

CHARACTERIZATION OF PLUTONIUM AEROSOL COLLECTED DURING AN ACCIDENT

Yung Sung Cheng,* Raymond A. Guilmette,[†] Yue Zhou,* Jun Gao,* Thomas LaBone,[‡] Jeffrey J. Whicker,[†] and Mark D. Hoover[§]

Abstract—This study determined the plutonium particle size distribution and dissolution rate of $^{238}\text{PuO}_2$ aerosol collected during the 16 March 2000 release of an undetermined amount of $^{238}\text{PuO}_2$ in a room within a plutonium facility at Los Alamos National Laboratory. The facility has been in operation since 1978 to support the development, fabrication, and testing of ^{238}Pu heat sources for the U.S. Department of Energy. Several workers were in the room at the time of the release and in vivo study of five of the workers began the day after the exposure event. Four of the subjects subsequently received chelation therapy. Over 30 fixed air filter samplers (FASs) and four continuous air monitors (CAMs) were operating in the room during the radiological release. One 47-mm-diameter glass fiber FAS filter and one 25-cm-diameter mixed cellulose ester CAM filter containing Pu aerosol from the incident were examined in the study described here. Total alpha radioactivity on the filters was determined by gross alpha counting. Isotopic identification of the ^{238}Pu was made by alpha spectrometry. Film autoradiography was used to characterize the spatial distribution of alpha-emitting particles on the filters. Track-etch autoradiography was used to estimate the distribution of alpha radioactivity in individual plutonium particles on the filters for particle size measurement. The glass fiber filter was then cut into six sections. Particles from two sections were resuspended in alcohol, dispersed as an aerosol using a Lovelace nebulizer, and characterized by aerodynamic diameter using a Lovelace Multi-jet cascade impactor. The measured activity median aerodynamic diameter from the cascade impactor was $4.8\ \mu\text{m}$ with a geometric standard deviation of 1.5. That agreed with the size distribution obtained from the alpha track detection technique. The remaining four filter sections were used in an in vitro dissolution study with synthetic serum ultrafiltrate. The retention of undissolved ^{238}Pu was consistent with a biphasic exponential function. The majority of the ^{238}Pu dissolved with a half-time of 900 d. The information on particle size distribution and solubility from this study was useful in assigning a radiation dose to the

exposed workers, supporting the decision to administer chelation therapy, and providing a model for characterizing accident-associated aerosols in the future.

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INTRODUCTION

AT APPROXIMATELY 2 p.m. on 16 March 2000, an undetermined amount of $^{238}\text{PuO}_2$ was released from a glovebox system in the Plutonium Facility (PF-4) at Los Alamos National Laboratory (LANL) (U.S. DOE 2000). The system was part of a U.S. Department of Energy program to fabricate and develop small ^{238}Pu heaters for civilian and defense applications. No plutonium was released outside the facility as a result of the incident. The release was later determined to have been caused by a loose Swagelok fitting on a dry-vacuum line connected to an airlock that serviced the glovebox. At the time of the release, an employee was performing a maintenance evaluation of the glovebox. While the employee was working on the glovebox, continuous air monitors (CAMs) in the room alarmed, indicating the presence of airborne radioactive material thus prompting the evacuation of the room.

Five of the eight workers in the room at the time of the release had positive nasal swipes. Chelation therapy at LANL's occupational medicine facility was initiated within 3 h of the incident for four of the employees. At the same time that workers were undergoing chelation therapy, efforts were initiated to characterize the particle size distribution and the solubility of aerosols released during the incident. Both particle size distribution and solubility information are useful for radiation dose estimate (ICRP 1994) but are not always available during plutonium accidents.

A number of historical studies have shown that the physicochemical properties of plutonium oxide particles can vary depending on the source and history of the material (Anderson et al. 1970; Ramsden et al. 1970;

* Lovelace Respiratory Research Institute, 2425 Ridgecrest Drive SE, Albuquerque, NM 87115; [†] Los Alamos National Laboratory, PO Box 1663, Los Alamos, NM 87545; [‡] Savannah River Site, Building 735-4B, Aiken, SC 29808; [§] National Institute for Occupational Safety and Health, Centers for Disease Control and Prevention, 1095 Willowdale Road, Morgantown, WV 26505.

For correspondence or reprints contact: Y. S. Cheng, Lovelace Respiratory Research Institute, 2425 Ridgecrest Drive SE, Albuquerque, NM 87115, or email at ycheng@lrri.org.

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Newton et al. 1983; Carbaugh et al. 1991; Guilmette et al. 1994; Hickman et al. 1995). Lung scans were taken to measure initial lung burden (Anderson et al. 1970; Newton et al. 1983; Carbaugh et al. 1991), and bioassay samples were taken from the exposed workers to assess the excretion rates of plutonium from the body and to estimate lung burden and radiation dose based on an assumed dissolution rate of plutonium aerosols (Anderson et al. 1970; Ramsden et al. 1970; Newton et al. 1983; Carbaugh et al. 1991; Guilmette et al. 1994; Hickman et al. 1995). In one case, samples were taken for a solubility test (Anderson et al. 1970), and in another case, airborne filter or personal samples were taken for measurement of activity concentration and particle size distribution (Ramsden et al. 1970).

Dissolution of plutonium oxide particles using lung simulants and solvents has shown that the dissolution rate of ^{239}Pu oxides is slow (absorption Type S according to the ICRP 66 definition), but ^{238}Pu oxide has higher solubility (Kanapilly et al. 1974; Miglio et al. 1977; Vashi et al. 1980). Also, the solubility of the plutonium oxide increases as the firing temperature increases when the oxide particles are formed (Miglio et al. 1977). The solubility could also change as the material ages (Park et al. 1974). In vivo solubility of plutonium oxides in the lung of laboratory animals and in exposed workers in general have shown similar dissolution behavior (Bair et al. 1962; Guilmette et al. 1984; Carbaugh et al. 1991). However, the dissolution behavior of ^{238}Pu oxide observed in vivo has shown properties of either absorption Type S (Newton et al. 1983) or Type M (medium absorption half-times) (Mewhinney and Diel 1983).

The purpose of this study was to characterize the radioactivity, including alpha spectra; the particle size distribution; and the dissolution rate of the plutonium aerosol collected on filters from the accident. This information was useful in assessing the exposure level and characteristics of the material inhaled by the workers. It was also essential for better estimating the radiation dose to the exposed workers and may be useful in guiding future decisions regarding chelation treatment.

CHARACTERIZATION OF THE COLLECTED AEROSOLS

Filter samples from the incident

Over 30 fixed air filter samplers (FASs) as well as four alpha continuous air monitors (CAMs) were operating in the room at the time of the incident. A 47-mm-diameter FAS filter (LANL sample identification number FAS 11) (Type A/E glass, Gelman Sciences, Ann Arbor, MI) from a sampler approximately 2 m from the release location and a 25-mm-diameter CAM filter (LANL

sample identification number CAM B) (SMWP, mixed cellulose ester, 5- μm pore size, Millipore Corp., Bedford, MA) containing plutonium aerosols from the incident were sent to Lovelace Respiratory Research Institute (LRRI) for analyses.

Gross alpha counting of the filters

The initial measurements performed on the filter samples were nondestructive gross alpha counting by the zinc sulfide method to quantify collected radioactivity. The counting system consisted of a Ludlum PRO031800 detector (Model 218, Ludlum Measurements, Inc., Sweetwater, TX) attached to an Eberline Smart Radiation Monitor (Model SRM-200, Eberline, Santa Fe, NM). The counting system was optimized and calibrated with a set of ^{239}Pu counting standards prior to counting the filter samples. The counting efficiency was 0.504 ± 0.021 counts per disintegration. Gross alpha counts were $3.84 \times 10^4 \pm 2.49 \times 10^3$ Bq (1.04 ± 0.07 μCi) for the FAS filter and $3.01 \times 10^3 \pm 6.68 \times 10^2$ Bq (0.81 ± 0.02 μCi) for the CAM filter.

Isotopic identification of the plutonium

The alpha energy spectrum of the sample collected on the CAM filter was evaluated using the alpha spectrometer feature of an Eberline Alpha-7 CAM (Eberline, Santa Fe, NM) (see Fig. 1). The slight attenuation of the peak toward the low-energy side is due in part to the presence of the thin protective layer of Mylar (mass thickness of 0.22 mg cm^{-2}) that was placed over the filter prior to placing the filter in the spectrometer, and in part to a small amount of shielding within the filter matrix, as compared to the ideal spectrum shown as a solid black

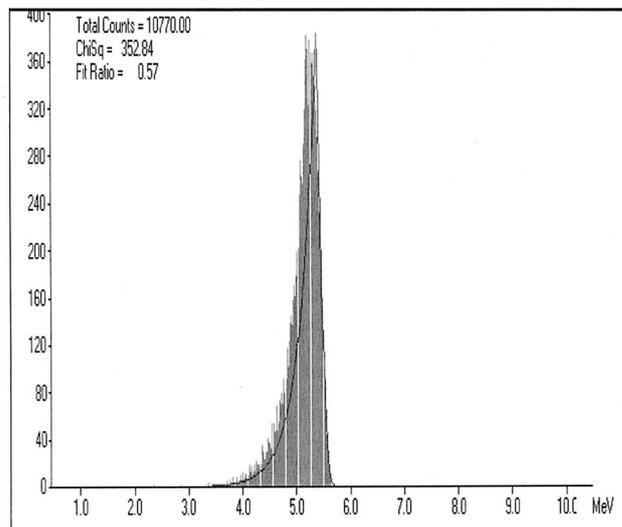


Fig. 1. Alpha spectrometry results demonstrating the presence of ^{238}Pu on the CAM filter sample.

line on the figure. A wipe sample from a filter separation paper that was packaged with the filter when it was removed from the CAM was also evaluated by alpha spectrometer and found to contain ^{238}Pu . That indicated that some plutonium may have been lost in the packaging and handling process. Alpha spectroscopy was not performed on the FAS filter because the coarser nature of the glass fiber filter matrix degrades the alpha energy spectrum.

Film alpha autoradiography

A qualitative assessment of the particle deposition patterns on the filters was made by film alpha autoradiography analysis. This procedure consisted of placing each filter/Mylar sandwich between two pieces of Polaroid, Type-57, high-speed, black-and-white film (Polaroid, Cambridge, MA). The emulsion side of the first piece of film was exposed to the collection surface of the filter, while the emulsion side of the second piece of film was exposed to the backside of the filter. The alpha particles collected on the filters exposed localized areas on the film, thus providing a means of characterizing the deposition patterns for each filter. The autoradiography of the two filter samples indicated the relatively uniform activity of the filters (Fig. 2).

Alpha track-etch autoradiography

Polyallyl diglycol carbonate (PADC) chips (Track Analysis Systems Limited, Bristol, UK) were exposed to ^{238}Pu alpha emissions by centering the chips on the glass fiber FAS filter. PADC chips are a clear, stable, and rigid plastic material with a density of 1.30 g cm^{-3} . Mylar was placed between the filter and the PADC chips to prevent cross contamination of the ^{238}Pu from the filter to the PADC chips. Exposure times of 30, 60, and 120 s were used. Following exposure, the PADC chips were etched

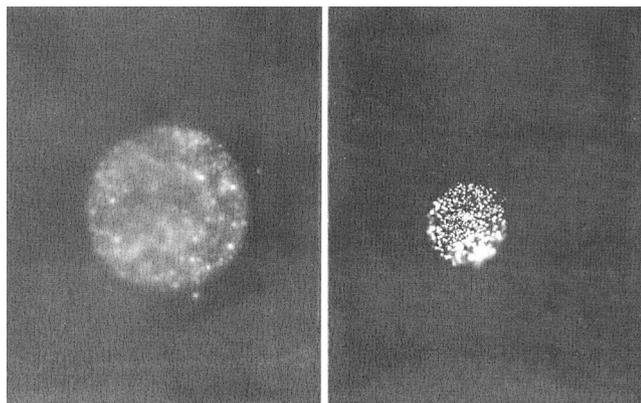


Fig. 2. Autoradiography of the 47-mm-diameter FAS filter (left) and the 25-mm-diameter CAM filter (right).

for 3 h using 6.25 N NaOH at $70\text{ }^\circ\text{C}$. To stop the etching process, the chips were then placed in 0.1 N HCL at room temperature and rinsed in distilled water.

The etched surface was then examined by transmission electron microscopy (TEM) (Model 109, Carl Zeiss Inc, Thornwood, NY). All of the alpha-track images on each PADC chip were recorded and stored in electronic files. Alpha tracks for each particle were identified and counted using image-analysis software (Optimas, Media Cybernetics, Silver Spring, MD). From the number of tracks detected from each particle, the particle volume equivalent diameter, d_{ve} , was calculated using the Leary equation (Leary and Smith 1951; Hayden 1976):

$$d_{ve} = \left(\frac{12MC}{\lambda N_o \pi \rho f t} \right)^{1/3}, \quad (1)$$

where M is the molecular weight of the compound (274 for $^{238}\text{PuO}_2$); C is the disintegration per second; λ is the decay constant; N_o is Avogadro's number; ρ is the density (11.46 g cm^{-3} for $^{238}\text{PuO}_2$); t is exposure time; and f is the number of radioactive atoms in the compound. This equation assumed a 50% counting efficiency on the chip and a spherical particle.

To compare the aerodynamic particle size measurement of the cascade impactor (see below) and facilitate dose calculation, the volume equivalent diameter (d_{ve}) was converted to the aerodynamic diameter (d_{ae}) using the following equation (Cheng et al. 1988; ICRP 1994):

$$d_{ae} = d_{ve} \sqrt{\left[\frac{\rho C(d_{ve})}{\chi \rho_o C(d_{ae})} \right]}, \quad (2)$$

where ρ and ρ_o are particle and unit densities, respectively; and χ is the dynamic shape factor. C is the slip correction factor (Allen and Raabe 1982).

The activity size distribution can be obtained by counting the number of alpha tracks for each particle because one track indicates one alpha decay. For dose calculation as well as for comparison to the cascade impactor measurement, the aerodynamic equivalent diameter (d_{ae}) was calculated from d_{ve} using eqn (2). Because the true dynamic shape factor (χ) was unknown, two calculations were made: assuming $\chi = 1$ for spherical shape, which is consistent with the assumption used in eqn (1), and assuming $\chi = 1.5$, which is a default value used in the ICRP lung model (1994) for the dose calculation. Fig. 3 shows the activity distribution of aerodynamic size for these two cases. The size distribution can be expressed as a lognormal distribution. The activity median aerodynamic diameter (AMAD) and σ_g are listed in Table 1.

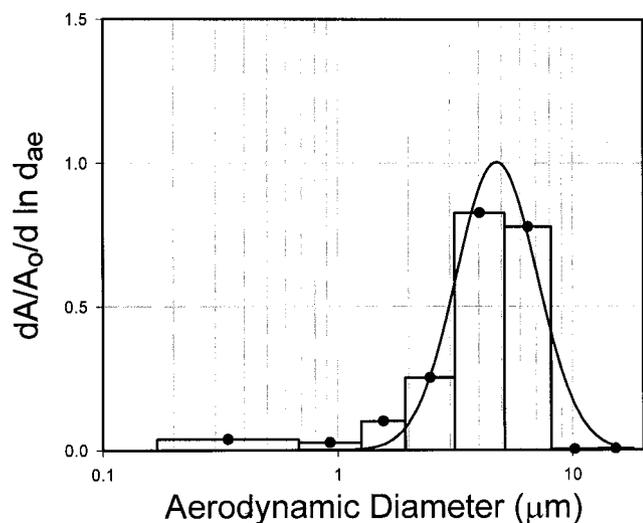


Fig. 3. Aerodynamic size distribution and the fitted lognormal distribution from the cascade impactor measurement of ^{238}Pu particles resuspended from the FAS filter.

Table 1. Activity distribution of d_{ae} assuming dynamic shape factors of either 1.0 or 1.5.^a

Dynamic shape factor χ	AMAD (μm)	σ_g
1	4.7	1.4
1.5	3.8	1.4

^a Note: AMAD and σ_g values were calculated from measurements of 945 individual particles.

Aerodynamic particle size determination by resuspension and aerosolization

The FAS filter was cut into six equal sections (Fig. 4). Sections 1 and 6 of this filter were used in the resuspension and aerosolization part of this study to size the aerosol that had been collected on the FAS. The filter was placed into a vial with ethyl alcohol and subjected to ultrasonic agitation for 10 min. Fig. 5 shows the aerosol generation and particle size measurement system. A Lovelace nebulizer (In-Tox Products, Albuquerque, NM) was used to generate the particles in suspension. Typically this nebulizer is run at 20 pounds per square inch (psi) and produces a flow rate of 1.5 L min^{-1} . The aerosol was then passed through a radial diluter and up through an activated carbon chamber, which dried the particles. A ^{85}Kr discharge tube was placed within the activated carbon chamber for charge neutralization. A Lovelace Multi-jet Cascade Impactor (LMJ) run at 14.0 L min^{-1} was used to collect and size the aerosol distribution.

The LMJ impactor consists of seven stages and a backup filter (Newton et al. 1977). The 50% cut-off aerodynamic diameter ranged from 0.68 to $12.8 \mu\text{m}$. The LMJ was started in conjunction with the release of the aerosol and allowed to run until the full 20 mL of

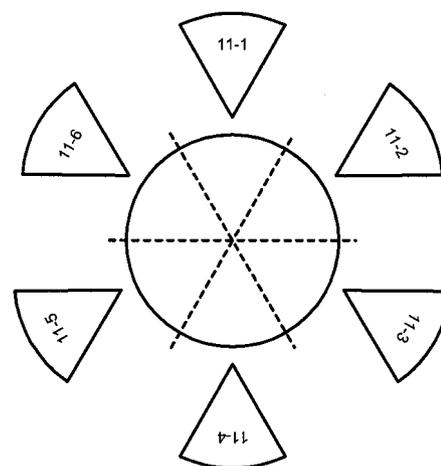


Fig. 4. Illustration of how the FAS filter was cut into six sections for use in the resuspension and dissolution studies (11-1: Resuspension/aerosolization; 11-2: Dissolution; 11-3: Dissolution; 11-4: Dissolution; 11-5: Dissolution; and 11-6 Resuspension/aerosolization).

generator solution was consumed. Following the sampling run, the impactor was disassembled, and the substrates and backup filter were counted to determine the activity distribution of the aerosol.

Fig. 6 shows the impactor data obtained from the two tests that were performed when the resuspended material was aerosolized from FAS sections 1 and 6. The particle size distribution can be described by a lognormal distribution as shown in Fig. 6. The best-fitted lognormal distribution, obtained using SigmaPlot 2000 software (SPSS, Chicago, IL), has an activity median aerodynamic diameter (AMAD) of $4.8 \mu\text{m}$ and σ_g of 1.5.

TEM and SEM analysis of the plutonium particle size distribution

An electrostatic precipitator was used to collect the resuspended aerosol onto an electron microscope substrate for transmission electron microscopic (TEM) and scanning electron microscopic (SEM) analyses. It was operated during the sampling period of the impactor measurement. Attempts were made to characterize the particle size and morphology of the plutonium particles. Both TEM and SEM were used; however, we could not positively identify plutonium on the filter. It was likely that the plutonium particles were a small fraction of material collected on the filter. The morphology of particles observed under the microscope showed non-spherical particle shape.

Comparison of the aerodynamic and track-etch particle size distributions

Fig. 7 shows a comparison of activity distributions of aerodynamic size from the impactor and alpha track

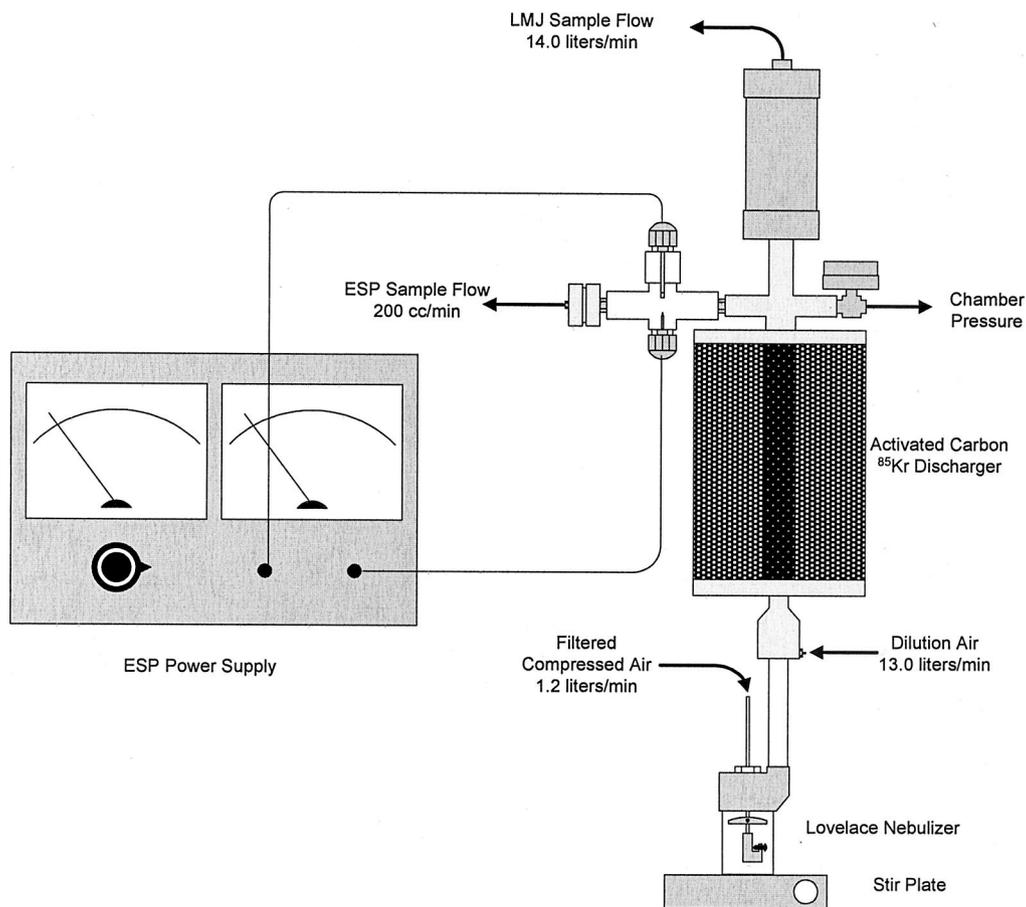


Fig. 5. Schematic of the system for aerosol generation and collection.

analyses. It appeared that the impactor data and the alpha track data agreed well if a dynamic shape factor of 1 was assumed. A slightly smaller size distribution was obtained when a shape factor of 1.5 was assumed. This does not mean that the shape factor of 1 is the correct value for dose assessment; most irregular-shaped particles have a shape factor greater than one. As shown below, both values of AMAD ($3.8 \mu\text{m}$ from the alpha track analysis and $4.7 \mu\text{m}$ from the cascade impactor analysis) were used to estimate the dose of exposed workers and then compared.

Determination of solubility by in vitro dissolution

Serum ultrafiltrate (SUF) solvent (Eidson and Me-whinny 1983; Cheng et al. 1997) was used in a static dissolution system as described by Kanapilly (1977) to determine the dissolution rates of the plutonium particles. The SUF approach is based on the seminal work of Gamble (1967), who described the chemical composition of extracellular fluid. The molar concentrations of the chemicals in SUF were as follows: 0.116 moles L^{-1} NaCl; 0.010 NH_4Cl ; 0.027 NaHCO_3 ; 0.005 glycine;

0.001 L-cysteine; 0.0002 Na citrate; 0.0002 CaCl_2 ; 0.0005 H_2SO_4 ; 0.0012 NaH_2PO_4 ; 0.0002 DTPA (diethylenetriaminepentaacetic acid), a chelating agent not present in serum, but added to decrease the adherence of dissolved actinide ions to the containers; and 50 ppm alkylbenzyltrimethyl ammonium chloride, an antibacterial agent not present in serum. Each container of solvents with carbonate had a 5% CO_2 airstream passed slowly over the liquid surface of the sample to maintain the pH at 7.3 ± 0.05 .

Duplicate dissolution tests involving two filter sections each were performed. The filter sections were sandwiched between two 47-mm diameter membrane filters (Tuffryn HT-100, $0.2 \mu\text{m}$ pore size, Millipore Corp., Bedford, MA) and secured in a Teflon filter holder (Free-Flow Filter Holder 04-112, In-Tox Products, Albuquerque, NM). The filter holders were placed individually into glass beakers containing 50 mL of solution at room temperature.

Solvent samples were collected at 1, 4, 8, 24, and 96 h in the first week; twice a week for the remainder of the first month; then once a week after the first month. At

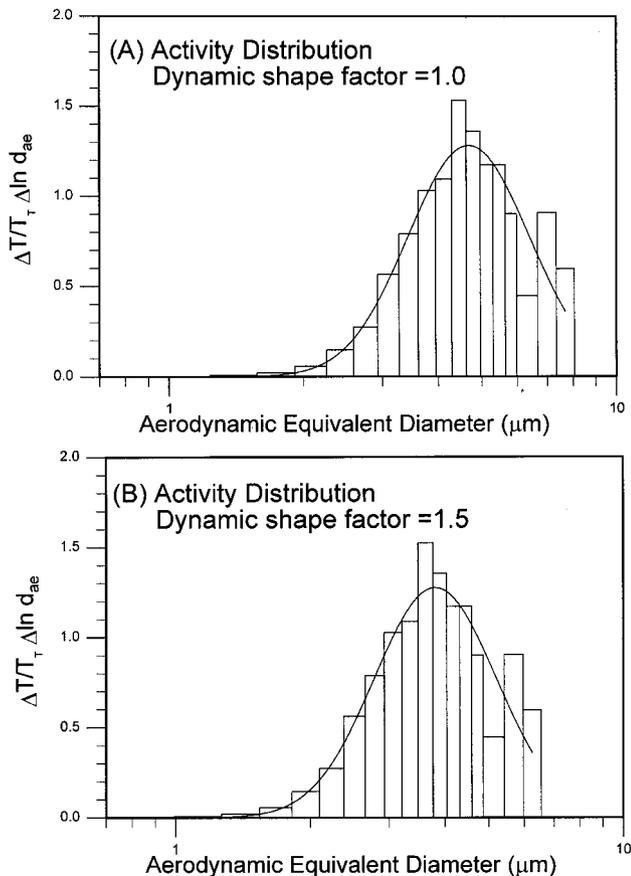


Fig. 6. Activity aerodynamic size distributions calculated from the alpha track measurement assuming different dynamic shape factors of (A) 1.0 and (B) 1.5.

the end of the experiment, all the solvent samples, the undissolved plutonium on the filters, plus the sample plastic holders and the plastic containers were analyzed for plutonium content. The analysis for plutonium in the solvent and container samples involved high-temperature ashing (550 °C) of each sample followed by digestion and dissolution in strong acid. The filter samples, which contained undissolved plutonium oxide particles, were further treated with concentrated HF to dissolve the plutonium oxide particles. Once dissolved, the plutonium activity in each sample was assayed using a two-phase solvent extraction cocktail described by Keough and Powers (1970) and alpha liquid scintillation counting. Dodecylbenzene was substituted for toluene as the organic solvent phase.

Fig. 8 shows dissolution rate for four filter sections: two for a 3-mo study and two for a 12-mo study. The dissolution rate was in the range of 0.02 to 0.06 per day in the first few hours, then it decreased to mostly between 10^{-3} and 10^{-4} per day. Filter sections 3, 4, and 5 appeared to have a similar dissolution rate, whereas filter section 2 had a slightly higher dissolution rate. The

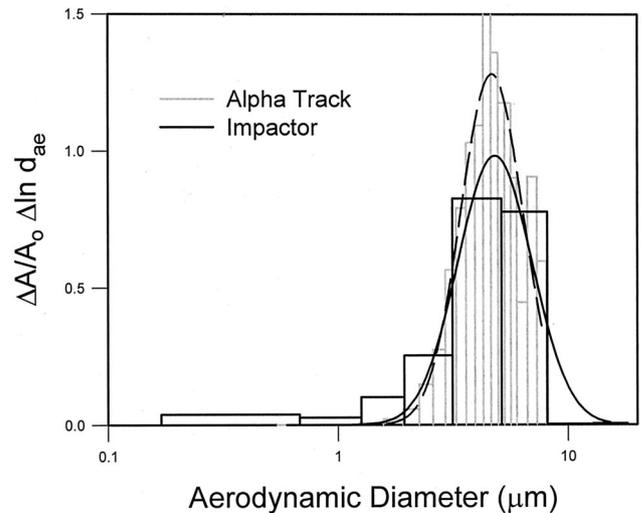


Fig. 7. Comparison of the activity aerodynamic diameter distributions obtained by cascade impaction and by alpha-track analysis (assuming a dynamic shape factor of 1.0).

dissolution rate was similar to the in vitro dissolution rate of $^{238}\text{PuO}_2$ using a similar dissolution set up and simulant, which was much higher than that of $^{239}\text{PuO}_2$ (Kanapilly et al. 1974).

Fig. 9 shows the retention of ^{238}Pu in the simulated lung serum fluid as a function of time for the two samples undergoing a 12-mo dissolution study. The retention data can be expressed by a biphasic exponential decay function using the curve-fitting procedure in SigmaPlot (SPSS Inc., Chicago, IL):

$$\frac{A}{A_0} = 0.0228 \exp(-0.298 t) + 0.9772 \exp(-6.09 \times 10^{-4} t), \quad (3)$$

where the r^2 value of the fit is 0.982. This indicates that 2.3% of ^{238}Pu dissolved with a half-time of 2.3 d, whereas 97.7% dissolved with a half-time of 1,138 d.

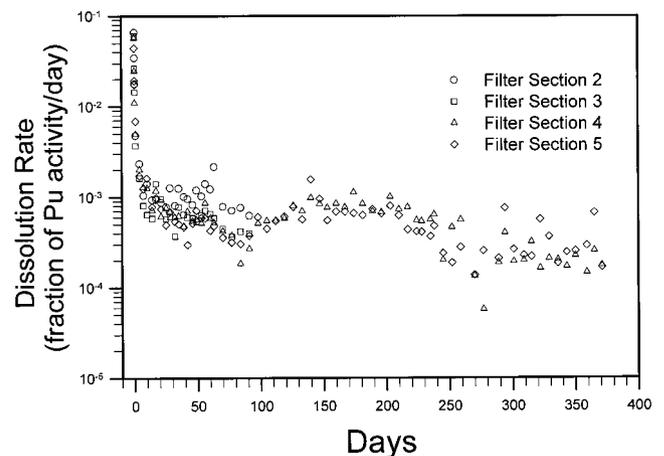


Fig. 8. Dissolution rate of ^{238}Pu in filter samples as a function of time in the simulated lung serum fluid.

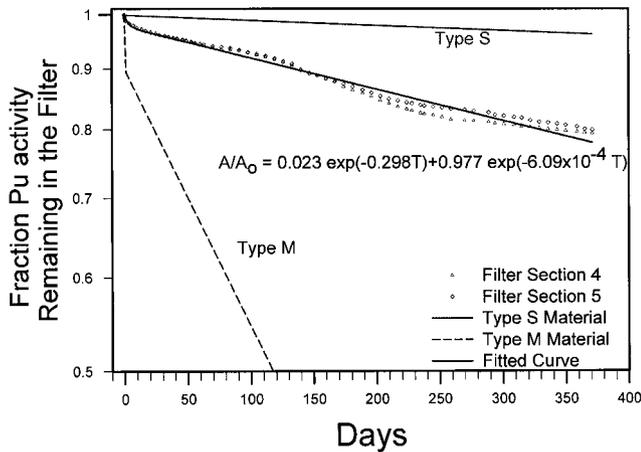


Fig. 9. Comparison of the retention of ^{238}Pu in the filter sample as a function of time in the simulated lung fluid and the ICRP 66 default assumptions for Type S and Type M materials.

Fig. 9 shows also the dissolution behavior of Type S and Type M material. In comparison to the dissolution types recommended by ICRP 66 (1994) (Table 2), this material has a dissolution property between Type M and Type S.

MODELING OF THE IN VIVO BEHAVIOR OF THE PLUTONIUM AEROSOL

The particle size distribution and dissolution rate of the measured plutonium particles were used to model the deposition pattern, chest count, and excretion data. The ICRP 66 lung dosimetric model was used to calculate the deposition fraction in the human respiratory tract (ICRP 1994). The calculation was performed for a male adult in light exercise mode. After the intake was calculated, urinary excretion was modeled using the measured dissolution rate and the ICRP 67 (ICRP 1993) biokinetic model for plutonium, modified for chelation treatment (LaBone 2002). Comparison of the predicted data for urine bioassay and chest counts was made.

Table 3 contains deposition fractions for regions of the respiratory tract calculated based on ICRP 66 Lung Dosimetric Model (ICRP 1994) for three different aerosols:

Table 3. Comparison of the calculated lung deposition fractions for the two estimated particle size distributions in this study and the default particle size distribution according to ICRP 66 (1994).^a

	3.8 μm ($\sigma_g = 1.4$)	4.7 μm ($\sigma_g = 1.4$)	5.0 μm ($\sigma_g = 2.5$)
ET	0.854	0.875	0.757
BB	0.026	0.025	0.018
bb	0.014	0.012	0.009
AI	0.057	0.038	0.045

^a ET: extrathoracic region; BB: bronchial region; bb: bronchiolar region; AI: alveolar-interstitial region.

- 3.8 μm AMAD ^{238}Pu oxide aerosol with a shape factor of 1.5, a density of 11.46 g cm^{-3} , and a geometric standard deviation of 1.4 μm ;
- 4.7 μm AMAD ^{238}Pu oxide aerosol with a shape factor of 1.0, a density of 11.46 g cm^{-3} , and a geometric standard deviation of 1.4 μm ; and
- 5.0 μm AMAD aerosol with a shape factor of 1.5, a density of 3.0 g cm^{-3} , and a geometric standard deviation of 2.5 μm (default ICRP 66 parameters for workplace aerosols).

All deposition fractions were calculated using physiological parameters for light exercise (breathing rate 1.5 $\text{m}^3 \text{h}^{-1}$).

All three types of aerosol are equally consistent with the urinary excretion and measured retention in the chest (data not shown), i.e., these bioassay data cannot be used to select which type of aerosol is the best estimate of the aerosol actually inhaled. However, the cumulative excretion of ^{238}Pu in the feces predicted with a D_{ae} of 3.8 μm aerosol most closely agrees with the observed cumulative fecal excretion (data not shown), and was therefore used to model the intake.

The urinary excretion following the intake was evaluated using the physical parameters of the 3.8 μm aerosol and physiological parameters for light exercise. The second term of the empirical dissolution function was used as presented previously, but the rate constant of the first term was changed from 0.298 d^{-1} to the ICRP 66 default of 100 d^{-1} . This change is required to adequately model the early urinary excretion. Note that because the first aliquots from the dissolution study were pulled at 1 h, a rate constant of 100 d^{-1} cannot be resolved. Fig.

Table 2. Comparison of the in vitro dissolution result from this study to the classification of dissolution behavior according to ICRP 66 (1994).

	Type F (fast)	Type M (moderate)	Type S (slow)	This study
Fraction dissolved rapidly, F_r	1.0	0.1	0.001	0.023
Rapid dissolution rate, s_r (d^{-1})	100	100	100	0.298
Slow dissolution rate, s_s (d^{-1})	0	0.005	0.0001	6.09×10^{-4}

10, using these respiratory tract parameters and the ICRP 67 systemic model (modified for chelation) (LaBone 2002), shows the observed (the points) and predicted (the line) urinary excretion. The error bars represent a 2σ measurement uncertainty. In Fig. 11, the intake calculated from the urinary excretion is used to calculate predicted retention in the chest (the line), which is compared to the observed retention (the points). Again, the error bars represent 2σ measurement uncertainty. Note that the line in Fig. 11 is not a fit to the points.

DISCUSSION

Characterization of plutonium aerosols from the accidental exposure at LANL provided important information on activity concentration, particle morphology and shape, and dissolution rate. This information was used to more accurately assess the exposure and to estimate radiation dose of the exposed workers. To these ends, the filter samples collected in the exposure area during the incident provided the most relevant samples for the study.

An extensive characterization of the two filter samples gave comprehensive data on the physical, chemical, and radiological properties. The activity counting and alpha spectrometry showed that the material collected in the room was comprised of ^{238}Pu particles; it was likely to be $^{238}\text{PuO}_2$ based on the process knowledge. Although the activities were high, we could not accurately calculate the concentration because the total exposure time was unknown.

Two techniques yielded useful information on the plutonium aerosol size distribution. Particles collected in

the filter were resuspended into an alcohol suspension, then the aerosol was sampled by a cascade impactor. Activity aerodynamic size distribution was obtained by counting the impactor stages. This technique provided the activity median diameter of $4.8\ \mu\text{m}$ and σ_g of 1.5. The advantage of this technique is that it directly measures the aerodynamic size, which is needed for a dose estimate without assumptions on the particle shape, chemical composition, and particle density. However, the efficiency of the resuspension and sampling was low (est. $<1\%$ of activity on the filter), which raised concerns about whether the resuspended samples were representative.

The alpha-track technique provided an independent means by which to check the results of the impactor analysis of particle size. We have shown here that we have obtained particle size data on volume equivalent diameter on both number and activity distributions. The size results were influenced somehow by the exposure time of the chip to the filter. Low exposure time limits and the detection limit of the small particle and extended exposure made it difficult to count many tracks for the large particles. Using the three exposure times and combining the data provided an average distribution that minimized these limitations. This technique yielded direct measurement of activity size distribution on volume equivalent diameters. The aerodynamic size distribution can also be estimated by assuming $^{238}\text{PuO}_2$ particles (density of $11.46\ \text{g cm}^{-3}$) and the dynamic shape factor. We obtained the activity size distribution based on the aerodynamic equivalent diameter. When the dynamic shape factor was assumed to be unity, the AMAD was

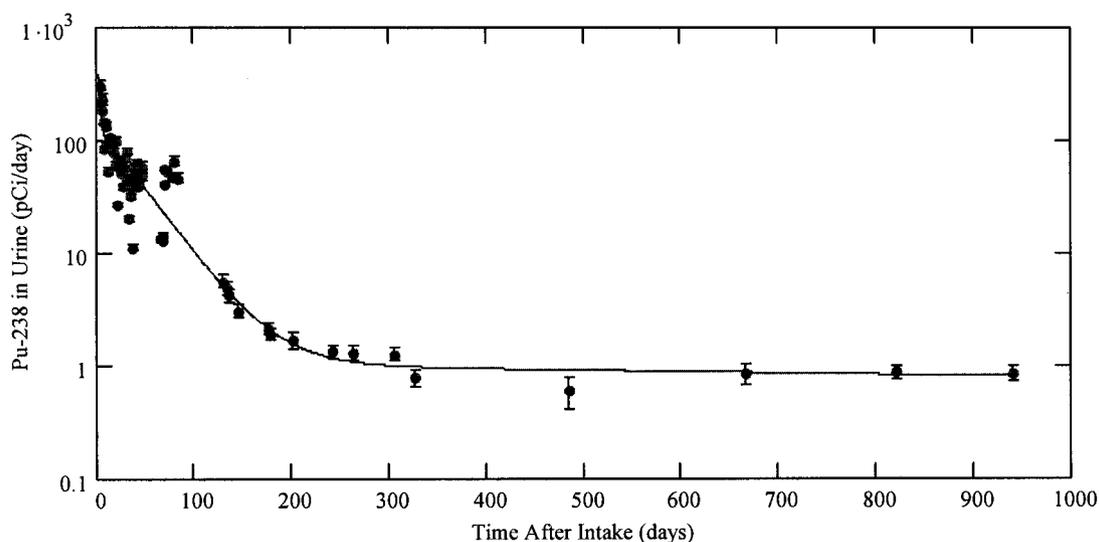


Fig. 10. Comparison of urine excretion data for an exposed worker (data points) and the model prediction from this study (curve).

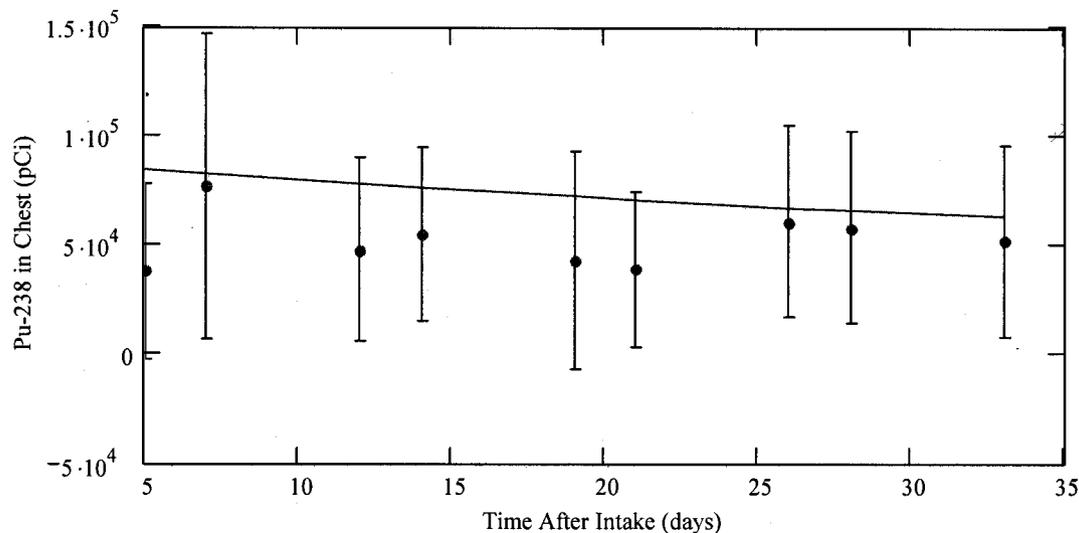


Fig. 11. Comparison of retention in the lung (chest counts) for an exposed worker (data points) and the model prediction from this study (curve).

4.7 μm ($\sigma_g = 1.4$) for the alpha-track measurement, which is very close to the impactor data (AMAD of 4.8 μm and σ_g of 1.5). A unit density was consistent with the assumption made in eqn (1) to convert the alpha-track number to the volume equivalent diameter. If a dynamic shape factor of 1.5 is used, which is the default value recommended by ICRP for occupational exposure when there is no measured value, the AMAD was smaller at 3.8 μm ($\sigma_g = 1.4$).

Overall, the impactor analysis and alpha-track detection gave consistent results on the activity aerodynamic size distribution, which minimized the error estimate of the particle size and confirmed both techniques. Additional advantages of the alpha-track technique are that it was noninvasive and the results were available within several hours following the incident. The filter samples were available after the alpha-track exposure for other studies. However, the uncertainty of the alpha track technique was that the particle shape factor was not known, which could influence its conversion from the physical diameter to aerodynamic diameter.

The *in vitro* solubility of this material indicates that it is between Type S and Type M, consistent with values reported in ICRP (1994). Comparison with urine bioassay data indicated that using the dissolution rate constants obtained from the *in vitro* measurement agreed with the experimental data if the fast rate constant was changed to the default value of 100 per day suggested by the ICRP 66 model (1994). With this modification, the *in vitro* result could be used to model the dissolution and excretion *in vivo* of this material. For future determination of *in vitro* dissolution rate, early time points such as

10 min would be needed to obtain a more accurate estimate of the fast rate constant.

The utility of the physical and dissolution data of airborne plutonium oxide particles was demonstrated by using these parameters to model the deposition, retention, and excretion in an exposed worker. Our results indicated that using physical parameters of mass median aerodynamic diameter (MMAD) = 3.8 μm ($\sigma_g = 1.4$, $SF = 1.5$) provided modeling results that agreed with the bioassay data and chest counts. However, a default value of MMAD = 5.0 μm ($\sigma_g = 2.5$, $SF = 1.5$) provided by ICRP 66 model also agreed with urine data and chest counts.

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