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Departmental Report
A Survey of an Air Monitoring Program

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MASTER

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ABSTRACT

An air monitoring program administered at Argonne National Laboratory was assessed by comparing personal air sampler (PAS) data, stationary air sampler (SAS) data, and bioassay data. The study revealed that the PAS and SAS techniques were equivalent when averaged over all employees and all workdays, but the standard deviation was large. Also, large deviations were observed in individual samples. The correlation between individual PAS results and bioassay results was low. Personal air samplers and bioassay monitoring played complementary roles in assessing the workplace and estimating intakes. The PAS technique is adequate for detection and evaluation of contaminated atmospheres, whereas bioassay monitoring is better for determining individual intakes.

Introduction

The objective of this report is to compare personal air sampling data to stationary air sampling data and to bioassay data that was taken during the decontamination and decommissioning of sixty-one plutonium glove boxes at Argonne National Laboratory (ANL) in 1995. The glove boxes were decommissioned to eliminate cost of routine surveillance and maintenance and to make nine laboratories available for other purposes. The air monitoring program during the operation consisted of workers wearing personal air samplers (PAS) and stationary air samplers (SAS) positioned throughout the project workplace. Bioassay sampling was performed to monitor for actual intake of airborne radioactive material by the workers during the course of work activities. The radioactive contaminants present inside the glove boxes were predominately $^{238,239,240}\text{Pu}$, with lower quantities of ^{241}Am and trace amounts of ^{235}U , ^{233}Pa , ^{243}Cm , ^{237}Np and ^{137}Cs .

The three basic pathways of intake for plutonium are inhalation, ingestion, and contaminated wounds. In the workplace, the most frequent route for plutonium intake is inhalation. The aerodynamic properties of the contaminated aerosol inhaled determine the distribution of deposition in the respiratory track. Once the particles have been deposited within the respiratory track, the transportability of the particle is also governed by its chemical form. Plutonium particles that are deposited in the nasopharyngeal and trachea-bronchial regions are rapidly cleared by ciliary mucous transport, swallowed into the gastrointestinal tract, and excreted in the feces. Smaller particles that deposit in the pulmonary region of the lung may be cleared by two routes. Particles may be rapidly phagocytized by alveolar macrophages (large cells that act as scavengers

that engulf debris, dead cells, and particles) and cleared up the bronchi and trachea by ciliary action, or they may enter into the blood by solubilization, cellular transport, or direct transfer.

Two inhalation classes have been established to describe the clearance of plutonium. Class W material is cleared in a period of weeks and Class Y material is cleared in years. Plutonium in the oxide form is assigned to inhalation class Y and all other compounds of plutonium to inhalation class W. For health and safety purposes, the primary radionuclides of concern were the isotopes of plutonium. To provide an appropriate safety envelope, the assumption was made that plutonium was in the most insoluble chemical form, plutonium oxide (PuO_2). This chemical form is assigned to the lowest annual limit intake of plutonium molecules (ICRP 30, part 4).

Plutonium particles that are ingested are expected to be excreted in the feces. The absorption of insoluble plutonium compounds in the gastrointestinal tract (GI) is extremely low, typically on the order of 10^{-5} (ICRP 30, 1979). Of the plutonium reaching the blood stream, it is expected to be deposited in the liver and skeleton with an assimilation of 45% of the body burden in each organ. The remaining 10% is assumed to be deposited into other tissues and early excreta in the urine (ICRP 30, 1979). The biological half-lives for plutonium in the liver and skeleton are 40 years and 100 years respectively (Raabe, 1994).

Plutonium presents a potential inhalation hazard if it becomes airborne. Therefore, it is important to design an effective air surveillance program to identify the loss of radiological controls and monitor for the potential intake of plutonium by workers. The Department of Energy Code of Federal Regulations 10 part 835 (10 CFR Part 835) lists work practices to minimize the intake of radioactive material and to comply with recommended internal radiation

quantities established by the International Commission on Radiological Protection (ICRP). These standards were developed to limit occupational exposure and, ultimately, the risks of radiation induced cancers and mutations (Raabe, 1994). Intake and exposure limits are represented by the annual limit intake (ALI) and the derived air concentration (DAC). The following definitions are from 10 CFR 835.

“ALI is the quantity of a single radionuclide which, if inhaled or ingested in 1 year, would irradiate a person, represented by *Reference Man (ICRP Publication 23) to the limiting value for control of occupation exposure.”

“DAC is the concentration of a radionuclide in air that, if breathed over the period of a work year, would result in the ALI for that radionuclide being reached. The DAC is obtained by dividing the ALI by the volume of air breathed by an average worker during a working year ($2.4 \times 10^3 \text{ m}^3$).”

Table 1 lists the ALI and DAC for the three isotopes of plutonium that were taken from ICRP 30 part 4. These limits should not be exceeded during any working year in order to not exceed current dose limits. The concept of maintaining doses to levels as low as reasonable achievable (ALARA) was developed to help design, assess and control working environments. Furthermore, ALARA helps maintain exposures below the recommended limits. The 10 CFR part 835 defines ALARA as:

“An approach to radiological control to manage and control exposures (individual and collective) to the work force and to the general public at levels as low as is reasonable, taking into account social, technical, economic, practical and public policy considerations.”

*Reference Man is a person with average anatomical and physiological characteristics and is the basis of limits that are defined in the report of the ICRP Task Group on Reference Man (ICRP Publication 23).

Table 1. The ALI (Bq) and DAC (Bq/m³) based on a 40 h/wk for isotopes of plutonium.

	Ingestion	Ingestion	Inhalation Class W	Inhalation Class Y
GI Assimilation Fraction	$f_1 = 1 \times 10^{-4}$	$f_1 = 1 \times 10^{-5}$	$f_1 = 1 \times 10^{-4}$	$f_1 = 1 \times 10^{-5}$
Radionuclide				
²³⁸Pu ALI T_{1/2}=86.4 yrs	3×10^5	3×10^6	2×10^2	6×10^2
DAC	-----	-----	9×10^{-2}	3×10^{-1}
²³⁹Pu ALI T_{1/2}=24,400yrs	2×10^5	2×10^6	2×10^2	5×10^2
DAC	-----	-----	8×10^{-2}	2×10^{-1}
²⁴⁰Pu ALI T_{1/2}=6580 yrs	2×10^5	2×10^6	2×10^2	5×10^2
DAC	-----	-----	8×10^{-2}	2×10^{-1}

MATERIALS AND METHOD

The 10 CFR part 835 lists requirements for air sampling when an individual is expected to receive an annual intake greater than or equal to 2 percent of ALI or an exposure to 40 DAC-hours per year. Under these conditions, the following equipment and practices were used to meet the requirements set forth in 10 CFR part 835. A containment structure was built to preclude spread of contamination. A workplace surveillance program using air sampling equipment (PAS and SAS), continuous area monitors, bioassay monitoring and personal protective equipment were employed to monitor exposures and keep intakes below the recommended limits.

Containment Structure

A containment structure (figure 1) was assembled to prevent the spread of contamination into the laboratory during the glove box size-reduction operations. The structure was composed of two compartments, a disassembly area and an airlock area. The airlock served as a buffer area between the clean laboratory environment and the highly contaminated disassembly area. The overall dimensions of the containment structure were 28 ft long, 12 ft wide, and 7 ft tall. The structure consisted of a series of metal panels with viewing windows along the wall. Skylights were used to provide adequate lighting. The internal walls of the containment structure were coated with strippable epoxy paint. The floors were layered with plastic fabric (puncture and tear resistant) to provide a removable floor surface for regular change during the project to minimize resuspension of loose contamination. The ventilation rate within the structure was designed to be at least 60 air changes per hour. The local exhaust filter systems, positioned inside the containment structure above the workstation, served as the primary barrier to remove the aerosols generated in the workplace.

Description of Air Sampling/ Air Monitoring Equipment

A personnel air sampler samples air near the breathing zone of a worker. The PAS used in this operation was a lapel type sampler composed of a light-weight compact pump worn on a worker with the filter head clipped to the lapel. MSA Flow-lite sample pumps with cellulose filters had an average air flow rate of 4 liters per minute (lpm) (Munyon, 1995). The PAS was used to estimate the radioactive particulate concentrations in the vicinity of the worker.

Figure 1. Containment structure for the size reduction of the glove boxes.

A stationary air sampler (SAS) is a monitor at a fixed location. The SAS used in this operation was developed at ANL. The sampling head consisted of a glass fiber filter and a Lucite block with a built-in rotameter. The SAS operated at a flow rate of 40 lpm. The SAS was used to measure radioactive aerosols in the ambient air.

Continuous air monitors (CAMs) are stationary, air monitoring units that measure radioactive material on a "real time" basis by continuously pulling ambient air through collection tubes or filters. They continuously measure the count rate of the radiation of interest (alpha, beta

or gamma). In this operation, Eberline Alpha-6 CAMs were positioned in the disassembly area, airlock and laboratory. The CAMs were used to warn workers, by sounding an alarm, in cases of excessive airborne contamination in the containment structure and surrounding areas. The alarm set point was based on the escape of contaminants in the disassembly area to ensure that workers were not exposed to concentrations in excess of the administrative limit of 0.1 DAC (10 CFR part 835).

Bioassay Analysis and Scheduling

Bioassay monitoring is the analysis of radioactive materials in the human body by in vivo measurements or by in vitro examination of material excreted or removed from the body (Raabe, 1994). Urine analysis and fecal analysis, and lung counting were employed for workers in this operation at ANL. A baseline bioassay was performed to determine the radionuclide content present in each individual prior to the project. Throughout the duration of this project, periodic urine and fecal samples were collected and analyzed for any increase of radioactive material. In the event of a potential exposure, special bioassay samples were collected to determine if there was a true intake. If the results were above the decision level (a final measurement of a radionuclide at or above which a decision is made that the radionuclide is present) more samples were collected to confirm the result and to determine the magnitude of the suspected intake. If applicable, a committed effective dose equivalent and specific committed doses were calculated and assigned to the appropriate worker. At the end of a project or upon exclusion of a worker, a termination bioassay was conducted to assess the final internal doses.

Personal Protective Equipment

Protective equipment was used to reduce exposure to airborne radioactive particulates. Protective clothing was worn by the workers to decrease the potential of radioactive contamination of the skin and personal clothing.

Respiratory protection was used to reduce concentrations of inhaled airborne radioactive material. Full-face, positive-pressure, air-purifying respirators with high efficiency particulate (HEPA) filters were used. The supplied air respirator included a face mask that covered the entire face up to the hairline. The supplied air came from ambient air filtered through the HEPA filter. These respirators were reported to have a protection factor of 1000 (ANL Report, 1995).

Operations

Each workday consisted of two shifts of three contract workers wearing respirators and PASSs. Three SASs were positioned inside the disassembly area of the containment structure. The SASs were identified by their position (figure 1). The north SAS was positioned on a microphone stand approximately one meter from the workers. The south SAS was stationed underneath a piece of ductwork in proximity to the wall. The west SAS was located downstream of the workers and between two of the room exhaust ports.

The filters on both the PAS and SAS were counted for alpha activity, using a Tennelec gas flow proportional counter, within an hour following the end of each shift. A second count was performed seven days later. The second count was conducted to allow for the decay of radon (^{222}Rn) and thoron (^{220}Rn) decay products that plated-out on the filter or attached to dust particles that were collected by the filter. The location, time and duration of sampling, flow rate and count rate were recorded on a data sheet. The activity (Bq) was calculated by dividing the count

rate by an average measured detector efficiency of 27%. The activity concentration was calculated by dividing the activity by the run time (total time of each shift) and the flow rate of the monitor. A gas-flow proportional counter was used because it was accessible and had an automatic sample feeder. The detector was calibrated using an ^{241}Am source traceable to the National Institute of Standards and Technology (NIST). Americium-241 was used because it has alpha energies similar to those emitted by the decay of plutonium isotopes, which are shown in table 2.

Table 2. Alpha energies (MeV) and corresponding intensities for ^{241}Am and $^{238,239,240}\text{Pu}$.

Radionuclide	Energy (MeV) [Intensity]	
^{241}Am	5.49 [85%]	5.44 [13%]
^{238}Pu	5.50 [72%]	5.46 [28%]
^{239}Pu	5.16 [88%]	5.11 [11%]
^{240}Pu	5.17 [76%]	5.12 [24%]

Routine bioassay samples were collected on alternating weeks from each worker from different shifts to determine if there were any measurable intakes.

RESULTS AND DISCUSSION

Personal Air Samplers versus Stationary Air Samplers

The work day, starting time and shift duration were the same for the PASs and SASs. The average of the concentrations of the six PASs for the combined morning and afternoon shifts were taken to obtain a representative concentration to describe the aerosol activity generated for each workday. Similarly, the average of the concentrations of the six SASs for the morning and

afternoon shifts were also calculated. A concentration frequency distribution for the PAS and SAS shows that the majority of the concentrations fall below 200 mBq/m³ (figure 2). On the 34th workday, activity concentrations higher than normal were measured by the PASs and SASs. The average concentrations, which are illustrated in figure 3, were 64,446.94 mBq/m³ (PAS) and 67,192.50 mBq/m³ (SAS). These high activity concentrations obscure trends that can be observed for other workdays for each monitor type. Table 3 shows the difference between the range and mean of the concentrations for the PAS and SAS including and excluding the 34th workday.

Figure 3 shows the average concentrations of PAS versus the corresponding average concentrations of SAS for each workday. The data are scattered and show a positive correlation, but no direct relationship. Concentrations measured by the PAS and SAS on the 34th workday might suggest that the PAS and SAS may be equivalent when measuring extremely high concentrations.

Table 3. The range and mean of the average concentrations for the duration of the project for the PAS and SAS including and excluding the 34th workday.

	Including 34th Workday		Excluding 34th Workday	
	Range (mBq/m ³)	Mean (mBq/m ³)	Range (mBq/m ³)	Mean (mBq/m ³)
PAS	1.86-64,446.94	1687.96 +/- 1427.54	1.86-1834.47	261.62 +/- 59.97
SAS	5.55-67,192.50	1735.68 +/- 1488.56	5.55-1116.11	248.03 +/- 43.24

Figure 2. Frequency distribution for the daily average concentrations of PAS and SAS for the duration of the project. The first bin (0) represents any value less than or equal to (0). The remaining bins represent all values greater than the previous entry up to and including that number.

Figure 3. Average concentrations of PAS versus average concentrations of SAS.

The ratio of the average concentration of the PASs to the average concentration of the SASs for each workday is shown in figure 4. The absence of data indicates that there was inadequate data collected for that workday. The frequency distribution of the ratios of the PAS to SAS (figure 5) shows that over 80% of the concentrations measured with the PASs are within a factor of 3 of the concentrations measured with the SASs. On the 39th workday, the PAS overcounted the SAS by a factor of 46 (figure 5). Table 4 displays the concentrations of each PAS and SAS for that day. The activity concentrations for the SASs in the morning and afternoon shift were low and indicate that there was little generation of radioactive aerosol. Higher activity measured on one PAS compared to the other two PASs in both the morning and afternoon shift is evidence that the radioactive aerosol was localized near the source. It was assumed that the PAS with the higher activity was worn by the worker cutting the gloveboxes.

One SAS was positioned near the exhaust ducts (figure 1) inside the containment structure. Studies have shown that the best location for consistent detection is near room exhaust ducts (Alvarez, 1994). Instead of choosing one SAS to represent the aerosol concentration in the room, it was decided that the average of the three SASs would yield the best estimate of activity aerosol produced in the room. To determine if the activity concentration for each PAS was closely related to the estimated activity concentration in the room, the ratio of each individual PAS to the average SASs for each shift were taken. Figure 6 shows the frequency distribution of

Figure 4. Ratio of the average concentration of PAS to average concentration of SAS for each workday.

Table 4. Activity concentrations (mBq/m³) for each PAS and SAS for the morning and afternoon shift for the 39th workday.

	PAS 1	PAS 2	PAS 3	South SAS	North SAS	West SAS
Morning	360.00	0	65.00	2.17	1.50	12.50
Afternoon	6.67	73.33	1520.00	3.00	2.17	23.33

Figure 5. Frequency distribution of the logarithm of ratio of the average concentration of PAS to average concentration of SAS. The first bin (-2) represents any value less than or equal to (-2). The remaining bins represent all values greater than the previous entry up to and including that number.

Figure 6. Frequency distribution of the ratio of the SAS average concentration to each PAS concentration of each shift. The first bin (-2) represents any value less than or equal to -2. The remaining bins represent all values greater than the previous entry, up to and including that number.

these ratios. Fifty percent of the PASs were within a factor of 2 of the average SASs while 32% were greater than a factor of 3.

The cumulative average concentrations of the PASs and SASs for each workday (figure 7a and 7b) show the differences between the two samplers and daily variation in concentrations. High activity measured on the 34th workday (figure 7a) shows a large jump between the concentrations, leading to the conclusion that the working conditions or method of work deviated from general work operations. Following the 34th workday, a shift occurs in the data that is more apparent in figure 7b. A linear regression performed on the data in figure 6b (before and after the 34th workday) showed a decrease in slope after the 34th workday for the PAS and SAS by a factor of 2 and 4 respectively. Prior to day 34, an ALARA goal was achieved, but, following the high activity incident, further attempts to reduce exposure were made. A structure was built and positioned around the saw to minimize the dispersement of radioactive particles. The reduction of the circulation of radioactive aerosol is demonstrated by the decrease in the slopes of the concentrations in figure 7b. This indication showed that a lower ALARA goal could be achieved.

Alpha (α) particles are monoenergetic and have a range of approximately 5 cm in air. Not all α -emissions emitted from radioactive decay on a filter will reach the detector to be counted because of detector geometry and attenuation while traversing through media such as air, filters, and detector windows. Sometimes radioactive particles become embedded in the filter medium and the filter attenuates the emitted α -particles (self-absorption). Correction was made for filter self-absorption by multiplying the count rate by 1.3 (historical value for ANL)

Figure 7a. Cumulative average concentrations of PAS and SAS as a function of workday.

Figure 7b. Cumulative average concentrations of PAS and SAS as a function of workday, excluding the 34th workday.

or a 30% increase. Self-absorption was examined to determine if 1.3 was a reasonable factor to apply. In addition, the self-absorption of the glass-fiber and cellulose filters was compared.

Radiochemical analyses of eight arbitrarily chosen filters were performed. Four filters were chosen from both types of samplers: glass-fiber filters #1 through #4 and cellulose filters #5 through #8. Each filter was counted in a low-background alpha/beta counter and then cut into quarters. Each quarter was counted and dissolved and the actinide elements purified. The samples were then counted using α -spectrometry.

Self absorption of α - particles in the filters was determined by comparing the radiochemical analysis and the activity measured by the gas-flow proportional counter (table 5) . The ratios of the detector measurements to the radiochemical measurements ranged from 0.89 to 1.51 with a mean ratio of 1.22. The average ratio for the glass-fiber filter and cellulose filter were 1.40 and 1.05 respectively. The self-absorption factor appeared higher in the glass-fiber filters than for the cellulose filters. The difference in self-absorption between the glass-fiber filter and cellulose filter might result from the particle collection properties. Cellulose filters have interconnecting pores of uniform size and collect most particles at or near the surface of the filter, whereas the glass-fiber filters have oriented-fiber glass mats in which an appreciable fraction of particles can become trapped. Also, the glass-fiber and cellulose filters have different particle collection efficiencies which range from 99.6% to >99.99% and 43% to 95.5% respectively. Surface-particle collection is advantageous for α sampling because it minimizes self absorption

Table 5. Results of radiochemical (RC) measurements of eight air filters @ 1 standard deviation. Filters #1 through #4 are glass-fiber and filters #5 through #8 are cellulose.

Filter	#1	#2	#3	#4	#5	#6	#7	#8
Total (α cpm)	49.1+/- 0.7	20.4+/- 0.5	19.0+/- .05	27.3+/- 0.6	6.7+/- 0.3	26.3+/- 0.5	56.2+/- 0.8	45.8+/- 0.7
1st Qtr (α cpm)	10.4+/- 0.6	3.4+/- 0.3	5.6+/- 0.4	4.7+/- 0.4	4.2+/- 0.4	4.9+/- 0.4	7.2+/- 0.5	4.2+/- 0.3
2nd Qtr (α cpm)	8.2+/- 0.6	7.7+/- 0.4	3.3+/- 0.4	4.2+/- 0.7	0.5+/- 0.1	12.9+/- 0.5	19.5+/- 0.7	24.0+/- 0.3
3rd Qtr (α cpm)	12.2+/- 0.6	5.1+/- 0.4	4.9+/- .04	13.4+/- 0.7	0.2+/- 0.1	7.1+/- 0.5	14.8+/- .07	2.5+/- .03
4th Qtr (α cpm)	14.8+/- 0.7	3.5+/- 0.3	4.0+/- 0.4	6.8+/- 0.5	1.3+/- 0.2	1.6+/- 0.2	13.0+/- 0.7	17.3+/- 0.8
RC measurement of total α -dpm	157.4	69.1	69.0	98.7	28.6	96.1	240.1	153.5
Detector estimate of total α -dpm	239	104	67	160	35	102	213	155
Detector estimate RC measurement	1.51	1.51	0.97	1.62	1.24	1.06	0.89	1.01

* alpha counts per minute (α -cpm)

* alpha disintegrations per minute (α -dpm)

within the filter. Also, a high collection efficiency is desired to obtain adequate samples. The self-absorption factor of 1.3 used by ANL is conservative because the activity measured by the detector overestimates the true activity determined by the radiochemical analysis. The same type of filters should have been used for both the PAS and SAS. Two different types of filters were used because two different monitoring groups at ANL were involved in this project.

For the assessment of potential inhalation and exposure, it is important to understand the size distribution of particles in an aerosol. The probability that a particle will be inhaled and deposited in the respiratory track (respirable particles) can be determined by the aerodynamic

diameter (AD). The AD is a "measure of the inertial behavior of a particle moving in air and is defined as the diameter of a unit density (1 g/cm^3) sphere that settles with the same velocity as the particle" (Lodge and Chan, 1986). A cascade impactor separates particles by AD, typically ranging from $0.5\mu\text{m}$ to $50\mu\text{m}$ (Hinds, 1982). Cascade impactors consist of impaction plates (stages) placed in series and connected to a vacuum pump and flowmeter. A filter or oily surface (used to prevent particle bounce) is used as a collection surface on the plate. Each successive stage has a smaller jet nozzle diameter (entrance of air flow) and a closer impaction plate to successively collect smaller particles. Large particles in an airstream are unable to follow the air around the right angle turns and are collected on an impaction plate in the first stage. This process is repeated as the airstream flows down the device. The filter positioned after the final stage collects the smallest particles. Each stage is described by an effective cut-off diameter (ECD). The ECD represents the AD for which 50% of the particles are collected and 50% pass through. The range of particles captured in each stage is determined by the manufacturer of the cascade impactor. A Sierra Series 210 Ambient 10-stage cascade impactor, with a flow rate of 3 lpm at 25 C and 1 atmosphere, was used to measure particle sizes. Samples were analyzed in a Tennelec gas-flow proportional counter four to seven days after removal. The particle size was characterized by the activity median aerodynamic diameter (AMAD) and its corresponding geometric standard deviation (GSD or σ_g). The AMAD is "the diameter of a unit density sphere with the same terminal settling velocity in air as that of the aerosol particle whose activity is the median for the entire aerosol" (ICRP 30, 1979). The AMAD was determined by counting each stage with a gas-flow proportional counter. The total activity was calculated by summing all the stages. The activity fraction for each stage was calculated by dividing the activity measured for

Table 6. Summary of aerosol particle size measurements using a 10-stage cascade impactor.

Sample No.	Sampling duration (days)	AMAD (μm)	GSD	Range of particle sizes (μm) @ 1GSD	Shape of distribution
1	3	3.75	1.6	2.3-6.0	lognormal
2	3	2.5	2.6	1.0-6.5	lognormal
3	3	4.3	2.5	1.7-11.0	lognormal
4	8	2.2	2.8	.8-6.2	lognormal
5	4	4.0	--	--	NLN
6	3	2.5	2.5	1.0-6.3	lognormal
7	3	2.2	2.5	0.9-5.5	lognormal

*AMAD~Activity Median Aerodynamic Diameter, μm

*GSD~Geometric Standard Deviation, σ_g

*Particle size range: m/σ_g to $m \sigma_g$

*NLN~non-lognormal distribution

each stage by the total activity. The cumulative activity fraction versus the ECD was plotted on log probability paper. A line was eye-fitted through the data giving most weight to data between 20% and 80% and less weight to data <10 % or >90%. A straight line indicates that the distribution is lognormal. The AMAD was obtained by taking the diameter at 50% of the activity fraction. The corresponding GSD was calculated using equation (1) below.

$$\frac{D_{84\%}}{AMAD} = \frac{AMAD}{D_{16\%}} \quad (1)$$

where $D_{84\%}$ or $D_{16\%}$ are the diameters at 84 percentile or 16 percentile.

The results of the cascade impactor are summarized in table 6. The particle size data and

distributions plots are shown in Appendix A.

Radioactive particulates have different physical and aerodynamic sizes and are usually described by a lognormal distribution because of the range of particle sizes, skewed shape and long tail. The particle sizes from the collected samples ranged from 0.8 μm to 11.0 μm . Respirable particles ($<10 \mu\text{m}$) dominated the aerosol. Sample No. 5 was non-lognormal and had the highest AMAD (4 μm). An AMAD of 4 μm indicates there was a wide range of particle sizes caused by a mixture of activity. The non-lognormal distributions may be caused by a mixture of two or more activity sources with different lognormal distributions.

Bioassay

Collected urine and fecal samples were analyzed by radiochemistry techniques. The actinide elements in the samples were purified. The inventory of radionuclides present, corresponding activity, and measurement uncertainty were measured by alpha spectrometry. An intake of an analyte (a specific radionuclide to be determined in a sample of interest) was determined by comparing the measured activity with the decision level (Bq) or minimum detectable amount (MDA). The American National Standard Institute (ANSI) defines the MDA as "the smallest amount of (activity or mass) of an analyte in a sample that will be detected with a probability β of non-detection (Type II error) while accepting a probability α of erroneously deciding that a positive (non-zero) quantity of an analyte is present in an appropriate blank sample (Type I error)." The MDA for each analyte and method of measurement was taken from the ANSI (HPS N13.30-1996). The activity in a sample that exceeded the MDA was followed by other measurements and bioassay sample collection to validate the initial result.

In the event the follow-up samples also exceeded the MDA, the time of intake was estimated (usually the midpoint between the last analysis and current analysis) and a dose was assessed using ICRP 54.

PAS vs Bioassay

In some cases, PASs are the only method of measuring individual exposure because of economic factors. The cost of a single bioassay sample (urine or fecal) for Pu, depending on the type of analysis, can range from \$300.00 to \$1,000.00 (Inkret and Lewis). The activities of the bioassay and corresponding PAS were compared to determine any correlation.

Daily intakes of plutonium, based on the PAS results for each of the seven workers, were calculated using equation (2). Daily results were summed for each worker to estimate total plutonium intake for the duration of the project.

$$A = [\tau \times C \times BR] / RPF \quad (2)$$

where

A = estimated intake activity (mBq)

τ = time (minutes) elapsed during a shift

C = concentration of the activity (Bq/m³) in air

BR = the breathing rate of an individual during moderate physical activity (m³/min)

RPF = the respirator protection factor

A value of 0.03 m³/min for the breathing rate (BR) of an individual during moderate physical

Table 7. Comparison of the activity intake of PAS and bioassay data for the seven contractor employees. ND (not detected) implies that no samples exceeded the baseline results.

Case	PAS	Bioassay (B)	B/PAS
A	1,066.7 mBq	600.0 mBq	0.56
B	466.7 mBq	400.0 mBq	0.86
C	216.7 mBq	800.0 mBq	4.0
D	100.0 mBq	1,016.6 mBq	10
E	73.3 mBq	ND	--
F	73.3 mBq	ND	--
G	35.0 mBq	ND	--

activity was taken from ICRP 23 (Reference Man). The respirator protection factor used was 1000 (ANL,1995).

Plutonium intakes based on bioassay results were summed for each worker to yield the total intake for the entire project. The measurements of all urine samples collected for bioassay were below the decision criteria used by ANL. The plutonium activity intakes for each worker, as estimated by PASs and measured by bioassay, are shown in table 7. There is no significant correlation between PAS and bioassay measurements. The results differed by a factor of 20. Factors that could have affected the results and produced large uncertainties in both the PAS and bioassay measurements include: inadequate representation of the breathing zone concentrations and fraction of respirable particles, variability in individual breathing rates, uncertainty in the respirator protection factor, and the uncertainty in mix of inhaled and ingested plutonium in fecal samples. Physical and physiological parameters, such as the metabolism of the radio-nuclides and the variations in excretion rates differ from person to person. Another difference

Table 8. Summary of the estimated intakes of contractor employees on the 34th workday for PAS and bioassay.

CASE	PAS	BIOASSAY	B/PAS
D	33.3 mBq	700.0 mBq	21.00
A	1,033.3 mBq	600.0 mBq	0.58
B	383.3 mBq	400 mBq	1.04

was the availability of data from PAS and bioassay. There were PAS data for each day the contract employees worked, whereas the bioassay data were limited to five samples per worker.

Special bioassays were conducted following the high activity measured on the 34th workday. This day accounted for the majority of the plutonium intakes estimated by the PAS during the project. Estimated intakes measured for the 34th workday (table 8) show little correlation between the PAS and bioassay results. Case B does show similar results.

Conclusion

The results of the comparison of the PAS and SAS are summarized in table 9.

Table 9. Summary of observations of the PAS and SAS.

PAS vs SAS	Observations
Concentration Frequency Distribution	Average concentrations of PAS and SAS are 261.62 mBq/m ³ and 248.03 mBq/m ³ respectively.
Ratio of PAS to SAS	>80% agree within a factor of 3
Self Absorption of Filters	conservative factor of 1.3
Particle Size	Most lognormally distributed. Ranged from 0.8μm to 11.0 μm @ 1GSD.

The mean concentrations for the PAS and SAS for the 53 workdays were 261.68

mBq/m³ and 248.03 mBq/m³ respectively. The majority of the activity concentrations between the PAS and SAS agree within a factor of 3. The PAS and SAS are equivalent when averaged over all employees and all workdays, but the standard deviation is large and individual samples can have a large deviation. The differences between the PAS and SAS can be explained by spatial variation in the concentrations as a result of change in ventilation patterns such as a worker changing position close to the source or the opening or closing of a door. Studies (Marshall, 1976; Marshall and Stevens, 1979) have shown that the concentration in an area can vary by orders of magnitude within one meter of the source, limiting the SAS to collect a fraction of what is collected by the PAS.

Correlation between air monitoring and bioassay appears poor. Other studies have also shown a lack of correlation (Alvarez, 1994). Lack of correlation may be a result of the anatomical, physiological and behavioral characteristics of an individual employee. Therefore, PAS and bioassay should play complementary roles in assessing estimated intakes. The PAS is used to detect and evaluate contaminated atmospheres, and bioassay is a definitive measure to determine the individual intakes and internal dose; hence, bioassay should be recommended as an additional measure to air monitoring where the risk of significant exposure is relatively high.

Possible improvements in the air monitoring program would include evaluation of the air flow dynamics in the work area to optimize position of air samplers (CAMs, PASSs, SASs, and cascade impactors) and the determination of the correlation between self-absorption factors and collection efficiencies in the various filter matrices.

Determination of ventilation patterns (usually with smoke candles) can assist in positioning air samplers at locations where there is an apparent high possibility of detecting a

contamination event. An air monitoring program that uses both PAS and SAS technologies should use the same type of filter medium or provide an inter-filter correlation of the two systems. Radiochemical analysis can be used to determine an appropriate self-absorption factor for a particular filter type.

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Appendix A

Day	PAS 1 Morning	PAS 2	PAS 3 mBq/m3	PAS 1 Afternoon	PAS 2	PAS 3 mBq/m3	Day	South Morning	North	West mBq/m3	South Afternoon	North	West mBq/m3	Avg PAS mBq/m3	Avg SAS mBq/m3
2	6.666667	23.33333	0	13.33333	30	30	2	0	18.33333	33.33333	1.833333	8.666667	81.66667	17.22222	23.97222
3	0	90	56.66667	33.33333	416.6667	10	3	5.166667	500	110	6.833333	38.33333	80	101.1111	123.3889
4	456.6667	490		111.6667	30	236.6667	4	43.33333	360	690	60	255	273.3333	265	280.2778
5	108.3333	0	0				5	13	113.3333	843.3333	0	0.016667	0	36.11111	161.6139
6	430	555	641.6667	38.33333	98.33333	65	6	93.33333	153.3333	2500	4.333333	126.6667	266.6667	304.7222	524.0556
7	63.33333	41.66667	48.33333	53.33333	48.33333	305	7	9.333333	21.66667	38.33333	11	36.66667	66.66667	93.33333	30.61111
8	63.33333	0	35	0	11.66667	0	8	4.333333	7.333333	3.166667	206.6667	5	7	18.33333	38.91667
10	0	0	11.66667	0	0	0	10	2.333333	16.66667	11.83333	0	0.666667	1.833333	1.861111	5.555556
11	255	2741.667	0	276.6667	60	296.6667	11	96.66667	650	386.6667	93.33333	116.6667	63.33333	605	234.4444
12	176.6667	141.6667		48.33333	0	103.3333	12	12.5	85	61.66667	9	13.16667	23.33333	94	34.11111
13	0	73.33333	0	15	6.666667	0	13	0.7	36.66667	11.5	3.666667	3.166667	7	15.83333	10.45
14	2381.667	133.3333	58.33333	0	0	38.33333	14	73.33333	1250	2116.667	66.66667	373.3333	1245	435.2778	854.1667
15	158.3333	0	18.33333	80	233.3333	60	15	73.33333	36.66667	56.66667	11.16667	123.3333	146.6667	91.66667	74.63889
16	0	0		0	0	36.66667	16	2850	11.83333	4.166667	1.333333	1.333333	6.5	7.333333	479.1944
17	233.3333	1198.333	138.3333	303.3333	418.3333	255	17	65	460	298.3333	40	358.3333	600	424.4444	303.6111
18	5	73.33333	175	73.33333	5	0	18	1750	86.66667	46.66667	18.33333	51.66667	171.6667	55.27778	354.1667
19	40	116.6667	301.6667	465	1073.333	0	19	165	448.3333	2128.333	13.83333	126.6667	158.3333	332.7778	506.75
20	4773.333	658.3333	201.6667	796.6667	841.6667	1186.667	20	88.33333	838.3333	1135	238.3333	1856.667	1933.333	1409.722	1015
21	66.66667	6.666667	0	6.666667	426.6667	15	21	31.66667	30	2.166667	6.5	11.16667	14.83333	86.94444	16.05556
22	25	10	0	116.6667	138.3333	1536.667	22	21.66667	10.33333	5.166667	10.33333	663.3333	248.3333	304.4444	159.8611
23	345	225	81.66667	8.333333	96.66667	60	23	48.33333	168.3333	126.6667	41.66667	100	68.33333	136.1111	92.22222
24	200	528.3333	111.6667	78.33333	18.33333	475	24	51.66667	281.6667	250	20	24.16667	20	235.2778	107.9167
25	0	230	720	26.66667	308.3333		25	116.6667	1038.333	728.3333	161.6667	1410	250	257	617.5
26	203.3333	8.833333		883.3333	1081.667	321.6667	26	51.66667	25	18.33333	138.3333	503.3333	265	499.7667	166.9444
27	40	103.3333	20	55	4.5		27	16.66667	110	80	0.066667	0.133333	0.166667	44.56667	34.50556
28	1148.333	783.3333		190	975	843.3333	28	258.3333	490	686.6667	68.33333	556.6667	711.6667	788	461.9444
29	253.3333	0	0	271.6667	265	235	29	2.333333	0	4.166667	10.5	423.3333	866.6667	170.8333	217.8333
30	118.3333	228.3333	41.66667	490	103.3333	201.6667	30	65	63.33333	40	85	331.6667	568.3333	197.2222	192.2222
31	46.66667	160	386.6667	83.33333	16.66667	20	31	20	628.3333	443.3333	43.33333	7.666667	30	118.8889	195.4444
32	248.3333	420	116.6667	86.66667	71.66667	98.33333	32	48.33333	2238.333	3095	26.66667	36.66667	118.3333	173.6111	927.2222
33	398.3333	1128.333	20	143.3333	791.6667	780	33	23.33333	115	30	86.66667	721.6667	1235	543.6111	368.6111
34	78.33333	36.66667	346.6667	6786.667	278366.7	101066.7	34	28.33333	236.6667	131.6667	44108.33	205783.3	152866.7	64446.94	67192.5
37	131.6667	1001.667	71.66667	638.3333	25	95	37	325	430	250	31.66667	1195	161.6667	327.2222	398.8889
38	391.6667	111.6667	80	1008.333	366.6667	263.3333	38	138.3333	55	181.6667	355	535	581.6667	370.2778	307.7778
39	360	0	65	6.666667	73.33333	1520	39	2.166667	1.5	12.5	3	2.166667	23.33333	337.5	7.444444
41	0	0		0	66.16667	41.83333	41	13.33333	15.66667	11	38.33333	6.833333	5.333333	21.6	15.08333
42	12.5	0		47.83333	112.1667	0	42	13.16667	1.5	8.666667	16.66667	16.16667	80	34.5	22.69444
43	62.66667	3.5	3.666667	136.5	197.3333		43	35	9.166667	60	398.3333	46.66667	166.6667	80.73333	119.3056
44	9.833333	19.5	20.5	0	9.333333	37.33333	44	110	130	31.66667	255	16.66667	16.66667	16.08333	93.33333
46	59.33333	0	45.5	10.83333	18.16667		46	73.33333	68.33333	105	6.5	10.5	9.333333	26.76667	45.5
47	0	25.5	36	77.83333	515	77.16667	47	75	25	31.66667	173.3333	14.33333	48.33333	121.9167	61.27778
48	6.333333	27.66667		53.5	322.1667	0	48	163.3333	101.6667	14.5	1.333333	1.333333	1.166667	81.93333	47.22222
50	0	0		0	18.83333	0	50	1.833333	0	40	11.83333	23.33333	45	3.766667	20.33333
51	0	0	18.66667	15.16667	0	72.33333	51	10.83333	15.83333	51.66667	18.33333	28.33333	145	17.69444	45
52	565.6667	295.1667		3344.333	5297.667	1528	52	241.6667	1078.333	1508.333	541.6667	1290	2036.667	2206.167	1116.111