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TECHNIQUES FOR MONITORING PLUTONIUM IN THE ENVIRONMENT

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July 1978

Prepared for the U. S. Department of Energy
under Contract W-7405-ENG-48

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Techniques for Monitoring Plutonium in the Environment

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1. INTRODUCTION

Plutonium is one of the principal materials of both commercial and military nuclear power. It is produced primarily in fission reactors that contain uranium fuel, and its importance arises from the fact that a large portion of the plutonium produced is fissile: like uranium 235, the mass 239 and 241 isotopes of plutonium can be caused to fission by neutrons, including those with low energy. Because such fission events also release neutrons, substantial amounts of energy can be extracted from plutonium in a controlled or an explosive nuclear chain reaction.

Now that commercial nuclear reactors provide a noticeable fraction of United States (and world) electrical energy, these reactors account for most plutonium production. For the most part, this material now remains in the irradiated fuel after removal from reactors, but should this fuel be reprocessed, the plutonium could be recycled to provide part and even most of the fissile content of fresh fuel. For the current generation of water-cooled reactors, the amount of plutonium to be recycled is substantial. In fast breeder reactors, designed to produce more fissile material than they destroy, considerably larger quantities of plutonium would be recycled. In other types of advanced reactors, particularly those which depend heavily on thorium as the material from which fissile material (primarily uranium 233) is produced, the amount of plutonium to be handled would be considerably reduced.

Because plutonium is a highly toxic substance, great care is taken to contain it at the sites and facilities where it is stored or handled. In addition, it is necessary that devices be available to monitor any releases from these facilities into environmental media and to measure concentrations of plutonium in these media. The radiation protection standards are so strict for plutonium that only small releases and low concentrations can be tolerated. Such considerations, discussed in the next section, require that monitoring instrumentation be extremely sensitive.

The hazard from plutonium arises largely from the alpha particles emitted during radioactive decay. The most sensitive plutonium monitoring devices are based on detection of this radiation and are used for air monitoring and for measurement of plutonium extracted by radioachemical analysis of water, soil, and biological samples. Instruments for area survey or for monitoring of human subjects often utilize the electromagnetic radiation (x-rays or gamma rays) which follow radioactive decay. Less often used techniques for plutonium monitoring include activation and electrochemical methods. Other techniques, such as mass measurement, are possible but not developed. It is worth noting that, for the most part, the techniques for monitoring plutonium may also be used for monitoring other nuclear materials, including transuranics that may be associated with plutonium.

Techniques for monitoring plutonium have been reviewed previously.[1] This article is based largely on work performed in connection with Lawrence Berkeley Laboratory's Survey of Environmental Instrumentation.[2,3] Detailed information on the properties and uses of plutonium may be found in references 4-14.

2. FUNDAMENTAL MONITORING CONSIDERATIONS

2.1 Properties of Plutonium

The plutonium isotopes of greatest importance are those with mass number 238 to 242. Decay properties of these isotopes are given in Table 1. ^{241}Pu has the shortest half-life of these, 13 years; the properties of its decay daughter, americium 241, are also given since survey instruments often monitor this isotope as a plutonium indicator. The other isotopes in Table 1 have half-lives from about 10^3 to 10^5 years, so that after production they persist for long periods of time. However the hazard presented by radionuclides is determined, not only by the toxicity of the nuclides being considered and by the amount produced in reactors, but also by the type of daughters arising from decay of reaction products and by the pathways which radionuclides can take to humans.

Plutonium in commercial nuclear power operations could have several chemical forms. The most probable form for nuclear fuel is the dioxide. Like the uranium dioxide now used as fuel for light-water reactors, PuO_2 is a ceramic that is relatively insoluble; the difficulty with which a sample can be brought into solution makes it relatively inaccessible to human beings; the primary hazard from the alpha-emitting isotopes, those of most concern, arises from the possibility of inhalation. However, low solubility also introduces difficulties for monitoring methods which require dissolution of a bulk sample. On the other hand, in some circumstances plutonium may be present in a more soluble form than the dioxide; in this case, it can become accessible to humans via water-based pathways and, moreover, it can cause damage at lower concentrations.

The dependence of toxicity on solubility arises from differences in uptake and retention. Airborn soluble plutonium can reach the bloodstream by passage across the lung mucosa; in water it is absorbed from the gastrointestinal tract. In either case, it is largely deposited on bone surfaces, where its potential for causing cancer is deemed greater than that of radium. Reducing the solubility of the plutonium decreases the amount deposited in bone, so that the critical organ becomes the site of direct exposure, the lung or GI tract; however, for insoluble plutonium deposited in lungs, substantial amounts find their way to the pulmonary lymph nodes, which may therefore be the most sensitive site.

Table 1. Properties of the Plutonium Isotopes*

Isotope	Half-Life	Immediate Daughter (Half-Life)	Main Decay Radiation and Energies
^{236}Pu	2.85 yr	^{232}U(72 yr)	69% 5.77 MeV α 31% 5.72 MeV α
^{237}Pu	0.12 yr	^{237}Np(2.1×10^{6} yr)	electron capture
^{238}Pu	86 yr	^{234}U(2.5×10^{5} yr)	72% 5.50 MeV α 28% 5.46 MeV α
^{239}Pu	2.44×10^{4}	^{235}U(7.1×10^{8} yr)	73% 5.16 MeV α 15% 5.15 MeV α 12% 5.11 MeV α
^{240}Pu	6580 yr	^{236}U(2.4×10^{7} yr)	76% 5.17 MeV α 24% 5.12 MeV α
^{241}Pu	13.2 yr	^{241}Am(433 yr)	β^- (0.031 MeV maximum)
^{242}Pu	3.8 10^{5} yr	^{238}U(4.5×10^{9} yr)	76% 4.90 MeV α 24% 4.86 MeV α
^{241}Am	433 yr	^{237}Np(2.1×10^{6} yr)	86 % 5.49 MeV α + 60 keV γ 13% 5.44 MeV α

*For more detailed information, see ref. 15. For isotopic composition of spent reactor fuel, see Table 3.

See references 4-8 for further discussion.

2.2 Radiation Protection Standards

Consideration of these pathways for plutonium and of the effect that decay can have on humans has led to the radioactivity concentration guides given in Table 2. These specify concentration limits for air and water which would lead to the maximum permissible occupational exposures were workers exposed to these concentrations for 168 hours per week. Practical occupational limits are obtained by dividing these limits by the portion of the week actually worked. On the other hand, guides for individual members of the general public are one tenth those given in the table; for population groups, the guides are lower again by a factor of three.[16] Plutonium particles may be difficult to monitor effectively at such low air concentrations simply because of the discrete nature of the activity: a substantial time may be required for a sampler to collect even a single particle.

For the alpha-emitting plutonium isotopes, the concentration guides are derived from a comparison with ^{226}Ra and its daughters if the plutonium is in soluble form. This leads to the "soluble" limits given in the table and also to a maximum permissible occupational body burden of 0.04 μCi for the (soluble) alpha emitters shown (as compared with 0.10 μCi for ^{226}Ra). The limits for insoluble plutonium are based on more general criteria for dose limitations, such as those which limit whole body exposures to 5 rem per year for radiation workers, 0.5 rem/year for members of the public, and 0.17 rem/year for population groups[16] or which have limits a factor of three higher for specific organs, including the lung. The maximum permissible insoluble plutonium lung burden corresponding to the occupational exposure of Table 2 is 0.016 μCi.

In practice, more specific limits apply to environmental emissions from commercial nuclear power operations. The Nuclear Regulatory Commission, in its review of licensing applications for light-water reactors, uses guidelines which typically limit resulting exposure of members of the general public to approximately 0.005 rem/year, about one percent of the dose limitations given above. In addition the Environmental Protection Agency has begun to develop standards which apply to the nuclear fuel cycle; for uranium-fueled light-water

reactor power plants, the Code of Federal Regulations, Title 40, Part 190, places a limit on total fuel cycle emissions of ^{239}Pu and other alpha-emitting transuranics of 0.5 millicuries per gigawatt (electric)-year. Standards for facilities using recycle plutonium have not yet been developed. More recent EPA regulations apply to exposures in the vicinity of plutonium-contaminated areas.[17] Consideration has been given to formulation of concentration standards for soil.[18] It has also been proposed[19] that waste streams containing more than 10 nCi/gm of plutonium and other long-lived transuranics be treated as transuranic wastes.

The standards for other alpha-emitting activities are often as stringent as those for plutonium. Many of these would have to be monitored at the same locations as plutonium and, indeed, many monitoring techniques apply equally well to these radionuclides. Of particular interest where plutonium is concerned is americium 241, the daughter of ^{241}Pu (a beta emitter). Plutonium 241 has a short half-life (13 years) and, within only a few decades, can contribute an amount of ^{241}Am activity that is comparable to the alpha activities initially present in many plutonium mixtures.

Plutonium does not occur naturally in significant quantities. ^{239}Pu is produced primarily by neutron capture on ^{238}U, followed by beta decay of ^{239}U and its daughter, neptunium 239; heavier isotopes are produced largely by subsequent neutron capture on plutonium itself. ^{238}Pu is produced by a sequence of neutron captures beginning with ^{235}U. Production of plutonium occurs almost entirely in nuclear reactors, whether commercial, research, or military. The isotopic mixture varies substantially, depending on the type of reactor and on the purpose (if any) for producing the plutonium. Table 3 indicates the amount and composition of plutonium that would be produced in commercial nuclear power plants of several types. As will be seen below, the efficacy of plutonium monitoring techniques can depend strongly on the isotopic mixture that is being observed. Many of the currently available techniques have, in fact, been developed for use with weapons grade plutonium, which is produced in such a way as to minimize the ^{240}Pu since this isotope releases relatively many spontaneous neutrons, making efficient detonation of a weapon difficult. As a result, a typical sample consists of the following mixture:[18] 93.6 percent ^{239}Pu,

Table 2. Maximum Permissible Concentrations for Plutonium, Radium, and Americium (168 hour occupational[a])

ISOTOPE		MPC_{air}	MPC_{water}
$^{238,239,240,242}Pu$	soluble	0.6 (bone)	0.05 (bone)
	insoluble	10 (lung)	0.3 (GI)
^{226}Ra	soluble	10 (bone)	0.0001 (bone)
	insoluble	20 (lung)	0.3 (GI)
^{241}Pu	soluble	30	2
	insoluble	10^4	10
^{241}Am	soluble	2	0.04
	insoluble	40	0.3

[a]Concentration limits for occupational exposures may be obtained by dividing the stated MPCs by the percentage of hours in a week actually worked. Limits for individual members of the general public are one tenth the stated MPCs; for populations, the limits are 1/30th the MPCs shown. (For the plutonium and radium alpha emitters, the critical organ is shown.) See Code of Federal Regulations, Title 10, Part 20.

Table 3. Typical Isotopic Composition of Plutonium Extracted from Nuclear Power Plants[a] (each entry gives isotope, mass in kilograms radioactivity in Curies)

Light-water reactor fueled with natural plus recycle uranium

	%	kg	Ci
238	2.4	6	1.0×10^{5}
239	58.4	142	8.7×10^{3}
240	24.0	59	1.3×10^{4}
241	11.2	27	2.8×10^{6}
242	3.9	10	3.7×10^{1}
	100	244	1.2×10^{5} (α) 2.8×10^{6} (β)

Light-water reactor fueled with natural uranium plus recycle plutonium

	%	kg	Ci
238	4.2	41	6.9×10^{5}
239	38.7	380	2.3×10^{4}
240	27.5	270	6.0×10^{4}
241	18.1	178	1.8×10^{7}
242	11.5	113	4.4×10^{2}
	100	982	7.8×10^{5} (α) 1.8×10^{7} (β)

Liquid-metal fast breeder reactor fueled with depleted uranium plus recycle plutonium and uranium

	%	kg	Ci
238	0.1	1.4	2.4×10^{4}
239	71.7	1460.6	9.0×10^{4}
240	25.1	510.8	1.1×10^{5}
241	2.4	48.4	4.9×10^{6}
242	0.8	15.5	6.0×10
	100	2037	2.3×10^{5} (α) 4.9×10^{6} (β)

[a]Yearly masses and radioactivity from fuel reprocessing for 1000 MWe plants (80% plant factor), assuming 150 day decay time, except for LMFBR, which assumes 30 days. Taken from ref. 9.

5.9 percent ^{240}Pu, 0.4 percent ^{241}Pu, 0.01 percent ^{242}Pu and 0.01 percent ^{238}Pu. Note that mixtures from commercial reactors (Table 3) contain considerably more of the isotopes other than 239.

The bulk of the world's plutonium inventory is contained in nuclear weapons, in nuclear reactors, and in materials associated with weapons and reactors. Environmental plutonium originates from these inventories: from nuclear weapons already exploded, from leakage at facilities which handle or process plutonium, or from special events such as the destruction (during takeoff) of a plutonium-bearing satellite. Nuclear explosions, including atomspheric tests, have contributed about 440,000 Ci of $^{238,239,240}Pu$ to the worldwide plutonium inventory (in an activity ratio of about 3:58:39). The burnup during 1964 of a satellite with a power cell deriving its energy from ^{238}Pu decay contributed about 17,000 curies of this isotope to the global inventory. Plutonium handling facilities are known to have released on the order of 10 curies to off-site environments.[20]

The bulk of this inventory was injected into the atmosphere in the mid-1960's, and this fact is reflected in the measured air concentrations and soil deposition rates. For example, during the 1970's concentrations of ^{239}Pu in surface air at New York City have been 0.1×10^{-18} Ci/liter or less, as compared with a peak value of 1.7×10^{-18} Ci/liter for 1963.[20] The latter was one tenth of the soluble plutonium concentration guide for large populations. As indicated, since 1963 measured concentrations have decreased substantially. Even at a given time, concentrations may vary by an order of magnitude from one place to another. Deposition rates have followed a similar pattern with the highest value, 0.6 nCi/m^2, occurring for 1963. Cumulative deposition in surface soils of the United States varies from 0.7 to 2.4 nCi/m^2,[20] depending on the state, except that results are higher for Nevada due to the presence of the Nevada Test Site. Most of this activity is contained in the top 5 cm of soil. Typical concentrations in surface soils are about 0.05 pCi/gm,[20] about 1 percent of the natural activide activity (due to uranium and thorium), which in turn comprises about half of the natural alpha activity in soil. In the vicinity of nuclear facilities or of nuclear testing, concentrations can be much higher. For example, about half of several curies released from the Rocky Flats plutonium facility is present at areal concentrations between 10 and 2000 nCi/m^2.[21] Many portions of the Nevada Test Site have

^{239}Pu soil concentrations of 100's or 1000's of pCi/gm.[22] In such areas, air concentrations can easily reach the general public concentration guides (see ref. 23); resuspension from plutonium-bearing soil may be an important contributor to air concentrations in such areas.[24] Individuals who spend large amounts of time in the vicinity of such facilities have the potential for accumulating the permissible body burden. In practice, only a few radiation workers have received such burdens. General public body burdens of ^{239}Pu were found to be a few pCi in 1970-71,[25] about a factor of 300 below the maximum permissible burden for populations. These burdens were accumulated primarily from inhalation of airborne plutonium; due to low plant uptake[26] concentrations in food are about one millionth the general public limits for water.

2.3 Monitoring Objectives

Monitoring techniques may depend strongly on applicable standards and on the conditions of monitoring. In the discussion that follows, this dependence will often be apparent, but it is useful to note some specific considerations:

1. Because radiation protection standards depend on the environmental medium, the sensitivity required will also vary; on the other hand, the basic monitoring technique can depend on medium because the range of the principal radiations associated with plutonium depends on the density of the medium.
2. The sensitivity sought often determines the time required to make a measurement; this time may be substantially lengthened if the technique requires chemical separation or if it is sensitive to other radionuclides than plutonium.
3. Both sensitivity requirements and time limitations depend on the conditions of monitoring—whether occupational or general public and whether routine or emergency.

A severe requirement on monitoring sensitivity is imposed by the fact that the radioactivity concentration guide for air in areas accessible to members of the general public is 6×10^{-17} Ci/ℓ for soluble plutonium, the more restrictive form. A liter of air with this concentration will contribute only 2×10^{-16} decays per second, so that many liters have to be sampled to achieve useful counting rates.

3. AIR MONITORING

The alpha particles associated with decay of plutonium isotopes are the signature used in monitoring for airborne plutonium. The universal technique is to collect the activity by pumping air through a filter. This filter typically collects at least 99 percent of the plutonium in the sampled air. Alpha particles may then be detected as they are emitted from this medium (as suggested in Figure 1) or the plutonium may be chemically separated from the filter medium and deposited on a substrate suitable for more precise measurements.

Fundamental considerations in these techniques are the alpha spectra from plutonium decay as well as the spectra from interfering activities. Figure 2 shows alpha energies and relative strengths for each of the plutonium mixtures of Table 3. It also indicates the position of important background alpha groups arising from daughters of radon 222, part of the ^{238}U decay series. The less important radon 220 (also called "thoron") daughters, part of the thorium 232 decay chain, are also shown. Isotopes of polonium are the most serious background since they attach themselves to particulates, which are then collected on air filters. Radon typically occurs at concentrations of 0.01 to 1 pCi/liter* in surface air,[25] far above the concentration guides for plutonium. The general public concentration guide for radon is 1 pCi/liter, as compared with the soluble plutonium guide of 6×10^{-5} pCi/ℓ. Because of the comparatively high ambient concentration of radon daughters, measurement of the total alpha activity can ordinarily only put an upper limit on plutonium concentrations.

The more useful air monitoring techniques discriminate against background alpha activities, particularly the radon daughters. The remainder of this section treats three techniques which leave the collecting filter intact: direct alpha spectroscopy, lifetime analysis (including holdup), and associated α-β discrimination. Radiochemical analysis of filters is treated immediately thereafter.

3.1 Direct Alpha Spectroscopy

Because filters do not collect radon gas efficiently, it is clear from Figure 2 that alpha spectroscopy, measurement of the energy of emitted alpha particles, can discriminate against natural background, provided

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*Concentrations may be considerably higher inside structures built with concrete and similar materials; see ref. 25.

the monitoring system has sufficiently good energy resolution. In a typical system, a semiconductor detector is used to measure the energy and--because of the detector arrangement for direct measurement from air filters--resolution is hundreds of keV, rather than the tens of keV often available from such detectors. As a result, the 6.0 MeV groups from ^{218}Po (also known as radium A) can interfere with measurement of plutonium, even of the ^{239}Pu 5.1 MeV alpha groups; this is indicated in figure 3, taken from early work.[27] The alpha activity of commercial mixtures of plutonium, and also of ^{241}Am, would be even more difficult to measure because the dominant alpha groups are at 5.5 MeV, even closer to the ^{218}Po (Ra A) group. The most useful plutonium monitoring systems have some method of discrimination, however crude.

Monitoring systems from commercial manufacturers are available both as continuous monitors, where the detector system operates while the air sampling occurs, and as off-line monitors, where a filter from a separate sampling unit is inserted into a counting instrument. Sampling rates are typically 50 to 100 ℓ/m. The filter is most often 47 mm diameter, and the detector is usually a large area (up to 750 mm^2) diffused junction or surface barrier detector, arranged as suggested in Fig. 1. Commercial units are usually small and self contained, except for the vacuum pump. Rather than accumulate an energy spectrum such as Figure 3, they ordinarily count the alphas within some preset energy "window" corresponding to the isotope(s) of interest. Many units compensate for the presence of a portion of the ^{218}Po decays in this window (due to various energy degrading processes), often by making a subtraction proportional to the number of counts in a ^{218}Po window. Such units can detect a 40-hour occupational concentration limit (2×10^{-15} Ci/l) in a few hours.[28] (In the event that daughters of ^{220}Rn, also called thoron, present significant background, the same set of windows may be used for background compensation. The primary "thoron" interference arises from decay of bismuth 212, with alpha particle groups at 6.05 and 6.09 MeV, close to the ^{218}Po group.)

Because of the relatively poor resolution of such systems, the semiconductor detector subsystem may be replaced by a gas proportional

counter. This may be particularly useful for large filter areas. On the other hand, it is possible to improve the resolution and sensitivity of semiconductor systems by using evacuated counting chambers to reduce the energy spread of detected particles caused by energy loss in air, and by using more sophisticated data analysis techniques, including lifetime analysis, to discriminate against radon daughters.[28] Finally, the presence of other alpha-emitting species, such as curium, can complicate these low resolution techniques by contributing alphas to either the foreground or background windows.

3.2 Lifetime Analysis

The fact that the radon daughters detected in plutonium measurements are very shortlived can be used to discriminate against them. The sequence of interest is:

$$\underset{\text{(RaA)}}{^{218}\text{Po}} \xrightarrow[3\text{ min.}]{\alpha} \underset{\text{(Ra B)}}{^{214}\text{Pb}} \xrightarrow[27\text{ min.}]{\beta} \underset{\text{(Ra C)}}{^{214}\text{Bi}} \xrightarrow[20\text{ min.}]{\beta} \underset{\text{(Ra C')}}{^{214}\text{Po}} \xrightarrow[164\ \mu\text{ sec.}]{\alpha}$$

The most primitive time discrimination technique simply holds up counting in a spectroscopic system until a large portion of the ^{218}Po has decayed. Each half hour of holdup reduces the amount of ^{218}Po on a filter by a factor of 1000, provided ^{222}Rn has not been adsorbed. A more sophisticated technique would use lifetime analysis to separate the short-lived (radon daughter) and long-lived (plutonium or other transuranic) activities. In particular, the 3-minute ^{218}Po activity can be distinguished by counting in consecutive time intervals and observing the decrease in the count rate in the plutonium window. However, if the thoron daughters are present in significant amount, a somewhat more complex discrimination scheme must be used because of the presence of ^{212}Bi; for this isotope, a subtraction of the 6.1 MeV alphas that appear in the plutonium window may be performed based on the 8.78 MeV alpha group from ^{212}Po, a daughter of ^{212}Bi. In general, for any mixture of thoron and radon daughters, the incorporation of lifetime analysis into a spectroscopic system can determine the daughter activities,[28-32] substantially improving the sensitivity for detection of plutonium and other long-lived transuranics. A system under development

at Lawrence Livermore Laboratory uses these techniques to measure a 40-hour concentration of plutonium in 30 minutes, an order of magnitude better than the simpler commercial monitoring systems.[28] The LLL system has a continuous filter strip that collects activity on one area and is then stepped into an evacuated counting chamber; while counting occurs, collection takes place on the next portion of filter. The system samples air at 566 liters/minute, and separating the chambers protects the array of semiconductor detectors from the sampled air, which--in a reprocessing plant--could be highly corrosive. Evacuating the counting chamber improves the energy resolution by about a factor of 5. The counting period is divided into two parts to discriminate against ^{218}Po counts in two windows: one for plutonium and americium (4.8 to 5.5 MeV) and one for curium (5.6 to 6.8 MeV). ^{212}Bi is subtracted on the basis of counts in a ^{212}Po window (8.0 to 8.8 MeV) if necessary.

3.3 Associated α-β Measurements

In the associated β method, alpha counts due to radon and thoron daughters are estimated by observation of the β rays that effectively accompany the alpha decay of ^{214}Po and ^{212}Po. These nuclides are formed by β decay and, because of their short half-lives (164 and 0.3 μsec respectively), almost immediately alpha decay. Observation of α-β coincidences may be used to estimate the amount of radon and thoron daughter activity that should be subtracted. Instruments employing this technique[33-36] typically count alphas with a scintillator-photomultiplier counter and betas with a scintillation counter or Geiger-Muller tube. Subtracting a weighted α-β count rate from the alpha count rate (without energy information) may suffer from several difficulties: if there is a large radon background, it will be necessary to subtract two large numbers, yielding a very uncertain smaller number; the relative detector efficiencies must be well known; the relative amounts of members of each decay chain must be known, since--in each case--only one member is measured. Some of these difficulties would be alleviated by incorporating α-β subtraction in an alpha spectroscopic system. However, because of the difficulties in making a reliable subtraction, this method is not widely used.

4. RADIOCHEMICAL ANALYSIS OF AIR, WATER, SOIL, AND BIOLOGICAL SAMPLES

Chemical separation of the transuranic elements from the matrix in which they are found has two advantages: these activities can be separated from one another and from background activities, particularly the uranium and thorium decay chains; for bulk samples, chemical separation removes the matrix that would interfere with detection of alpha particles. Alphas of a few MeV have a range on the order of 10^{-3} cm, and any sample with more than a small fraction of this thickness will seriously degrade energy resolution. Radiochemical analysis is usually followed by electrodeposition of a thin layer of the radioisotopes, permitting high-resolution alpha spectroscopy.

For off-line plutonium monitoring, this approach leads to the highest sensitivity, although it has the disadvantage that substantial time, equipment, and technical competence are required for the dissolution, separation, and counting procedures. Sample preparation depends on the type of sample, but the counting arrangement is relatively independent. Semiconductor detectors of the sort used in the direct alpha monitoring discussed above are employed, but the system may be designed for much higher resolution, even tens of keV, by use of evacuated counting chambers and very thin deposition layers.

A serious condition for the effectiveness of radiochemical analysis is that the plutonium or other activity of interest actually be dissolved. Fired plutonium dioxide, the principle form for commercial nuclear power, is notoriously insoluble.[37] For this reason, vigorous dissolution techniques are often appropriate. A second condition is that the technique be effective in extracting very small amounts of plutonium: a picocurie of LWR plutonium has a mass of about 1 picogram. The efficiency of plutonium extraction and deposition is often checked by the addition to the sample of a "tracer", typically a known quantity of ^{236}Pu or ^{242}Pu, neither of which contributes substantially to the activity of either reactor or weapons plutonium. The alpha groups from these isotopes can be resolved from those of ^{238}, ^{239}, ^{240}Pu in a high-resolution system, providing a measure of the extraction efficiency, assuming the tracer and the "unknown" plutonium activity are equally well dissolved. It is also appropriate that sufficient tracer be added that counting

statistics on the tracer groups does not degrade the precision of the final result.[38]

Two classes of chemical separation are used: total dissolution techniques, which bring the entire sample into solution, and leaching, which selectively extracts the substance(s) of interest from the sample matrix. Leaching is the less vigorous approach, hence more liable to error; this technique should be checked by total dissolution. Samples are typically leached or dissolved with nitric acid plus some combination of hydrofluoric, hydrocholric, and perchloric acids. Specific elements are often extracted from the resulting solution by ion exchange techniques. Carrier solutions are then prepared for electrodeposition onto a planchet which may be observed in an alpha spectrometer.[39-41] Also used is a total dissolution method for soil samples which, after an initial treatment with nitric and hydrofluoric acid, utilizes a high-temperature "fusion with anhydrous potassium fluoride" and subsequent "pyrosulfate fusion" to dissolve the sample completely; separation procedures lead to three fractions ready for electrodeposition: thorium; protactinium, uranium, neptunium, and plutonium; americium, curium, and californium.[42]

With careful high-resolution alpha spectrometry, positive identification of particular radionuclides is possible with only a few counts in the corresponding alpha peak(s). For an hour-long counting period this implies minimum sensitivity on the order of 0.2 pCi, the amount that an air filter collects in an hour at one liter/sec if plutonium is present at the general public concentration limit of 6×10^{-17} Ci/liter. In contrast, the alpha air monitors described in Section 3 have sensitivities on the order of one hour at the occupational concentration of 2×10^{-15} Ci/liter, corresponding to a collected activity of 8 pCi or more. Longer counting periods, of course, improve the sensitivity of radioachemical analysis. For media other than air, the available sensitivity is more than adequate: the general public concentration limit for plutonium in water is 50,000 pCi/liter. In any case, the primary disadvantages to radioachemical analysis are the time, equipment, and personnel required. For many applications, waiting many hours or days for analytical results is not practical.

5. FIELD SURVEY AND PERSONNEL MONITORING

In many circumstances, a relatively rapid determination of the radionuclide content of a sample or an area is required. This ordinarily precludes complex chemical analysis; on the other hand, direct measurement via alpha particles is impractical because of the short range of alpha particles in matter. The electromagnetic radiation which often follows alpha decay is a much more appropriate signature because it is less seriously degraded by passage through bulk material. This radiation includes both x-rays and gamma rays, emitted from decay daughters left in an atomic or nuclear excited state.

Atomic excitations of uranium, the daughter of plutonium alpha decay, yield L x-ray lines at 13.6, 17.3, and 20.2 keV,[43] which may be used for monitoring plutonium since they follow 14% of ^{238}Pu decays, 5% of ^{239}Pu, and 12% of ^{240}Pu.[43,44] Gamma rays following plutonium decay are rarely used for monitoring because of their much lower branching ratios; for ^{239}Pu, the most prominent gamma ray, 52 keV, is only a 0.02% branch.

^{241}Pu decays to ^{241}Am; decay of ^{241}Am has a 37% probability of producing L x-rays (13.9, 17.8, and 20.8 keV), and 36% probability of leading to a 60 keV gamma ray.[43] Depending on circumstances, this isotope may be used as an indicator for plutonium or--if the ratio of plutonium to americium is not known--it may constitute an interfering activity.

An important consideration in photon spectrometry is the geometry of the detector relative to the source. Detectors can typically average over a large area, unless collimators are employed, and interpretation of results depends on this averaging, on the isotopic composition of the contamination, and on its depth distribution in the survey area.

5.1 Photon Spectrometry with Scintillation Detectors and Gas Counters

Rapid area surveys are often performed with portable instruments based on thin large-area scintillation detectors, most often NaI(Ti) crystals. The most straightforward version of such an instrument couples such a crystal to a photomultiplier, allowing energy analysis of radiation interacting with the crystal. The archetype of this instrument, designed

for monitoring of plutonium ground contamination by observation of americium photons, is the FIDLER (Field Instrument for Detection of Low Energy Radiation)[45] which uses a 12.7 cm diameter, 0.16 cm thick crystal. Energy windows are set around 17 and 60 keV, the principal ^{241}Am photon groups. In practical applications, the sensitivity of the FIDLER is found to be a few hundred nCi/m^2 of ^{241}Am [46] although lower minimum detectable area distributions have been cited. Similar instruments have been developed based on a CaF_2(Eu) scintillator.[47,43]

More sophisticated systems have been developed to help eliminate background due to high energy photons which deposit only part of their energy in the thin scintillator. In the "phoswich" detector, illustrated in Figure 4, a thin NaI(Tl) detector is backed by a thick CsI(Tl) detector, which stops high-energy photons. CsI(Tl) has a longer decay time than NaI(Tl), so that its signal may be used to suppress the high-energy photon background in the thinner detector. This system has ordinarily been used for plutonium lung assays, in which circumstances sophisticated detectors and shielding arrangements are feasible. Under these circumstances the phoswich detector can reduce background by factors of three or more and achieve sensitivities better than the roughly 6 nCi minimum detectable ^{239}Pu achievable with an unbacked NaI(Tl) detector.[48] Recently a portable survey instrument has been developed using a phoswich type detector,[49] making a similar reduction in background as compared with the FIDLER.

Detectors relying on photon-induced ionization in a rare gas yield somewhat better resolution than thin NaI detectors. Large-area gas proportional counters have been used for plutonium lung assay, yielding minimum detectable ^{239}Pu activities of 6 nCi and resolution of 13% at 13.6 keV.[50,51] Gas proportional scintillation (as opposed to charge) counters have recently achieved considerably better resolution, about 7% in the L x-ray region,[52,53] and may be found to be useful in both laboratory and field surveys.

5.2 Photon Spectrometry with Semiconductor Detectors

Semiconductor detectors made of silicon or germanium, often lithium drifted, offer 2 to 4% energy resolution for low-energy

photons, superior to that of scintillators or gas proportional counters. On the other hand, semiconductor devices are typically very small, with active areas on the order of 10 cm^2, compared with the 100 cm^2 or more characteristic of the scintillators. Germanium and lithium detectors suffer the additional disadvantage that they must be cooled with liquid-nitrogen during use or even, in the case of Ge(Li), at all times.

The sensitivity of these detectors is typically superior to that of other systems, largely because of their better energy resolution (see Figure 5). A field comparison[46] between a FIDLER, with 130 cm^2 area, a thick Ge(Li) detector, with volume 70 cm^3 and cross-section 15 cm^2, and a 4 detector array of thin high purity (intrinsic) germanium, 0.25 cm thick with total area 33 cm^2, gave the following as minimum detectable concentrations of ^{241}Am in various conditions: 200-700 nCi/m^2 for the FIDLER, 75-200 for the Ge(Li) detector, and 40-100 for the germanium array, all with 2000 sec counting times. Planar high purity germanium arrays with total areas up to 100 cm^2 have been employed for environmental survey or sample monitoring, but it is not to be expected that these would have minimum detectable concentrations much less than the roughly 50 nCi/m^2 obtainable with a four detector array.

This limit is 5 pCi/cm^2, which - depending on the distribution in the soil - corresponds to a few pCi/cm of ^{241}Am, and somewhat more of plutonium. Consideration has been given to a more sensitive system,[54] consisting of an array of silicon detectors flanked by thick anticoincident NaI(Tl) detectors to suppress the background caused by high energy photons. Such a system would be used for assay of soil samples without performing radiochemical analysis, and could have a minimum detectable concentration of about 1 pCi/gm for plutonium alone and about 8 pCi/gm for plutonium in the presence of americium; the sensitivity to americium would be considerably better than that for Pu alone. Other investigators have employed simple Ge(Li) and Si(Li) detectors for soil assay, yielding results in agreement with radiochemical analysis when concentrations are greater than 5 pCi/gm of americium and greater than 50 pCi/gm of plutonium in the presence of americium.[55]

Plutonium lung burdens have also been measured with semiconductor detectors, including: Ge(Li),[56] Si(Li),[57] and high-purity germanium,[58] yielding sensitivity superior to scintillation detectors.

6. TECHNIQUES OTHER THAN DECAY MONITORING

The techniques discussed above are those generally used for environmental measurements of plutonium. Other techniques are available, but are not often used, either because of their difficulty or because of their lack of sensitivity. These include: neutron activation techniques, resulting either in gamma ray production or in fission; photometric techniques, including x-ray fluorescence; and electrochemical techniques, including coulometry,[59] poloragraphy,[60] potentiometric and amperometric titration.[61,62] Methods currently used are also described in ref. 39.

7. NEW TECHNIQUES FOR AIRBORNE PLUTONIUM PARTICULATES

Most air monitoring techniques rely on the fact that plutonium particles can be collected on filter materials. More subtle use can be made of this particulate character, either as an intergral part of the measurement technique or as a means to separate the plutonium from other portions of the collected sample. Substantial attention has been given to the form in which plutonium is present in fuel cycle operations.[63,64] Much of the plutonium, by mass, is present as particles with aerodynamic diameters of one to a few μm; however a substantial portion may have smaller size, particularly in chemical operations.

A technique now being studied at Battelle Pacific Northwest Laboratory[65] would use a mass spectrometer to separate plutonium compounds from other substances. The plutonium would be ionized by a filament onto which plutonium particulates impacted after passage through a thin capillary tube. The ion source thus serves as the sampling unit. (See also ref. 66.) The minimum detectable amount for such a system is much smaller than for other techniques. However, because such a source operates at high vacuum, only a small stream of air, on the order of 10 cm^3/sec, can be sampled. At the 40-hour occupational concentration limit,

1 μm (aerodynamic diameter) ^{239}Pu particles occur only about once in several liters and much more rarely if the particles are larger. Under such conditions, the unit may have to sample for long periods before encountering a plutonium particle.

The particulate character of plutonium may, however, be used as a discriminator. Impaction techniques, in which a stream of air rapidly changes direction and large particles impact onto a surface, have been used for size measurements.[63] They may also be used to separate plutonium from the very small particles which carry most of the radon-daughter background.[67] The more advanced of such systems are "virtual impaction" techniques, in which, at each separation stage, the larger particles impact into a slowly moving air stream for filter collection and counting.[68,69] One such system, designed at Argonne National Laboratory,[68] uses an alpha detector to measure the collected activities, achieving a sensitivity about an order of magnitude better than the alpha spectroscopic units described above. Semi-automatic virtual impactors have been deployed for measuring conventional pollutants.[70] It should be noted that virtual impaction concentrates the activity of interest into a small air stream, a preconcentration that would be useful for the mass spectrometric impaction technique described above, as well as for other techniques.

8. SUMMARY

Plutonium is permitted to be present in environmental media only at very low concentrations. The principal means for measuring plutonium isotopes detect alpha particles emitted during decay, X-rays immediately following decay, or X-rays and gamma rays emitted following decay of ^{241}Am, the daughter of ^{241}Pu. In most cases substantial efforts are required to overcome the background due to the uranium and thorium decay daughters, particularly--in the case of air monitoring--the radon daughters.

Alphas are typically used as the signature for the presence of plutonium in air; in monitoring alpha energies directly from an air filter, instruments of moderate sensitivity are available to detect a 40-hour occupational concentration if present for a period of hours. However, the period required can be shortened by an order of magnitude by using specially designed and rather complex systems. Much greater sensitivity

is available if the alphas are monitored after radiochemical analysis of the filter sample. The same method provides great sensitivity for measurement of plutonium in bulk samples, whether water, soil, or biological samples: subpicocurie amounts can be measured in hour-long counting periods. X-rays and gamma rays, whether from plutonium or americium, are typically used for survey efforts. Scintillation crystals, gas counters, and semiconductor detectors are available for measuring radiation energies and can effectively measure soil distributions from several hundred nCi/m^2 to about 40 nCi/m^2, with semiconductor arrays the most sensitive. This corresponds to several pCi/gm, not nearly as sensitive as radiochemical techniques which are, however, not appropriately compared with portable systems. Fixed semiconductor detector systems may achieve greater sensitivity, as low as 1 pCi/gm.

Other types of monitoring techniques are available, but not widely used. Perhaps the most significant improvement in monitoring techniques is to take explicit account of the fact that plutonium is present in air as particles: at low concentrations, air sampling techniques may be plagued by the fact that plutonium is concentrated into particles; on the other hand, impaction techniques may be used to concentrate these particles into a relatively small air stream, free of background activities. This can moderately improve air monitoring capabilities. In general, plutonium monitoring techniques are adequate for their purpose, to measure elevated levels in the environment. The primary difficulty is that substantial time is often required for carrying out measurements. However, even in this respect, improvements are being made.

This work was supported by the Division of Biological and Environmental Research of the Department of Energy.

REFERENCES

1. R. J. Budnitz, "Plutonium: A Review of Measurement Techniques for Environmental Monitoring," IEEE Trans. on Nucl. Sci. NS-21 (1), 430 (1974).

2. Survey of Instrumentation for Environmental Monitoring, Lawrence Berkeley Laboratory report LBL-1, Vol. 3: Radiation, Oct. 1973.

3. A. V. Nero, "Measurement and Instrumentation Techniques for Monitoring Plutonium and Uranium Particulates Released from Nuclear Facilities," Lawrence Berkeley Laboratory report LBL-5218, August 1976 (NTIS).

4. H. C. Hodge, J. N. Stannard, and J. B. Husch (eds.), Uranium, Plutonium, Transplutonium Elements, Springer-Verlag, New York, 1973.

5. B. J. Stover and W. S. S. Jee, Radiobiology of Plutonium, J. W. Press, Salt Lake City, 1972.

6. W. S. S. Jee, The Health Effects of Plutonium and Radium, J. W. Press, Salt Lake City, 1976.

7. International Commission on Radiological Protection, The Metabolism of Compounds of Plutonium and Other Actinides, Pergammon Press, New York, 1977.

8. "Plutonium and Other Transuranium Elements: Sources, Environmental Distribution and Biomedical Effects," U.S. AEC report WASH-1359, Dec. 1974 (NTIS).

9. T. H. Pigford and K. P. Ang, "The Plutonium Fuel Cycles," Health Physics 29, 451-468 (1975).

10. Transuranium Nuclides in the Environment, symposium (San Francisco, Nov. 17-21, 1975) International Atomic Energy Agency Proceedings Series, Veinna, 1976.

11. A. V. Nero, A Guidebook to Nuclear Reactors, University of California Press, Berkeley, forthcoming (1978).

12. "Final Generic Environmental Statement on the Use of Recycle Plutonium in Mixed Oxide Fuel in Light Water Cooled Reactors," (5 vols.) U.S. NRC report NUREG-0002, Aug., 1976 (NTIS).

13. "Final Environmental Statement, Liquid Metal Fast Breeder Reactor Program" (3 vols.), U.S. ERDA report ERDA-1535, Dec. 1975, which also includes "Proposed Final Environmental Statement" (7 vols.), U.S. AEC report, Dec. 1974 (NTIS).

14. "Proceedings of the Rocky Flats Symposium on Safety in Plutonium Handling Facilities," U.S. AEC report CONF-710401, 1971 (NTIS).

15. C. M. Lederer, J. M. Hollander, and I. Perlman, Table of Isotopes, (6th ed.), Wiley, New York, 1967.

16. For further discussion, see ref. 4 and, in addition: National Council on Radiation Protection and Measurements, "Basic Radiation Protection Criteria," NCRP report No. 39, 1971; National Academy of Sciences--National Research Council, "Health Effects of Alpha-Emitting Particles in the Respiratory Tract," U.S. EPA report EPA 520/4-76-013, Oct. 1976; International Commission on Radiological Protection; "Recommendations of the International Commission on Radiological Protection (Adopted September 17, 1965)," ICRP Publication 9, Pergammon Press, New York, 1966.

17. "Proposed Guidance on Dose Limits for Persons Exposed to Transuranium Elements in the General Environment", U. S. EPA report EPA 520/4-77-016, Sept. 1977 (NTIS).

18. J. W. Healy, "A Proposed Interim Standard for Plutonium in Soils," Los Alamos Scientific Laboratory report LA-5843-MS, Jan. 1974.

19. "Proposed Rule Making on Transuranium Waste Disposal", Federal Register, 30 FR 32921, Sept. 21, 1974.

20. M. E. Wrenn, "Environmental Levels of Plutonium and the Transplutonium Elements," in ref. 8.

21. Philip W. Krey, "Remote Plutonium Contamination and Total Inventories from Rocky Flats," Health Physics 30, 209-214 (1976).

22. L. L. Eberhardt and R. O. Gilbert, "Statistical Analysis of Soil Plutonium Studies, Nevada Test Site," Battelle Pacific Northwest Laboratory report BNWL-B-217, 1972.

23. H. L. Volchak and R. H. Knuth, "The Respirable Fraction of Plutonium at Rocky Flats," Health Physics 23, 395 (1972).

24. P. L. Phelps and L. R. Anspaugh in "The Dynamics of Plutonium in Desert Environments" (P. B. Dunaway, ed.) NVO-142, p. 221, 1974.

25. National Council on Radiation Protection and Measurements, "Natural Background Radiation in the United States," NCRP Report No. 45, November 1975.

26. B. G. Bennett, "Environmental Pathways of Transuranic Elements," in ref. 8.

27. W. A. Phillips and C. L. Lindekin, "Plutonium Alpha Air Monitor Using a Solid State Detector," Health Physics 9, 299 (1963).

28. J. F. Kordas and P. L. Phelps: "A Review of Monitoring Instruments for Transuranics in Fuel Fabrication and Reprocessing Plants," Lawrence Livermore Laboratory report UCRL-52123 (Rev. 1), March 1977; "An Online High Sensitivity Measurement System for Transuranic Aerosols," IEEE Trans. on Nucl. Sci. NS-24, 670-672 (1977).

29. E. C. Tsivoglou, H. E. Ayer, and D. A. Holaday, "Occurrence of Nonequilibrium Atmospheric Mixtures of Radon and Daughters," Nucleonics 11, 40 (1953).

30. D. E. Martz, D. F. Holleman, D. E. McCurdy, and K. J. Schiager, "Analysis of Atmospheric Concentrations of RaA, RaB and RaC by Alpha Spectroscopy," Health Physics 17, 131 (1964).

31. N. Jonassen and E. E. Hayes, "The Measurement of Low Concentrations of the Short-Lived Radon-222 Daughters in the Air by Alpha Spectroscopy," Health Physics 26, 104-110 (1974).

32. R. J. Budnitz, "Radon-222 and its Daughters--A Review of Instrumentation for Occupational and Environmental Monitoring," Health Physics 26, 145-163 (1974).

33. E. Tanaka, S. Iwadate, and H. Miwa, "A High-Sensitivity Continuous Plutonium Air Monitor," Health Physics 14, 473 (1968).

34. J. H. Spaa, "A Rapid Indicating Instrument for the Stepwise Measurement of Airdust Radioactivity," Health Physics 4, 25 (1960).

35. M. O. Rankin, "The Use of Coincidence Counting Techniques for Analyzing Low Level Plutonium Contamination on Filters," Nucl. Inst. and Methods 24, 221 (1963).

36. E. D. Gupton, "Alpha Air Monitor for Plutonium-239," Oak Ridge National Laboratory report ORNL-TM-2011, 1967.

37. J. W. Healy, "Problems of Plutonium Measurement in the Environment," IEEE Trans. Nucl. Sci. NS-19 (1), 219 (1972).

38. International Atomic Energy Agency, Rapid Methods for Measuring Radioactivity in the Environment, IAEA, Vienna (1971), comment by C. W. Sill, p. 199.

39. J. H. Harley (ed.), HASL Procedures Manual, USAEC Health and Safety Laboratory Report HASL-300, 1972, updated to 1977.

40. "Nevada Applied Ecology Group Procedures Handbook for Environmental Transurannics," U. S. ERDA Nevada Operations Office report NVO-166 (2 volumes), October 1976.

41. N. A. Talvitie, "Radiochemical Determination of Plutonium in Environmental and Biological Samples by Ion Exchange," Anal. Chem. 43, 1827 (1971).

42. C. W. Sill, K. W. Puphal, and F. D. Hindman, "Simultaneous Determination of Alpha-Emitting Nuclides of Radium through Californium in Soil," Anal. Chem. 46, 1725 (1974).

43. H. L. Beck, J. E. McLaughlin and K. M. Miller, "In-Situ Determinations of Environmental Plutonium and its Related Nuclides," IEEE Trans. on Nucl. Sci. NS-23, 676-682 (1976).

44. M. G. Strauss, I. S. Sherman, E. J. Swanson and R. H. Pehl, "Considerations in Measuring Trace Radionuclides in Soil Samples by L X-Ray Detection," IEEE Trans. on Nucl. Sci. NS-25, to be published in 1978.

45. J. F. Tinney, J. J. Koch, and C. T. Schmidt, "Plutonium Survey with an X-Ray Sensitive Detector," Lawrence Livermore Laboratory report UCRL-71362, 1969.

46. J. A. Kirby, L. R. Anspaugh, P. L. Phelps, G. A. Armantrout and D. Sawyer, "A Detector for In-Situ Spectrometric Analysis of ^{241}Am and Pu in Soil," IEEE Trans. on Nucl. Sci. NS-23, 683-689 (1976).

47. B. S. Burton and M. N. Anastasi, "Development and Construction of an Instrument to Detect Fissionable Materials," Air Force Weapons Laboratory report AFWL-TR-71-145, 1972.

48. T. Ishihara, T. Iinuma, E. Tanaka, and S. Yashiro, "Plutonium Lung Monitor Using a Thin NaI(Tl) Crystal of Large Area," Health Physics 17, 669 (1969).

49. M. A. Wolf, "Portable Survey Instrument for Low Levels of Transuranic Contaminants," Los Alamos Scientific Laboratory report LA-6860-MS, July 1977.

50. T. Tomitani and E. Tanaka, "Large Area Proportional Counters for Assessment of Plutonium Lung Burden," Health Physics 18, 195-206 (1970).

51. S. S. Yaniv, A. Brodsky and D. Sashin, "Analysis of Pu-Am Spectra in a Xe Proportional Counter: Effect of Characteristic X-Rays," Health Physics 23, 705-714 (1972).

52. A. J. P. L. Policarpo, M. A. F. Alves, M. C. M. Dos Santos and M. J. T. Carvalho, "Improved Resolution for Low Energies with Gas Proportional Scintillation Counters," Nucl. Inst. and Meth. 102, 337-348 (1972).

53. H. E. Palmer, "Recent Advances in Gas Scintillation Proportional Counters," IEEE Trans. on Nucl. Sci. NS-22, 100-103 (1975).

54. M. G. Strauss, I. S. Sherman, E. J. Swanson and R. H. Pehl, "Considerations in Measuring Trace Radionuclides in Soil Samples by L X-Ray Detection," IEEE Trans. on Nucl. Sci. NS-25, to be published.

55. F. P. Brauer, J. M. Kelley, R. W. Goles, and J. E. Fager, "Measurement of Environmental ^{241}Am and the Pu/^{241}Am Ratio by Photon Spectrometry," IEEE Trans. on Nucl. Sci. NS-24, 591-595 (1977).

56. W. H. Tyree and R. W. Bistline, "Plutonium-239 Detection in Vivo with a Germanium-Lithium Array," Rocky Flats report RFP-1488, 1970.

57. R. W. Bistline and W. H. Tyree, "A Comparison of Low-Energy Photon Detection for Plutonium and Americium Wound Counting," Rocky Flats Report RFP-1068, 1967.

58. W. H. Tyree, R. B. Falk, C. B. Wood, and R. W. Liskey, "A High-Purity Germanium Detection System for in vivo Measurement of Americium and Plutonium," Rocky Flats report RFP-2525, 1976.

59. J. R. Stokely and W. D. Shults, "Controlled-Potential Coulometric Determination of Plutonium in the Presence of Iron," Anal. Chem. 43, 603 (1971).

60. K. Koyama, "Square Wave Polagraphy of Plutonium," Anal. Chem. 32, 523 (1960).

61. C. E. Pietri and A. W. Wenzel, "Improved Methods for the Constant-Current Potentiometric Determination of Plutonium," Talanta 14, 215 (1967).

62. C. E. Herrick, C. E. Pietri, A. W. Wenzel and M. W. Lerner, "Improved Amperometric Procedure for Determining Plutonium," Anal. Chem. 44, 377 (1972).

63. J. C. Elder, M. Gonzales, and H. J. Ettinger, "Plutonium Aerosol Size Characteristics," Health Physics 27, 45 (1974).

64. W. B. Seefeldt, W. J. Mechan, and M. J. Steindler, "Characterization of Particulate Plutonium Released in Fuel Cycle Operations," Argonne National Laboratory report ANL-75-78, May 1976.

65. Battelle Pacific Northwest Laboratory report BNWL-2000, Part IV, Feb. 1976; J. Stoffels, private communication; cf. also ref. 28.

66. W. E. Davis, "Continuous Mass Spectrometric Analysis of Particulates and Other Impurities in Air and Water Using Surface Ionization," General Electric Corporation, 1975.

67. G. K. Rusch, W. P. McDowell, and W. G. Knapp, "The ZPR-9 Airborne Plutonium Monitoring System," IEEE Trans. on Nucl. Sci. NS-23, 690 (1976).

68. T. J. Yule, "An On-Line Monitor for Alpha-Emitting Aerosols", IEEE Trans. on Nucl. Sci. NS-25, to be published, 1978.

69. D. Talley, G. J. Newton, H. G. Yeh, and S. J. Waligora, "Development of a Five-Stage, Round Jet, Virtual Cascade Impactor," in "Inhalation Toxicology Research Institute Annual Report, 1975-1976," Lovelace Biomedical and Environmental Research Institute report LF-56, Dec. 1976.

70. B. W. Loo, J. M. Jaklevic, and F. S. Goulding, "Dichotomous Virtual Impactors for Large Scale Monitoring of Airborne Particulate Matter," Lawrence Berkeley Laboratory report LBL-3854, 1975.

Figure Captions

Figure 1. SCHEMATIC ARRANGEMENT FOR ALPHA DETECTION FROM AIR FILTERS

A semiconductor detector is ordinarily used to count, and perhaps measure the energy of, alpha particles emitted from the filter medium. The variation in path length indicated may notably affect the amount of energy deposited in the detector.

Figure 2. RELATIVE STRENGTH OF ALPHA GROUPS FROM TYPICAL PLUTONIUM MIXTURES

Schematic spectra for the mixtures of Table 3 are given. Within each mixture, the height of each line is proportional to the strength of that group. (The mass numbers indicate which isotope accounts for each group.) Also shown are schematic spectra for the radon and thoron decay chains.

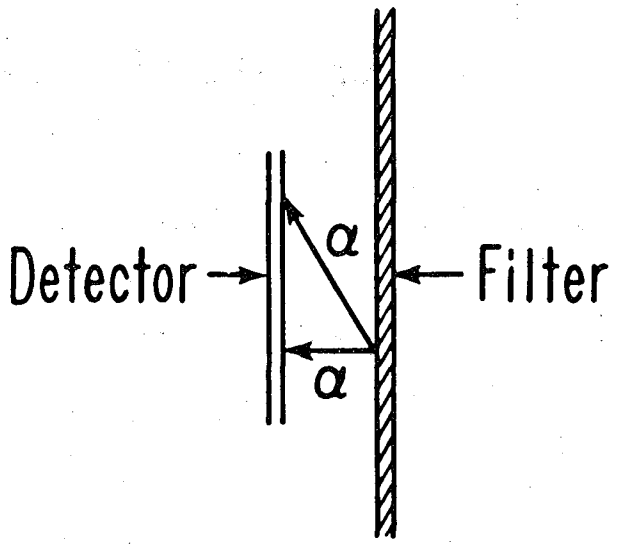
Figure 3. RADON DAUGHTER INTERFERENCE IN PLUTONIUM ALPHA DETECTION

The illustration indicates how, in an ordinary alpha detection system, radon daughter activity can interfere with monitoring of plutonium. In this case, because of relatively poor resolution, ^{218}Po (Radium A) contributes counts in the region of the ^{239}Pu groups.

Figure 4. PHOSWICH DETECTOR

For low energy electromagnetic radiation, whether X-rays or low energy gamma rays, background may be reduced by using a thin CsI(Tl) scintillator as the primary detector and a thick NaI(Tl) scintillator in anticoincidence (to minimize the effect of high energy radiation leaving small amounts of energy in the thinner detector).

Figure 5. ^{239}Pu and ^{241}Am SPECTRA TAKEN WITH A Ge(Li) DETECTOR
Semiconductor detectors are increasingly being used for field measurement of plutonium/americium electromagnetic emissions. The illustration shows spectra accumulated with a 5 cm^2 Ge(Li) detector (cf. ref. 55). (Figure courtesy of Battelle Pacific Northwest Laboratory.)



XBL 768-3257

Figure 1

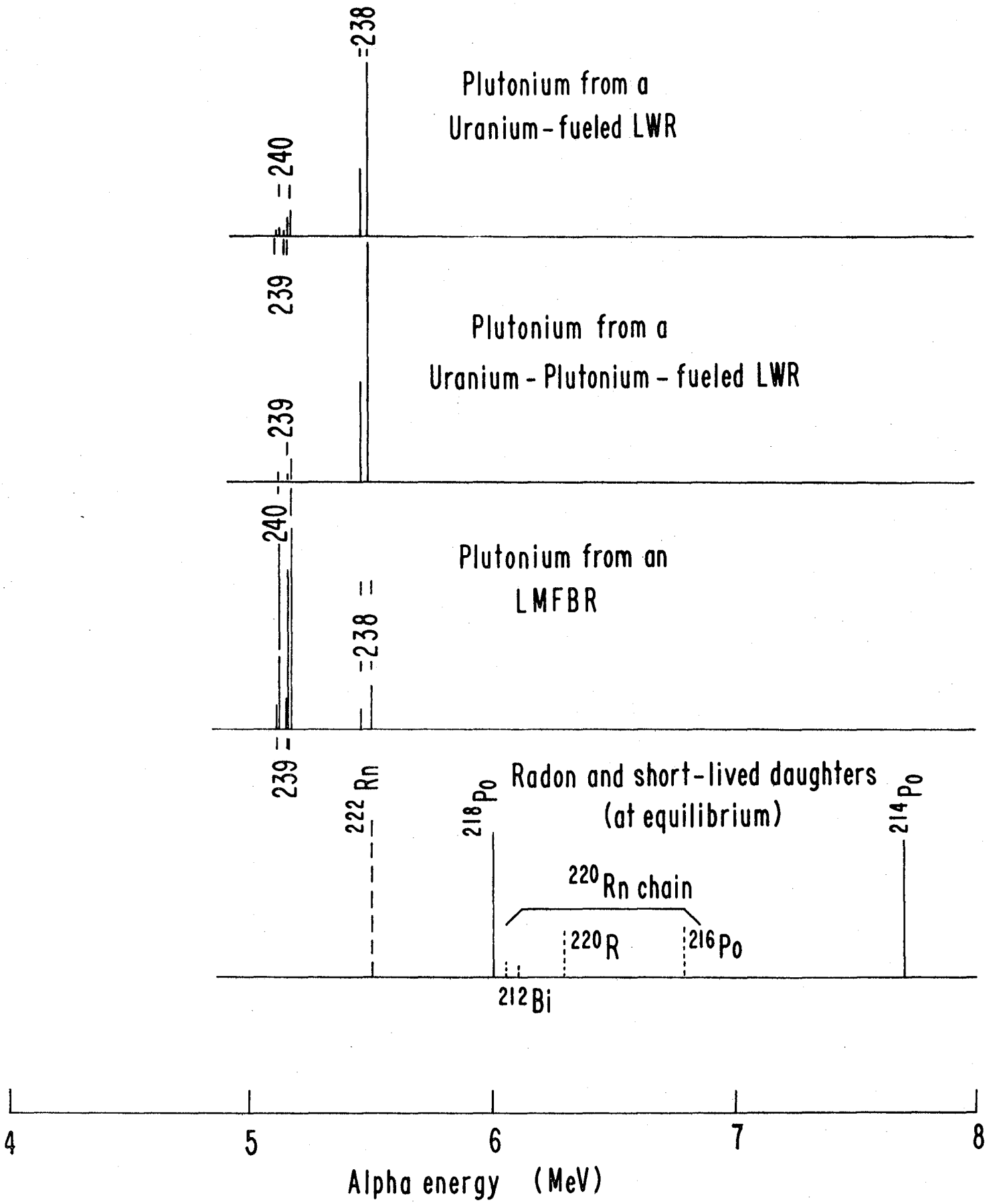


Figure 2

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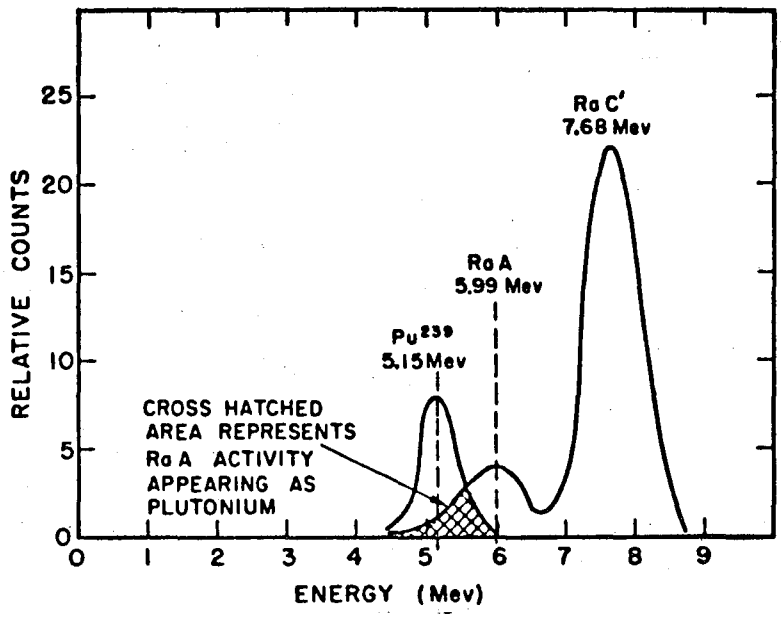
RELATIVE COUNTS
25
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ENERGY (Mev)
Ra C'
7.68 Mev
Ra A
5.99 Mev
Pu239
5.15 Mev
CROSS HATCHED
AREA REPRESENTS
Ra A ACTIVITY
APPEARING AS
PLUTONIUM

Figure 3

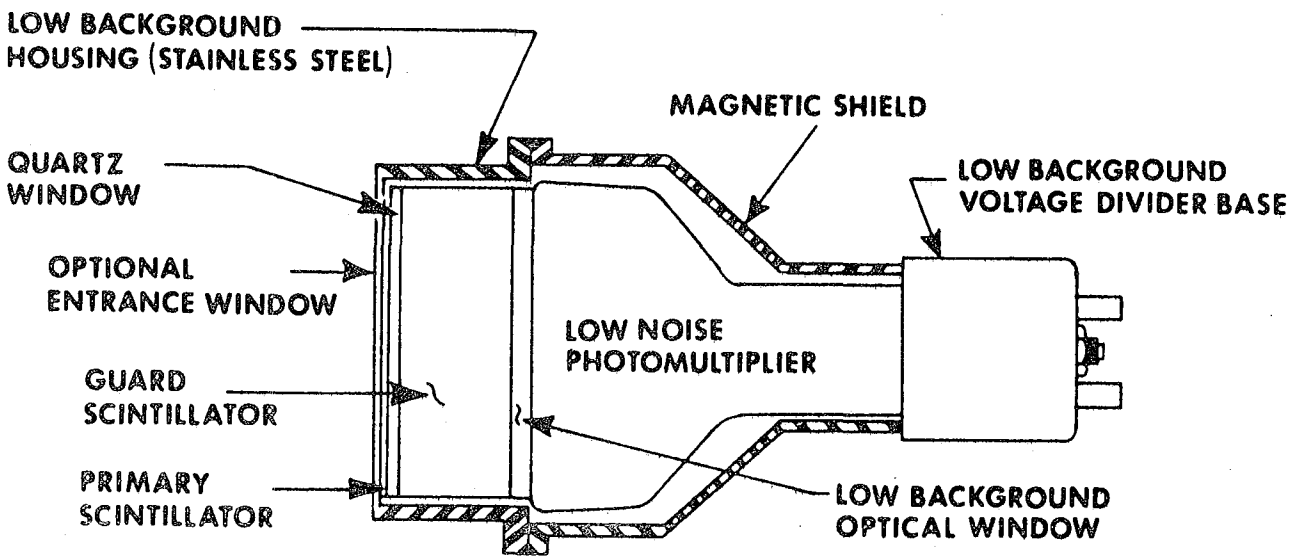
LOW BACKGROUND
HOUSING (STAINLESS STEEL)
MAGNETIC SHIELD
QUARTZ
WINDOW
LOW BACKGROUND
VOLTAGE DIVIDER BASE
OPTIONAL
ENTRANCE WINDOW
LOW NOISE
PHOTOMULTIPLIER
GUARD
SCINTILLATOR
PRIMARY
SCINTILLATOR
LOW BACKGROUND
OPTICAL WINDOW

Figure 4

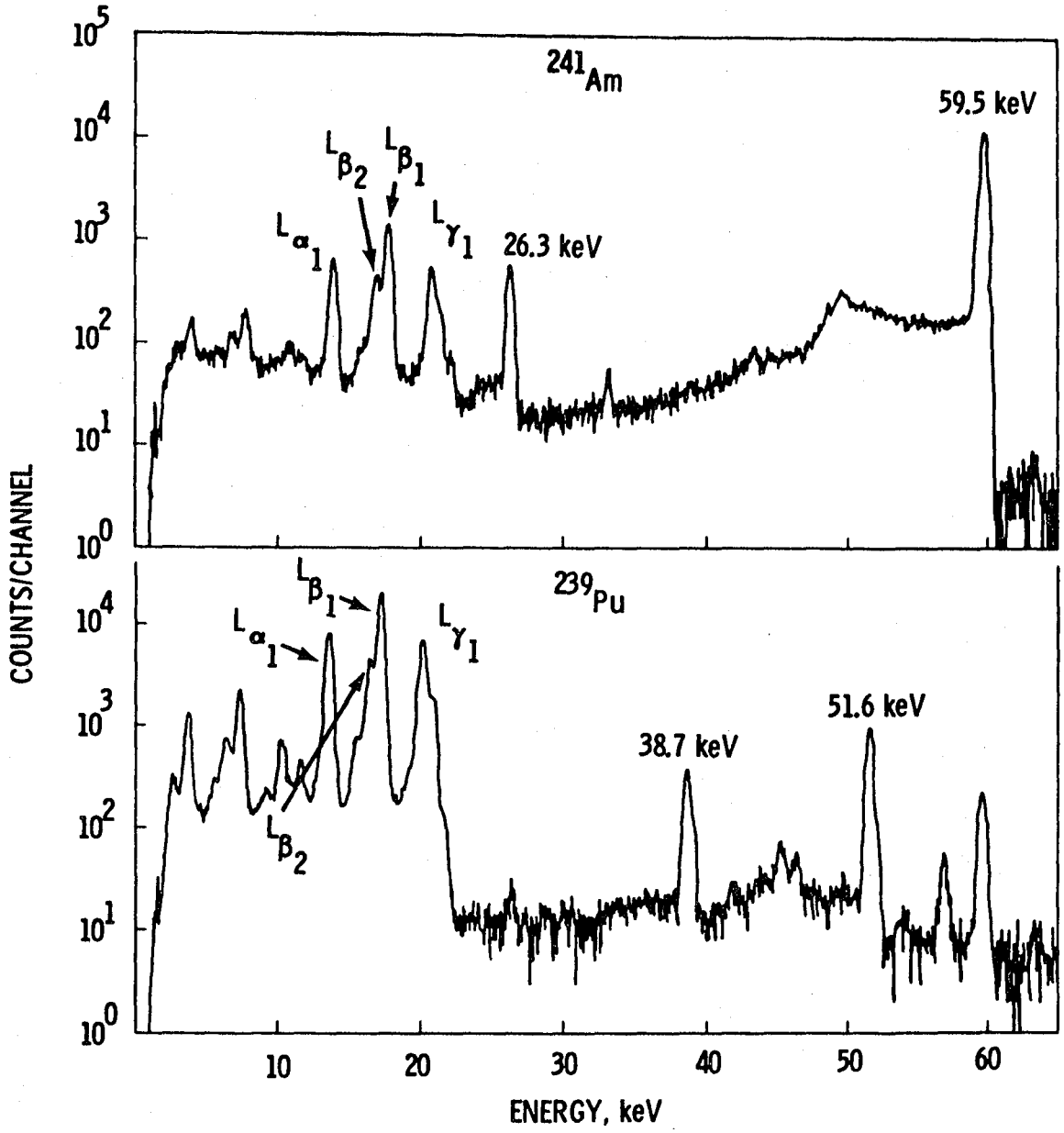
241Am
59.5 keV
Lβ2
Lβ1
Lα1
Lγ1
26.3 keV
239Pu
Lβ1
Lα1
Lγ1
Lβ2
38.7 keV
51.6 keV
COUNTS/CHANNEL
ENERGY, keV
10
20
30
40
50
60

Figure 5

This report was done with support from the United States Energy Research and Development Administration. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the United States Energy Research and Development Administration.

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