⁹Pu) are significantly higher than for high energy measurements and can represent doses in excess of the annual occupational limits for actinides such as ²³⁹Pu. For aged Pu mixtures the ²³⁹Pu activity is commonly derived from measurements of the ²⁴¹Am activity based on assumptions regarding the Pu to Am ratio of the mixture. The calculated detection level for ²³⁹Pu is lower based on this method assuming the uncertainty in the ratio is zero. However, the uncertainty in the ratio is frequently not known and not factored into the determination of the MDA.

SYSTEM DESIGNS

System designs will vary depending on the reason for the measurements. Maximum sensitivity with minimum count times is desired for routine measurements of large numbers of subjects. For quantifying the amount of material in the body maximum accuracy is required and a system that has a small dependence on the activity distribution may be warranted since this is usually the largest uncertainty in the measurement process. Arrays of detectors placed in close proximity to the body provide the best sensitivity for detecting low levels of radioactive material in the body. However, the accuracy

of these types of systems can depend strongly on the activity distribution in the subject. A single detector that scans the body can also provide adequate sensitivity at less cost compared to an array. Arrays of detectors placed a fixed distance from the subject or above and below the subject will be less dependent on the location of the activity in the person. The standard one and two metre arc counting geometries can be used effectively to minimise the dependence of the results on the activity distribution for high energy photons. Due to the inverse square law and other geometry factors, the sensitivity of the system will limit the applicability for quantifying low levels of activity.

The design of *in vivo* counting systems generally includes shielding to reduce the background count rates and optimise the capability for a system. This can range from massive shielded cells with 15 cm to 41 cm thick walls, floors and ceilings made of pre-World War II plate steel with interior graded shields such as are used at many US DOE facilities to local shielding of the detectors with lead used in commercial systems.

For the quantification of actinide materials in the lung it is important to account for the photon absorption in the tissues overlying the lungs. Estimates of this tissue thickness are commonly calculated based on weight to height ratios. More accurate tissue thickness estimates can be obtained using diagnostic ultrasound equipment. Magnetic resonance imaging (MRI) is also being used to evaluate chest wall thickness and in special situations may have advantages over ultrasound. However, from a practical sense this technique may never be routinely used in operational *in vivo* counting programs. The radiation interaction characteristics of the calibration phantom are assumed to represent each individual adequately.

The measurement of radioactive material in a source organ or tissue may require adjustments based on the contribution to the count rate from activity in other organs or tissues. Collimating the detector(s) can limit the solid angle subtended by the detector(s) to discriminate against extraneous counts. But the collimation will decrease the detection efficiency at the same time. Count rate contributions from other organs and tissues can be approximated using phantom measurements. For example, the count rate contribution to a lung count from activity in the liver can be estimated from lung measurements of a phantom containing liver activity and no activity in the lungs.

THE FUTURE

The use of detectors with larger intrinsic germanium crystals, gas scintillation proportional counters and ambient temperature solid state detectors offer promise for the future development of *in vivo* monitoring equipment. As crystal growth technology develops, larger area planar germanium crystals will improve the detection efficiency for low energy photons and the larger

volume coaxial germanium crystal designs will continue to improve detection efficiency for high energy photons.

Gas scintillation proportional counters offer large surface areas for low energy photon detection and do not require liquid nitrogen for operation. At this time it is not clear if the detector sensitivity will be adequate for *in vivo* actinide monitoring. It is possible that the large surface area may allow only one or two detectors to be used for actinide lung monitoring compared to the six or more germanium detectors frequently used at present. An ambient temperature solid state detector with resolution capabilities comparable to current intrinsic germanium detectors may be achievable with a crystal made of a material such as gallium arsenide. The capability of manufacturing high purity gallium arsenide crystals with adequate dimensions and achieving a uniform electric field remain major challenges to obtaining acceptable resolution. Cadmium telluride is currently used in small monitors for wound measurements of low energy photons and could someday be developed into larger detectors. Depending on market forces the development of ambient temperature solid state detectors could proceed over the next decade to the point where they could be used for *in vivo* monitoring.

WORKPLACE MEASUREMENTS

One of the challenges for internal dosimetrists that has been neglected to a great extent in the past is the establishment of a strong cooperative relationship with the field health physicist. However, with a combination of changes in internal dosimetry including the requirement to add external and internal dose to control total lifetime dose the costs of enhanced bioassay programmes and worker cooperation; the use of the field health physicist as part of the facility dosimetry team is extremely important.

Workplace measurements that provide the most support for internal dosimetry programmes fall in two categories: (1) those that are a surrogate for bioassay and *in vivo* measurements (e.g. air monitoring measurements) and (2) those for which derived investigated levels (DILs) have been established to trigger investigations and perhaps special bioassay programmes.

In the US, the newly revised *Standards for Protection Against Radiation* of the Nuclear Regulatory Commission apply to all power reactors and also to by-product, special nuclear and source materials licenses in 29 of the 50 states. Under this regulation, it is permissible to assess internal dose based on representative air monitoring^(19,20).

Thus, workplace air measurements may be used to assess internal doses using

$$E = \frac{\bar{C} t}{P \times \text{DAC}_{90,\text{adj}} \times 2000 \text{ h}} \times 20 \text{ mSv}$$

where

E	denotes effective dose (mSv);
\bar{C}	denotes the average representative radionuclide concentration in air (Bq.m^{-3});
t	denotes the time (h) the worker breathed air at concentration \bar{C} ;
P	denotes the protection factor for the respirator used, if any $1 \leq P \leq 1000$;
$\text{DAC}_{90,\text{adj}}$	denotes the appropriate 1990 ICRP derived air concentration for the radionuclide (Bq.m^{-3}), adjusted for particle size (note: currently, the US uses the 1979 ICRP 30 DACs and 50 mSv in place of the 1990 DACs and 20 mSv adjusted for particle size); and
2000 h	is the number of hours in a working year.

While the technology for breathing zone air monitoring has not changed dramatically in recent years, recently, it has been shown that very significant gains in apparent detection capability can be made by optimising the use of counting data in the computation of $\bar{C}^{(21,22)}$. By pooling all data for a year's sampling for one worker, pooled decision levels (DLs) and minimum detectable average concentrations ($\text{MDC}\bar{C}$ s) can be used. These levels can permit a reduction in the DL and $\text{MDC}\bar{C}$ by as much as \sqrt{n} , where n is the number of measurements made in a year. In one sample case, a breathing zone air monitoring programme capable of detecting 9.8 mSv without pooled analysis, was shown to be capable of detecting 0.6 mSv when uncensored data were properly pooled. This result is well under the goal of 1 mSv.

The second, and perhaps more important, use of workplace monitoring is to trigger investigations and special bioassay programmes in an effort to minimise detection capability problems due to technology shortfall in routine bioassay programmes. Workplace measurements useful in this context are air monitoring (both general and breathing zone), surface contamination measurements, skin contamination surveys of workers, whole or partial-body contamination monitoring results, self-survey results, counts of nasal swabs and observations by workers or supervisors of off-normal conditions that may result in loss of containment of radioactive materials. Alone, or in combination, these techniques lead to prompt detection of intakes that might not otherwise be detected by routine bioassay measurements. Perhaps the greatest progress in the last decade has been in the detection of containment on or in the worker by the use of new, large detector area partial body personnel contamination monitors at exits of radiologically controlled areas. These have been developed because portable survey instruments have proven to be inadequate due to (1) survey time involved,

(2) lack of skill and (3) small number of cases of contamination detected.

The problem of detecting contamination with portable survey instruments was addressed by Clive Dray⁽²³⁾. He noted that the surface area of the body of the average person is 18,000 cm², so if a 100 cm² detector is used to observe each part for just one second, monitoring would take 3 min, not the 3 s usually taken. If an alpha detector is placed 3 cm from a radium source it will detect nothing due to absorption of the alpha particles by the air. If a 100 cm² beta detector is moved 5 cm from a spot of ³⁵S contamination the response will change by a factor of 10. This movement could change the reading from 5 to 50 counts per second and even three centimetres makes over a factor of two change. The smaller the probe the worse the effect for a point source. As a result of this, the NE Technology Limited developed a personnel contamination monitor using gas flow proportional counter detectors which will cover 50% of the surface of the body at one time. The theoretical maximum efficiency for detecting 80 Bq of ⁶⁰Co ranges from 14.8% for a detector area of 452 cm² to 16.3% for a detector area of 829 cm².

In a paper on performance of personnel contamination monitors prepared by M. Cox *et al*⁽²⁴⁾, the capability of several personnel monitors was discussed. Using multiple gas flow proportional or plastic scintillator detectors to cover major portions of the body (e.g. one half), the following measurement capabilities have been achieved:

- alpha/beta radiations 37 Bq per 10 s
- beta/gamma radiations 74 Bq per 10 s

Whether or not they trigger special bioassay measure-

ments, workplace measurements may provide information on the time at which an intake occurred or the time course of intake for protracted intakes. For most intakes, time course of intake is a variable that must be known to permit correct interpretation of bioassay data. Workplace measurements can also help provide a diagnosis of containment or control problems to manage future intakes. Particle size measurements made in the workplace may be needed as input to internal dose assessments as well as particle dissolution studies for assignment of solubility classes D, W and Y.

For measurements of radon progeny in mines and other work places, measurements of potential alpha energy concentration, equilibrium ratios and the ultra-fine or unattached fraction are needed. These measurements are crucial in determining dose to the various portions of the respiratory tract under either old or new ICRP respiratory tract models since *in vivo* or *in vitro* bioassay is impractical for these short-lived radionuclides.

SUMMARY

An attempt has been made to characterise the current and future status of *in vivo* and *in vitro* measurement programmes coupled with the associated radioanalytical methods and workplace monitoring. Developments in these areas must be carefully integrated by internal dosimetrists, radiochemists and field health physicists. Their goal should be uniform improvement rather than to focus on one specific area (e.g. dose modelling) to the neglect of other areas where the measurement capabilities are substantially less sophisticated and, therefore, the potential source of error is greatest.

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