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Performance of Multiple HEPA Filters Against Plutonium Aerosols

for Period January 1 through June 30, 1973

los alamos
scientific laboratory
of the University of California
LOS ALAMOS, NEW MEXICO 87544

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by

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PERFORMANCE OF MULTIPLE HEPA FILTERS AGAINST PLUTONIUM AEROSOLS

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Harry J. Ettinger, Project Manager, John C. Elder, and Manuel Gonzales

ABSTRACT

Field sampling has provided general criteria defining plutonium aerosol size characteristics and activity concentrations from typical plutonium operations. Two fabrication facilities have aerosols with activity median aerodynamic diameter's (amad) ranging from 2 to 5 μm; the two research and development facilities indicate amad's ranging from 1 to 4 μm; and a recovery facility consistently shows a sub-micron aerosol with a typical amad of 0.3 μm. This recovery facility also produces aerosols as small as 0.1 μm amad, has the highest activity concentration, and constitutes the most difficult air cleaning problem.

Using laboratory produced plutonium test aerosols with size characteristics similar to those defined by the field sampling program, multiple HEPA filter systems were evaluated to provide quantitative data defining performance of successive stages of HEPA filters, and filter performance as a function of particle size. Test data show that the first and second HEPA filter each provide overall efficiencies in excess of 99.99%, while the third HEPA filter provides an average efficiency in excess of 99.8%. These performance levels exceed AEC requirements. Data defining performance of the first and second HEPA filters as a function of plutonium aerosol size show that HEPA filter efficiencies are in excess of 99.99% for sub-micron plutonium aerosols.

Theoretical calculations estimating alpha radiation dose to the filter fibers from particles collected on the fibers indicate a dose on the order of 10^6 rads per minute.

I. SUMMARY

Field sampling and analysis of data from LASL, Rocky Flats, and Mound Laboratories has been completed to provide general criteria defining plutonium aerosol size characteristics and activity concentrations from typical plutonium operations. The two fabrication facilities have aerosols with activity median aerodynamic diameter's (amad) ranging from 2 to 5 μm; the two research and development facilities indicate amad's ranging from 1 to 4 μm; and the single recovery facility consistently shows a sub-micron aerosol with a typical amad of 0.3 μm. This recovery facility also produces aerosols as small as 0.1 μm amad, has the highest activity concentration, and constitutes the most difficult air cleaning problem.

Various data handling techniques were developed to indicate the variations in aerosol size characteristics at each facility. These included definition of the weighted average aerosol size parameters, evaluation of daily variations in aerosol size characteristics, and estimation of

the validity of using a lognormal size relationship. Based on criteria developed, approximately 85 to 90% of the samples obtained can be approximated by a lognormal size distribution.

Initially difficulties with the experimental test system were experienced due to alpha radiation damage to the plastic nebulizers and other system components, and background counting problems due to the collection of radon-thoron daughter products, but these have been eliminated. Size characterization of the ball milled oxide indicates plutonium aerosols ranging in activity median aerodynamic size (amad) from 0.7 to 1.6 μm, with a geometric standard deviation ranging from 2.1 to 2.9, and an activity concentration of approximately 2×10^{12} dpm/m^3, can be prepared. Various modifications of the ball milling procedures including use of centrifugal wet milling techniques are being tested to determine if this range of aerosol sizes can be increased to better represent the sub-micron aerosol found at Location 11. Use of Nuclepore filters for sampling was discontinued when testing indicated the collection efficiency of this media was only 90% against sub-micron particles. Millipore AA filters are now being used.

Using laboratory produced plutonium test aerosols with size characteristics similar to those defined by the field sampling program, multiple HEPA filter systems were evaluated to provide quantitative data defining performance of successive stages of HEPA filters, and filter performance as a function of particle size. These data show that the first and second HEPA filter each provide overall efficiencies in excess of 99.99%, while the third HEPA filter provides an average efficiency in excess of 99.8%. Two tests indicating efficiencies less than 99.8% for the third HEPA filter appear to be artifacts due to the extremely poor statistics involved when counting samples downstream of the third HEPA filter. These samples indicate less than 0.5 counts per minute, and minor contamination, or background changes, will occasionally indicate a spurious high count resulting in a suggested lower filter efficiency. Data defining performance of the first and second HEPA filters as a function of plutonium aerosol size show that HEPA filter efficiencies are in excess of 99.99% for sub-micron plutonium aerosols.

While these data provide reasonably conclusive evidence that multiple HEPA systems perform at or above AEC requirements for plutonium aerosols as small as 0.4 μm, additional tests are scheduled using higher plutonium aerosol concentrations, longer sampling periods, and smaller aerosols. The experimental system is being modified to permit generation and sizing of plutonium aerosols as small as 0.1 μm, to provide data for conditions similar to those existing in the plutonium recovery facility (Location 11). Performance data for these smaller aerosols are necessary to guarantee the performance of air cleaning systems for some plutonium operations.

Theoretical calculations are being performed to estimate alpha radiation dose to the filter fibers from particles collected on the fibers. Initial calculations suggest that this dose is on the order of 10^6 rads per minute, and this will be compared with published data indicating the effects of gamma radiation on HEPA filters.

II. FIELD SAMPLING

A. Background

Field sampling to determine Pu particle size characteristics and alpha activity concentration has been performed immediately upstream of the exhaust HEPA filters at five locations: two each at Mound Laboratory and Rocky Flats Plant, and one at LASL. These locations were selected to monitor Pu aerosols produced by typical research and production operations utilizing both ^{238}Pu and ^{239}Pu. Samples were obtained during the most active periods of the working day, when activity concentrations could be termed

"worst normal" and most source operations would be normally contributing plutonium aerosols to the process ventilation system. Many variables were expected to affect size characteristics and activity concentration, resulting in a range of these parameters for each facility. These include types of operations, isotope mixtures, chemical form, quality of prefiltation, and quantity of material handled. The relationship between some of these variables and the individual sampling sites are summarized in Table I. The predominant chemical form at each plant was reported to be PuO_2, although a detailed chemical analysis of each sample was not performed. Fabrication plants perform primarily mechanical operations such as grinding, welding, machining, and foundry, whereas the recovery plant predominantly performs chemical conversion processes, such as acid dissolution, precipitation and drying, and calcination of oxides in powder form. Research and development operations included mechanical and chemical processes similar to those at fabrication and recovery plants but on a much smaller scale and not on a continuous basis.

Aerodynamic diameter was considered the significant aerosol parameter of concern and is reported in preference to any form of the physical (microscopic) diameter. Inertial impaction, the chief mode of particle collection by HEPA filters operating at rated capacity, is a function of aerodynamic behavior of the particles.[1] Activity median aerodynamic diameter (amad) is of primary concern since it defines the fractional amount of radioactivity in the size range where inhalation of plutonium presents a significant hazard. Amad is also a convenient unit because it is not affected by changes in isotopic ratio, particle shape, or particle density.

Particle size characteristics were determined by radiometric analysis of each of the nine stages of Andersen impactors (eight impaction stages plus backup membrane filter). Errors due to possible rebound of particles were minimized by covering the impaction surface with filter media. Impactor calibrations for 1.0 cfm and 2.75 cfm flow were based on experimental results by others.[2,3,4] Details of the sampling program were discussed in a previous

TABLE I

SUMMARY OF OPERATING CONDITIONS AT EACH SAMPLING LOCATION

Location	Operations	Isotope	Prefilter Efficiency	Relative Quantities Handled
00	R & D	Both	Unknown	Small
04	R & D	238	High[a]	Moderate
08	Fabrication	238	High[a]	Moderate
11	Recovery	239	Unknown[b]	Large
14	Fabrication	239	Unknown	Large

[a] Routine monitoring and replacement.

[b] Probably unreliable due to presence of high concentrations of corrosive acid vapors.

progress report.[5]

As a general assessment of the field sampling program, these comments are applicable:

1. The number of sampling locations (5) was probably adequate to indicate typical particle size characteristics of aerosols challenging exhaust HEPA filters from major operations of concern at AEC facilities.

2. The number of observations at each location (>30) was adequate for statistical confirmation of typical aerosol size characteristics. At one location (04) additional observations might have been desirable due to the wide variation in aerosol size characteristics.

3. Sampling results provide an adequate description of typical field aerosols to be simulated in the laboratory experimental program to define the performance of multiple HEPA filters.

4. Sampling apparatus provided adequate range and versatility to accomplish the desired measurements in a field situation with minimal supervision of sampling procedures.

B. Plutonium Aerosol Size Characteristics

The range in particle size characteristics can be developed in several ways, each of which may contain useful features, depending on the proposed application. Both methods employed for the field sampling data are based on effective cutoff diameter (ECD) of each impactor stage[6] which defines aerodynamic particle size collected with 50% efficiency on a given impactor stage. To describe a size distribution utilizing impactor data, it is convenient and sufficiently accurate[6] to assume step function cutoffs at the ECD (all particles collected on a stage as smaller than the ECD) and assign the ECD of a stage to the next downstream stage. The amount of radioactivity on a stage is the activity associated with particles in a size interval defined by the ECD of that stage and the next stage upstream.

Figures 1 through 5 express the mean percent activity ($\bar{W}_i$) as a function of aerodynamic diameter for each sampling location. This expresses the particle size distribution as the mean of all samples at that sampling site. Frequency expressed in units of $\bar{W}_i$ is derived by summing percent activity (W_i) collected on the i th stage of the impactor during all N observations and dividing this sum by N. This method yields equal weighting for all observations and prevents overpowering the overall size distribution by a limited number of samples which have exceptionally high activity concentrations.

The two research and development facilities, 00 (Fig. 1) and 04 (Fig. 2), exhibit similar particle size characteristics with the predominant size interval 1.1 to 2.1 μm for both locations. Both locations exhibited about 10% activity collected on the backup filter (0.43 μm and smaller). Location 04 is characterized by a rather uniform distribution of activity over the other size intervals. As will be noted again later, this smearing effect over a broad range of particle sizes is also reflected in a higher σ_g. This indication of higher dispersity could be expected from a facility doing a wide variety of processes.

Results at the two fabrication facilities, Locations 08 (Fig. 3) and 14 (Fig. 4), are comparable to each other, but distinctly different from data obtained from the research and development operations. The predominant size interval at these locations is 3.3 to 4.7 μm, the largest particle size consistently observed at any of the five locations. It should be noted, however, that significant quantities of small particles may also be present at these locations, as indicated by high $\bar{W}_i$ on the backup filter for Location 14 (>10%).

The remaining sampling location (11), a recovery facility, produced a consistently smaller aerosol, with the highest activity percentages collected on the backup filter (under 0.12 μm). To measure size characteristics of the smaller aerosol at

Location 11 (Fig. 5), the impactor sample flow rate was increased from 1.0 cfm to 2.75 cfm to provide a smaller ECD for each stage.[5] At this higher flow rate, the last stage of the impactor allowed only 23% of the mean percent activity to reach the back-up filter, which is assumed to be 100% efficient for particles smaller than 0.12 μm aerodynamic diameter.

Figs. 6 through 10 summarize the variations in particle size in terms of aerosol amad and σ_g. Aerosol size characteristics are most commonly expressed in terms of these parameters if the particle diameters are lognormally distributed. Since limits of lognormality could not be precisely defined mathematically, a semi-quantitation criterion for lognormality was established requiring each impactor data set to contain no data points which deviated more than 15% from the least squares best fit line. Though somewhat arbitrary, the method yields good agreement with visually adopted lines of best fit and data considered acceptable for graphical solution. Application of the 15% deviation criterion resulted in rejection of approximately 10 to 15% of the impactor data. The amad and σ_g of data passing this test for lognormality are presented as frequency histograms in Figs. 6 through 10. These histograms are updated versions of similar graphs in our previous reports.[5] These histograms show variations in size distribution in greater detail than in the mean percent activity method previously detailed in Figs. 1 through 5.

With possibly one exception, general characteristics indicated by this analysis show (as in the mean percent method) the similarity of size characteristics for similar operations. The r & d facilities (00, Fig. 6, and 04, Fig. 7) appear somewhat less alike than in the earlier comparison, primarily because lognormally distributed data from Location 04 were sparse and widely spread. This broad variation in both amad and σ_g accompanied by a relatively high incidence of non-lognormal distributions might be expected at a facility doing a broad spectrum of chemical and mechanical activities. Amad's from Location 00 fall within a fairly narrow size range, which was not expected.

The two fabrication plants (08, Fig. 8, and 14, Fig. 9) regularly emit a fairly large aerosol (predominant amad 3 to 4 μm) with a relatively low dispersity as defined by σ_g. The amad of the aerosol from Location 11, the recovery plant (Fig. 10), consistently was sub-micron with large variations in σ_g. Table II summarizes the general results of these data, detailing the high percentage of amad and σ_g observations falling within fairly narrow limits, as well as the mean value.

The mean percent activity results detailed in Figs. 1 through 5 can also be graphically analyzed using the log probability criteria to obtain a mean amad and σ_g. It may be argued that this additional averaging step eliminates the detailed features of the original size distributions. However, this treatment condenses the data presented in Figures 1 through 10 into five sets of amad's and σ_g's, as presented in Table III. These mean values were obtained by least squares fit of $\overline{W}_i$ with a weighting proportional to sample size. Maximum deviation of any data point from the line of best fit was 10.2%, indicating these data meet the 15% deviation limit established for lognormally distributed data.

Not every value of amad and σ_g in Table III agrees closely with the corresponding all-sample mean from Table II or with the mean value intuitively selected from Figures 6 through 10. Notably different are the amad's of Location 04 (2.3 vs 2.9 μm) and Location 11 (0.34 vs 0.5 μm); and the σ_g at Location 11 (5.4 vs 3.9). Since the results were obtained by different methods (albeit using the same raw data), some differences could be expected. The magnitude of these differences are not large enough to seriously alter how

TABLE II

SUMMARY OF SIZE CHARACTERISTICS OF LOGNORMALLY DISTRIBUTED PLUTONIUM AEROSOLS

Activity median aerodynamic diameter (amad)

At Location___,	___%	of___observations	fall in the range ___ μm to___μm	All sample mean (μm)
00	86	77	1.0 to 3.0	1.9
04	62	26	1.0 to 4.0	2.9
08	92	48	3.0 to 5.0	4.1
11	89	18	0.1 to 1.0	0.5
14	84	49	2.0 to 4.0	2.6

Geometric standard deviation (σ_g)

At Location___,	___%	of___observations	fall in the range ___to___	All sample mean
00	86	77	1.5 to 3.0	2.1
04	81	26	1.5 to 3.5	3.0
08	92	48	1.5 to 2.5	1.7
11	67	18	1.5 to 4.0	3.9
14	71	49	1.5 to 3.0	2.9

these size characteristics will be used to define typical plutonium source term size characteristics and provide the basis for the laboratory experimental program to evaluate the efficiency of multiple HEPA filters.

C. Activity Concentrations

As described in the earlier report,[5] activity concentration measurements by membrane filters were obtained simultaneously with impactor samples. These results are presented as bargraphs of mean and maximum concentration in Figure 11. The highest activity concentrations were noted in one of several ducts entering three-stage HEPA plenums at Locations 11 and 14. These locations consistently produced activity concentrations of ^{239}Pu in the range 10^6 to 10^7 dpm/m^3. High activity levels combined with the small particle size makes Location 11 the most difficult air cleaning problem of the five sampling locations.

TABLE III

MEAN Pu AEROSOL SIZE CHARACTERISTICS BASED ON MEAN PERCENT ACTIVITY

Location	Type	Isotope	amad (μm)	σ_g
00	r & d	Both	1.8	2.2
04	r & d	238	2.3	3.5
08	fabri.	238	4.0	1.7
11	recov.	239	0.34	5.4
14	fabri.	239	2.7	2.4

D. Effect of Prefiltration

Better prefiltration at the glovebox in some locations was expected to have a noticeable effect on particle size, primarily by collection of large particles at the glovebox. Referring to the estimated quality of prefiltration noted in Table I, Location 04 and 08 should reflect lower mean percent activity in the larger size intervals (>7.0μm) if the higher quality of prefiltration truly has a marked effect on particle size. As seen in Figures 2 and 3, the mean percent activity is over 5% for particle sizes over 7.0 μm, showing this not to be the case.

The data were also examined for days at Location 04 and 08 when activity concentrations were suddenly quite high, indicating possible breakthrough of prefilters. Comparing size characteristics of samples with high activity and samples with normally lower activity did not show larger size associated with higher activity. A possible explanation, rather than filter failure, might be sharply increased activity in the glovebox line.

E. Experimental Apparatus & Techniques

Details of operation of the Andersen impactor and its calibration at 1.0 and 2.75 cfm and an evaluation of particle rebound were previously detailed.[5] Several additional equipment-related investigations conducted since the last report are described below.

1. Spectrum Analysis at Location 00

Alpha spectrum analysis of impactor samples from Location 00 (the only location handling both ^{238}Pu and ^{239}Pu) detected wide day-to-day variation in isotopic ratio.[5] Interest in separating the composite size distribution into distributions for each isotope prompted preparations for spectrum analysis of all impactor samples. This required a silicon surface barrier detector, vacuum chamber, and multi-channel analyzer. The analyzer has been borrowed and the other two items are on order. The surface barrier detector will be a ruggedized cleanable model which will allow analysis of the relatively high-level samples collected.

2. Resolving Time Measurements

Many of the samples counted on the gas flow proportional counter have exceeded 10^5 cpm. Since some counting systems undergo a change in resolving time with increasing count rate, resolving time measurements were made up to 1.2×10^6 cpm. Resolving time did not vary significantly from 10μsec up to that count rate. All resolving time corrections have been based on 10μsec resolving time.

3. Impactor Rebound

Additional experimental data comparing potential errors due to rebound from impactor surfaces coated with vinyl MF and Type E glass fiber filter indicated the possibility of a slight rebound effect from vinyl MF. In general, however, this comparison and earlier investigations with duplicate impactors failed to detect any significant rebound problem. Possible explanations for better adherence by Pu aerosol than by spherical polystyrene latex aerosol noted in previous studies of rebound[7] are (1) lower overall mass deposit on impaction surfaces, (2) lower particle resilience with less likelihood of elastic collision, and (3) irregular shape providing greater contact area.

III. EXPERIMENTAL TEST PROGRAM

A. Test System

Preliminary tests using the experimental test system to evaluate the performance of multiple HEPA filters against plutonium aerosols indicated difficulties due to alpha radiation damage to the plastic nebulizers. The damage resulted in large drops of generator solution passing into the test system duct leading to HEPA filter #1, creating a relatively large pressure drop across the HEPA filter, and an error in monitoring system air flow. Modifications to the aerosol generating system, and changing HEPA filter #1 after every two runs have eliminated this problem. Due to the

effects of radiation on the plastic ReTec nebulizers, these are repaired or changed every 3 to 4 runs.

The test system is shown in Figures 12 and 13. Figure 12 details the first module composed of a glovebox housing the aerosol generators, (1); sampler #1, (2); and HEPA filter #1, (3). Each test HEPA filter has a design flow rate of 25 cfm, and its construction and filtration velocity is identical to the typical 1000 cfm units used in most air cleaning systems. The only difference is that the 1000 cfm units are generally open faced, while the test filter is designed for in line installation with 2-inch pipe nipples at each end. Figure 13 shows the second module and its major components which consists of sampler #2 (4) immediately upstream of HEPA filter #2 (5); sampler #3 (6) immediately upstream of HEPA filter #3 (7); sampler #4 (8) downstream of HEPA filter #3; and a vacuum pump (9). Samplers #1, #2 and #3 are dual samplers simultaneously collecting a gross membrane filter sample for aerosol concentration, and an Andersen impactor sample for measuring aerosol aerodynamic size characteristics. The gross filter measurements determine overall HEPA filter efficiencies, while impactor data are used to calculate HEPA filter efficiency as a function of plutonium aerosol aerodynamic size. Sampler #4 consists of nine 2" open face glass fiber filters and is designed to filter all the exhaust air. This was required because of the very low levels of activity existing at this point which precludes impactor measurements to define aerosol size characteristics downstream of the third HEPA filter.

Initially Nuclepore filters with 0.8μm pore size were used as the gross samplers and 9th stage (backup filter) to the Andersen impactor. To check the collection efficiency of the Nuclepore filters, Millipore AA filters were located downstream while sampling an 0.8μm (amad) $^{238}PuO_2$ aerosol. Approximately 10% of the activity passed through the Nuclepore filter and was collected on the Millipore AA filter. On the basis of these data the sampling system was modified to use Millipore AA filters. Millipore AA filters are readily soluble in acetone, making it possible to plate small aliquots of the sample on aluminum plates for counting.

Sampling times are normally one minute during each 10 minutes at gross filter sampler #1. The Andersen impactor at this location samples for one minute near the beginning and near the end of each run which lasts 60 to 90 minutes. Sampler #2 (gross filter and Andersen impactor) collects a 30 minute sample, while samplers #3 and #4 operate during the entire duration of the run.

In the initial experimental runs, contamination was observed on the 3rd and 4th samples. Several dummy runs (no aerosol generated) were made to check the source of this contamination which was found to have a half-life of 10.8 hours which corresponds to thoron daughter decay products. Decay times of 4 to 7 days are now being allowed prior to counting to eliminate this problem. Recounts were completed on these initial samples which had not been discarded. All samples from samplers #3 and #4 are now being counted a minimum of one thousand minutes to provide better statistics for these samples which contain extremely low counts. Typically sampler #4 (downstream from HEPA #3) indicates approximately 0.5 cpm. It is necessary to minimize the error associated with this measurement to accurately define filter efficiency for HEPA #3.

B. $^{238}PuO_2$ Suspensions

To approximate plutonium aerosols with 0.1 to 5 μm amad's measured under field conditions, $^{238}PuO_2$ powders were dry ball milled for various time intervals and suspended in water to a concentration of 2.5 mg/ml. Ultrasonic agitation of the suspension broke up agglomerates, and addition of anionic surfactant kept the suspensions well dispersed. Selective ball

milling provided some control of size characteristics over the range of interest with some limitations at either end (<0.5 μm and >2.5 μm amad). Table IV summarizes the grinding procedures tested, and the resulting aerosol produced using the ReTec nebulizer and the test procedures previously detailed. It is obvious from Table IV that we cannot produce an aerosol with an amad much smaller than 0.7 μm using these techniques. However, even for 0.7 μm aerosols, a significant fraction of the particles are as small as 0.4 μm, and Andersen impactor sampling data provide information regarding the performance of HEPA filters for aerosols of this size. Because of the importance of developing data for test aerosols with amad's as small as 0.1 μm, a new centrifugal mill has been installed to permit wet milling with ethanol. By milling wet, re-agglomeration will be minimized and with the additional mill energy input, $^{238}PuO_2$ with amad's as small as 0.1 μm should be available.

C. Results

Overall HEPA filter efficiencies determined by gross filter data for each filtration stage are shown in Table V. HEPA filter stages are numbered 1-3 with stage 0 representing the aerosol concentration and size characteristics upstream of HEPA filter #1. Aerosol size characteristics in terms of amad and σ_g generally decrease at succeeding stages. Activity concentrations upstream of HEPA #1 ranged between 10^{10} to 10^{12} dpm/m^3. As expected filter efficiency is highest for the first stage, but the measured HEPA filter efficiencies remain well within the present minimum AEC performance guidelines for each stage;[8] i.e., 99.95% for first stages and 99.8% for succeeding stages. These data are summarized in Table VI.

Minimum efficiency noted for the third stage is slightly below the 99.8% guideline. This is due to statistical problems encountered with count rates below 1 cpm downstream of the third HEPA filter, and counting problems due to gaseous contaminants from radon-thoron daughters. Contamination probably accounts for the two tests indicating an efficiency less than 99.8%. Greater confidence in third HEPA stage efficiencies was obtained using longer run times, with four times the aerosol concentration (10 mg/ml), and longer counting times. The modifications in the test procedure have resulted in consistently higher efficiencies for the third HEPA filter. These data indicate that a third HEPA filter in series will satisfy existing AEC guidelines.[8]

Table VII shows HEPA filter efficiencies as a function of aerosol aerodynamic size. The first column denotes the impactor stages for an 8-stage impactor plus a backup filter (MF #2). The next column gives the impactor particle collection interval for each stage in μm. Mean efficiencies of HEPA filter #1 are well above the minimum criteria with the minimum efficiency occurring against particles collected on Stage #7 of the impactor or 0.43 to 0.65 μm size range. HEPA filter #2 also shows mean efficiencies above the required 99.8% with the minimum occurring against particles smaller than 0.43 μm represented by the impactor backup filter. Overall HEPA filter efficiencies based on total impactor activity were 99.998% for both cases. Although impactor data downstream of HEPA #3 are not available, the efficiencies reported for HEPA #3 are essentially against particles <1.1 μm and of that, particles <.43 μm account for about 40% of total activity.

IV. EFFECT OF ALPHA RADIATION ON HEPA FILTER MEDIA

An investigation of the effect of alpha radiation on glass fiber media in HEPA filters has been initiated. Prolonged exposure of glass fibers to particles of high specific activity may cause embrittlement, spallation, or some other damage mode which results in parting of fibers.

The approximate dose rate at the surface of a $^{238}PuO_2$ sphere was estimated as

follows:

(1) If the sphere is large compared with the alpha range (about 11 μm in PuO_2), then the dose rate inside the sphere is given by the rate of energy release per gram.

(2) Specific activity

$= 15.5 \text{ Ci/g} \times 2.2 \times 10^{12} \text{ α/min/Ci}$

$= 3.42 \times 10^{13} \text{ α/min/g}$

(3) Energy released

$= 5.5 \text{ MEV/α} \times 3.42 \times 10^{13} \text{ α/min/g}$

$= 1.88 \times 10^{14} \text{ MEV/min/g}$

(4) Energy Conversion:

1 rad = 100 erg/g

1 MEV = 1.6×10^{-6} erg

(5) Dose rate

$$= \frac{(1.88 \times 10^{14})\ (1.6 \times 10^{-6})}{100}$$

$= 3.0 \times 10^{6}$ rad/min

Although dE/dx is not as high in the adjacent fiber as in the particle, the distances are small compared to the alpha range and the fiber will probably absorb energy at a fairly high rate, say within one order of magnitude of absorption in the particle. For the purposes of further investigation of alpha radiation effects, an absorbed dose rate of 10^6 rad/min and an exposure time of one year are being specified.

Further information is being sought to relate the dose rate to material damage. Comparison of alpha absorbed dose and gamma exposures previously reported[9, 10] may allow indirect evaluation of potential alpha damage, if the gamma exposures can be related to calculated alpha absorbed dose. Cheever detected negligible HEPA efficiency

TABLE IV

Batch	Action	Size (amad)	σ_g
BND 177 AR	Original material from Savannah River used as received after sonication and addition of surfactant.	1.2μm	2.4
BND 177 100	Original material ball milled 100 hours, sonication, surfactant.	0.87μm	2.6
BND 177 360	Original material ball milled 360 hours, sonication, surfactant.	0.86μm	2.3
BND 177 MIX	Mixture of residuals from previous batches.	0.8μm	2.7
BND 176 DS 000	Special material supposedly much larger (∿14μm). Sonicated, surfactant added. Apparently larger particles either not aerosolized or not getting to samplers.	0.7μm	3.8
BND 177 720	Original material ball milled 720 hours, sonication, surfactant.	0.8μm	2.1
BND 177 1440	Original material ball milled 1440 hours, sonication, surfactant.	0.8μm	2.1

loss following ^{60}Co gamma irradiation at 10^7 to 10^9 R. If alpha absorbed dose is in a range where material damage is suspected, a laboratory study might be initiated using HEPA filters already heavily deposited with ^{238}Pu in the present multiple HEPA study.

V. FUTURE WORK

(1) Development of plutonium test aerosols with amad's as small as 0.1 μm.

(2) Testing of multiple HEPA filter systems using higher activity concentrations to provide better count statistics for the third HEPA filter, and smaller test aerosol to simulate the aerosols at Location 11.

(3) Evaluation of the need for additional field sampling sites and testing of more than 3 filters in series.

(4) Investigation of the effect of alpha radiation damage on HEPA filter media will continue.

(5) Literature review of the performance of sand and deep bed filters, and their applicability to air cleaning for plutonium facilities.

(6) Separation of particle size distributions by isotope will be performed by spectrum analysis at Location 00.

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TABLE V

OVERALL HEPA FILTER EFFICIENCY

Run	HEPA Filter Stage	Plutonium Aerosol amad (μm)	Plutonium Aerosol σ_g	Activity Concentrations dpm/m³	Plutonium HEPA Filter Efficiency
P2-1	0	0.7	2.26	1.6775×10^{11}	
	1	0.6	1.50	5.7595×10^{5}	99.99966
	2	0.7	1.8	14.30	99.99752
	3	--	--	7.228×10^{-2}	99.49495 *
P2-3	0	1.3	2.94	1.274×10^{11}	
	1	0.59	1.6	3.101×10^{5}	99.99976
	2	0.57	1.84	5.179	99.99833
	3	--	--	C O N T A M I N A T E D	

* probable contamination from radon-thoron daughters

TABLE V (continued)

OVERALL HEPA FILTER EFFICIENCY

Run	HEPA Filter Stage	Plutonium Aerosol amad (μm)	Plutonium Aerosol σ_g	Activity Concentrations dpm/m³	Plutonium HEPA Filter Efficiency
P2-4	0	1.3	2.7	1.1132×10^{11}	
	1	0.45	2.04	3.3270×10^{5}	99.99970
	2	0.48	2.54	2.424	99.99927
	3	--	--	C O N T A M I N A T E D	
P2-5	0	0.65	2.2	3.024×10^{11}	
	1	0.64	1.6	5.5329×10^{6}	99.99817
	2	0.55	1.4	1.006×10^{2}	99.99818
	3			C O N T A M I N A T E D	
P2-6	0	0.75	2.7	2.6168×10^{11}	
	1	0.59	1.6	3.7528×10^{6}	99.99857
	2	0.51	1.5	83.64	99.99777
	3	--	--	4.604×10^{-2}	99.94496
P2-7	0	1.61	2.70	1.0107×10^{11}	
	1	0.64	1.70	4.5096×10^{5}	99.99955
	2	0.43	1.31	4.615	99.99898
	3			1.003×10^{-2}	99.78267
P2-8	0	0.79	2.51	7.4584×10^{10}	
	1	0.67	1.74	1.4116×10^{6}	99.99811
	2	0.56	1.47	1.4994×10^{1}	99.99894
	3			7.2400×10^{-2}	99.51714 *
P2-9	0	0.84	2.07	3.1028×10^{11}	
	1	0.45	1.93	4.8581×10^{6}	99.99843
	2	0.42	1.66	1.2391×10^{2}	99.99745
	3			3.4600×10^{-2}	99.97208
P2-10	0	0.80	2.09	4.4013×10^{11}	
	1	0.52	1.67	6.2614×10^{6}	99.99858
	2	0.36	1.79	1.2902×10^{2}	99.99794
	3			2.4100×10^{-2}	99.98132
P3-1	0	0.71	2.12	$.9510 \times 10^{12}$	
	1	0.66	1.58	2.0030×10^{7}	99.99789
	2	0.42	1.79	5.2739×10^{2}	99.99737
	3			5.7856×10^{-2}	99.98903

* probable contamination from radon-thoron daughters

TABLE V (continued)

OVERALL HEPA FILTER EFFICIENCY

Run	HEPA Filter Stage	Plutonium Aerosol amad (μm)	Plutonium Aerosol σ_g	Activity Concentrations dpm/m^3	Plutonium HEPA Filter Efficiency
P3-2	0	0.77	2.18	1.2840×10^{12}	
	1	0.61	1.65	2.2651×10^{7}	99.99824
	2	0.60	1.40	6.7250×10^{2}	99.99703
	3			4.6788×10^{-2}	99.99291
P3-3	0	1.45	2.79	4.3785×10^{11}	
	1	0.82	2.00	1.1916×10^{7}	99.99728
	2	0.50	1.60	1.5438×10^{2}	99.99870
	3			5.6183×10^{-2}	99.96361
P3-4	0	0.78	2.55	4.3887×10^{11}	
	1	0.57	1.75	5.0336×10^{6}	99.99885
	2	0.50	1.65	8.2236×10^{1}	99.99836
	3			2.2300×10^{-2}	99.97288
P3-5	0	0.80	2.54	1.3696×10^{12}	
	1	0.58	1.69	1.2991×10^{6}	99.99991
	2	0.49	1.49	1.9823×10^{1}	99.99847
	3			1.9314×10^{-2}	99.90257

TABLE VI

OVERALL HEPA FILTER EFFICIENCY

Range of Size amad (μm)	Range of Size σ_g	HEPA Filter Stage	#Runs	Efficiency Range (%) Mean	Min	Max
0.70 - 2.1	2.07 - 3.0	1	14	99.99876	99.99728	99.99991
0.45 - 0.82	1.5 - 2.04	2	14	99.99817	99.99703	99.99927
0.37 - 0.70	1.27 - 1.84	3	11	99.86492	99.49495	99.99291

TABLE VII

HEPA FILTER EFFICIENCY AS A FUNCTION OF AEROSOL SIZE

Sampling Impactor Stage Number	Aerodynamic Diameter Range μm	Mean Efficiency(%) HEPA #1	HEPA #2
0	>11	99.99976	99.99974
1	7.0 - 11	99.99981	99.99688
2	4.7 - 7.0	99.99988	99.99912
3	3.3 - 4.7	99.99985	99.99819
4	2.1 - 3.3	99.99971	99.99970
5	1.1 - 2.1	99.99901	99.99978
6	0.65 - 1.1	99.99758	99.99851
7	0.43 - 0.65	99.99702	99.99812
MF#2	<0.43	99.99827	99.99728
OVERALL	-	99.99861	99.99830

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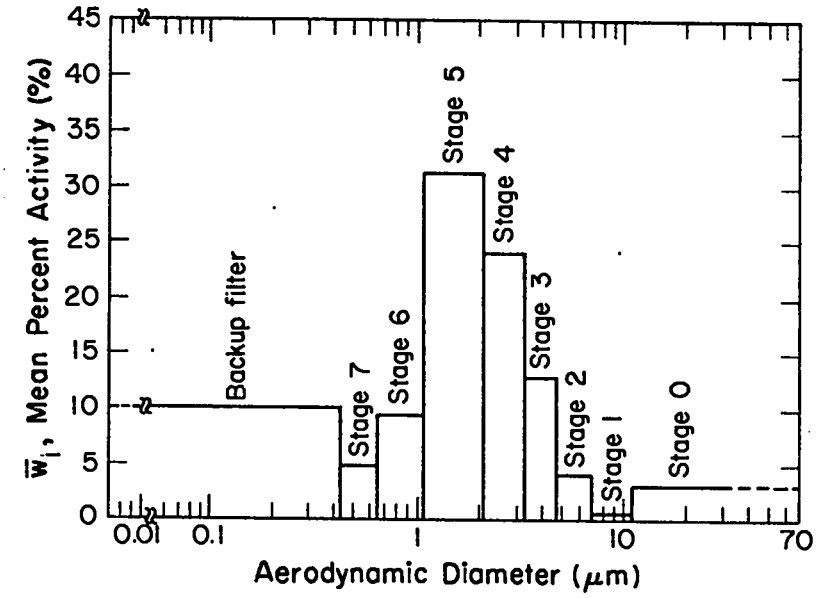

Figure 1: ^{238}Pu and ^{239}Pu Aerosol Size Characteristics by Radiometric Analysis of All Impactor Samples at Location 00, A Research and Development Facility.

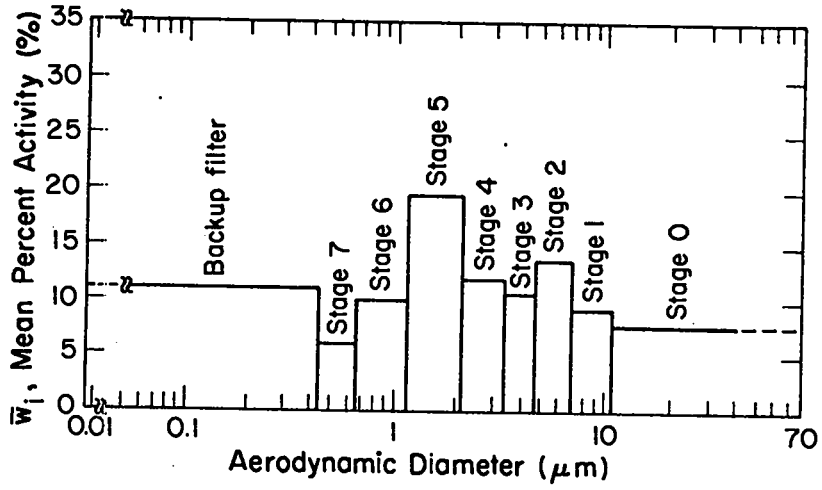

Figure 2: ^{238}Pu Aerosol Size Characteristics by Radiometric Analysis Of All Impactor Samples At Location 04, A Research and Development Facility.

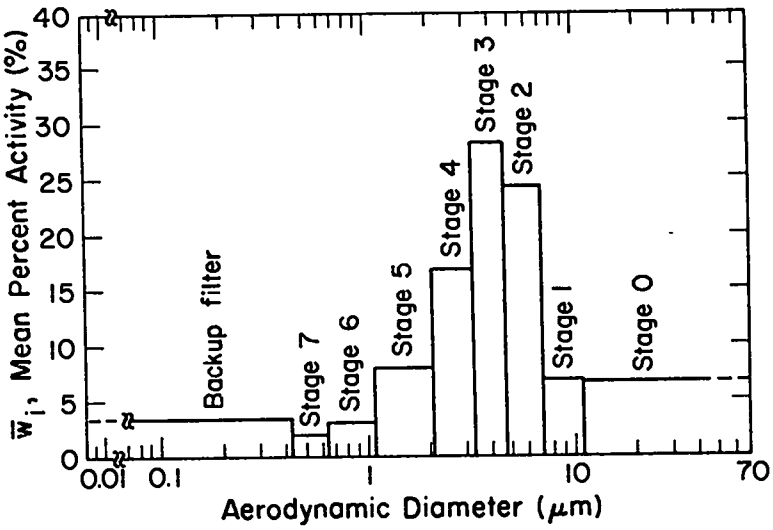

Figure 3: ^{238}Pu Aerosol Size Characteristics By Radiometric Analysis of All Impactor Samples at Location 08, A Fabrication Facility.

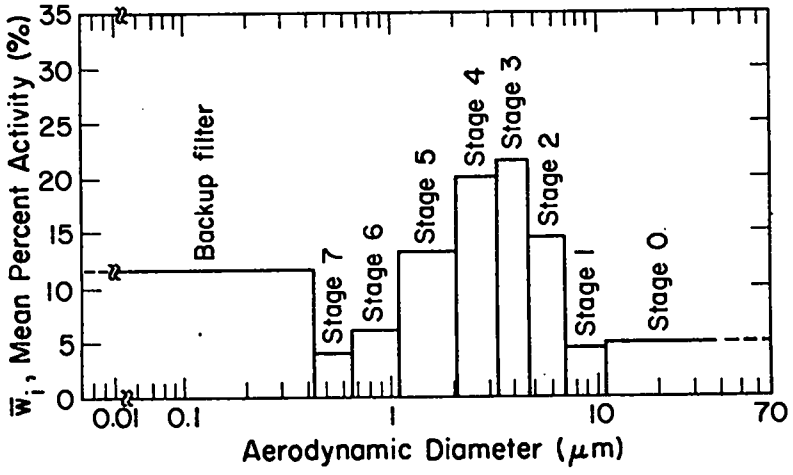

Figure 4: ^{239}Pu Aerosol Size Characteristics By Radiometric Analysis of All Impactor Samples at Location 14, A Fabricating Facility.

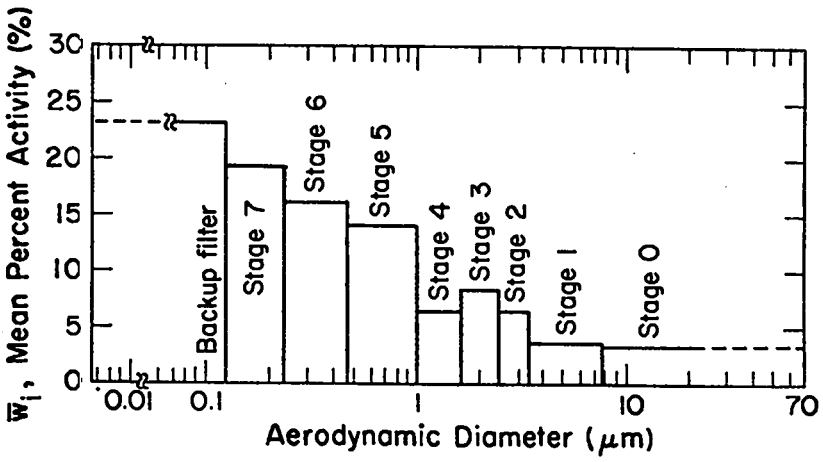

Figure 5: ^{239}Pu Aerosol Size Characteristics By Radiometric Analysis of All Impactor Samples at Location 11, A Chemical Recovery Facility. Impactor operated at 2.75 cfm.

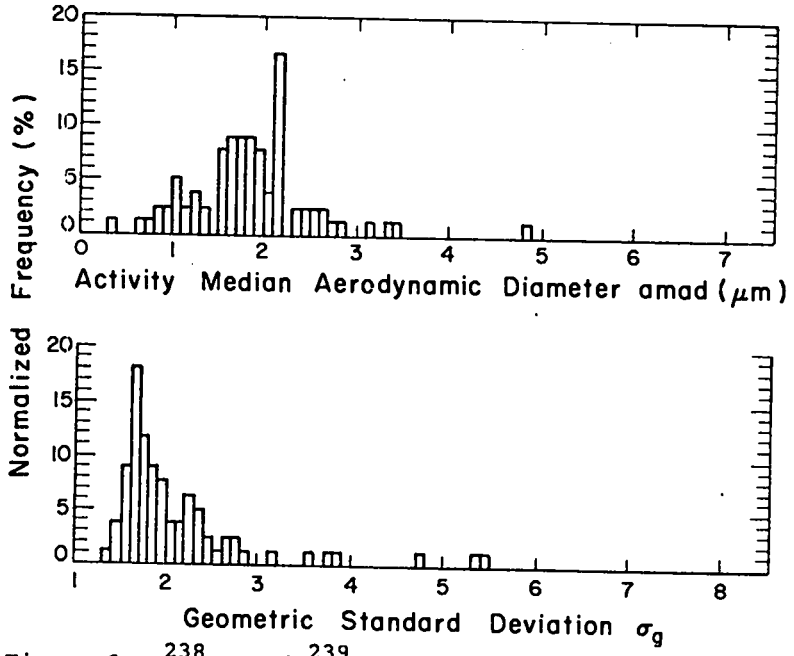

Figure 6: ^{238}Pu and ^{239}Pu Aerosol Size Characteristics of Lognormally Distributed Impactor Data From Location 00, A Research and Development Facility.

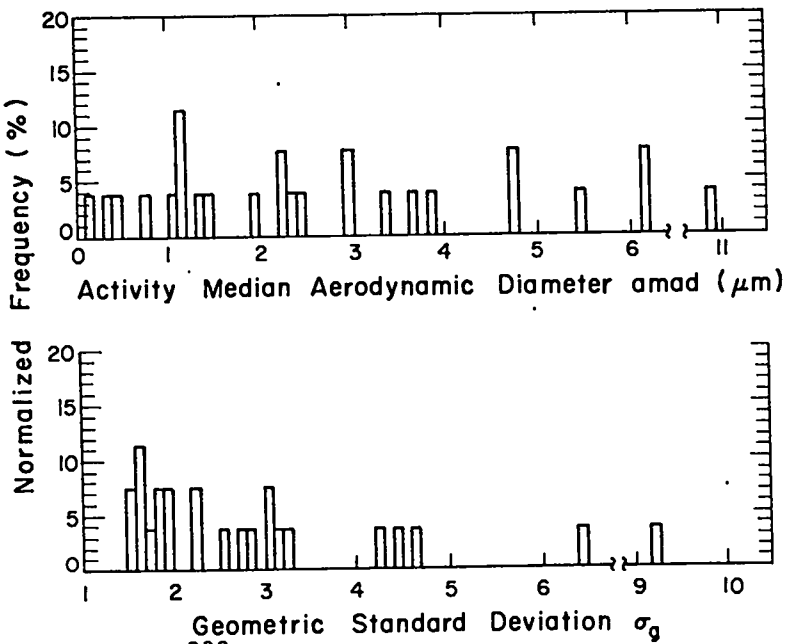

Figure 7: ^{238}Pu Aerosol Size Characteristics of Lognormally Distributed Impactor Data From Location 04, A Research and Development Facility.

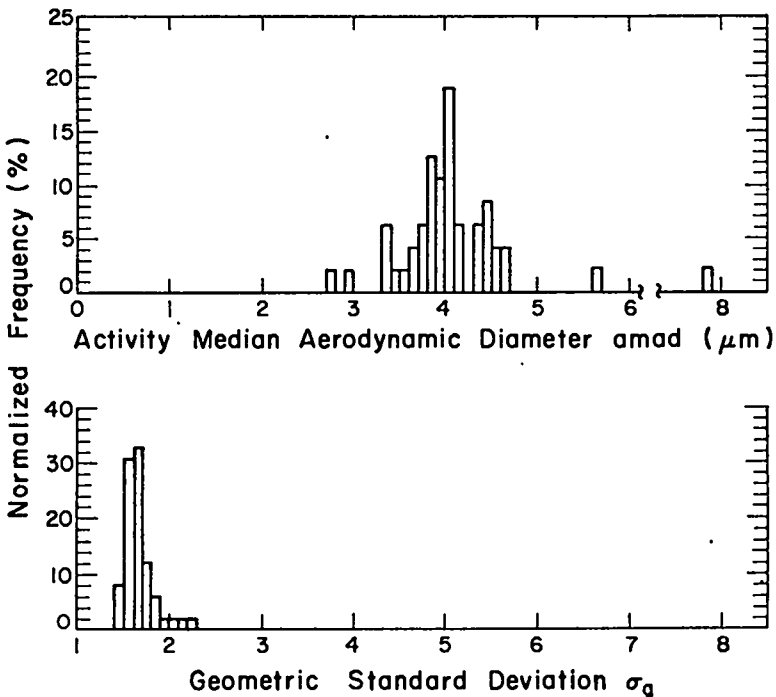

Figure 8: ^{238}Pu Aerosol Size Characteristics of Lognormally Distributed Impactor Data From Location 08, A Fabricating Facility.

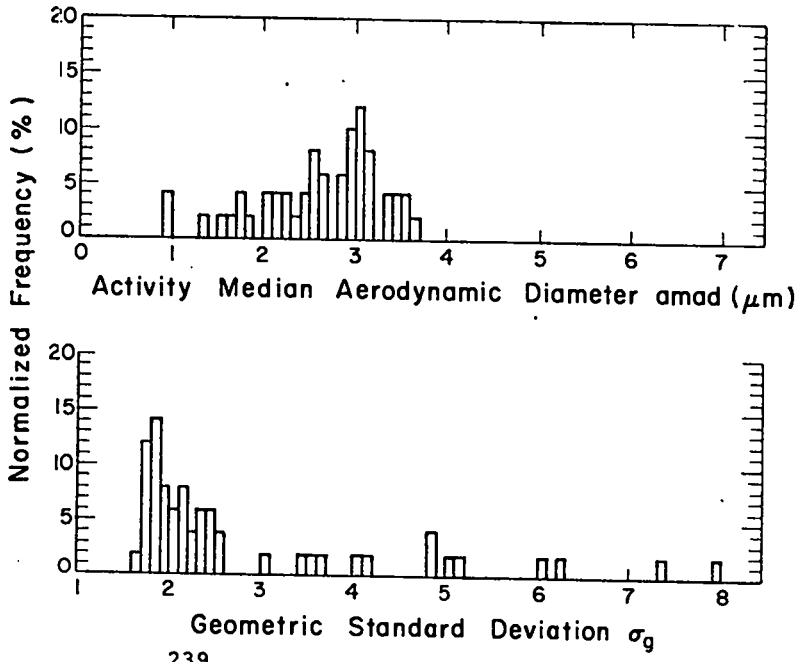

Figure 9: ^{239}Pu Aerosol Size Characteristics of Lognormally Distributed Impactor Data From Location 14, A Fabricating Facility.

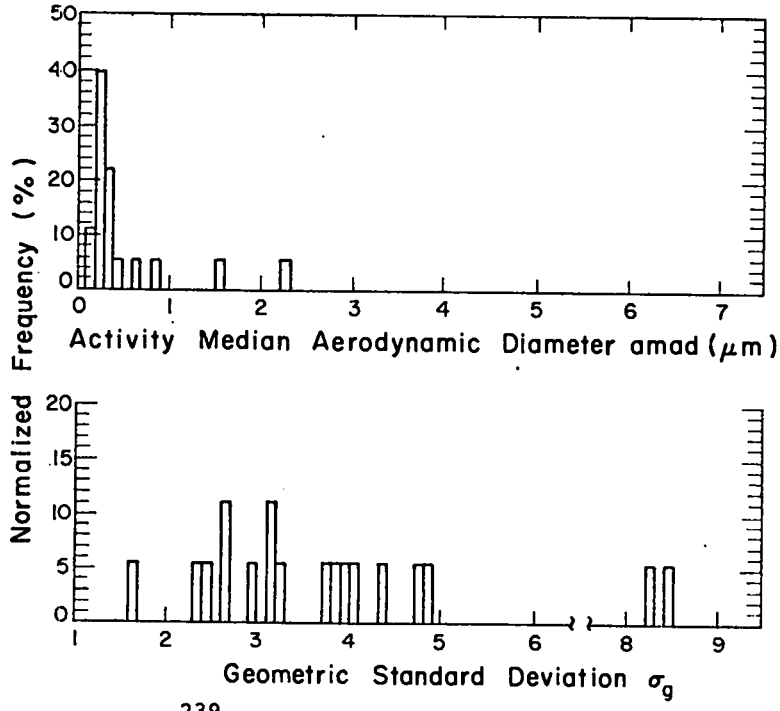

Figure 10: ^{239}Pu Aerosol Size Characteristics of Lognormally Distributed Impactor Data From Location 11, A Recovery Facility.

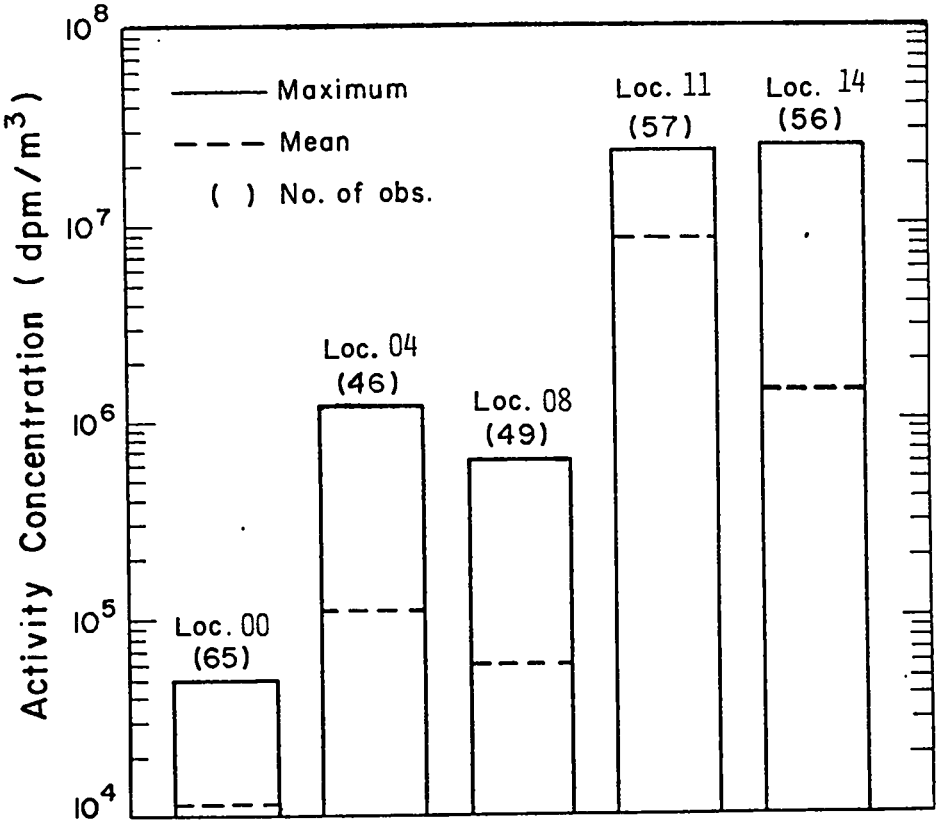

Figure 11: Maximum and Mean Alpha Activity Concentration at Five Pu Processing Facilities.

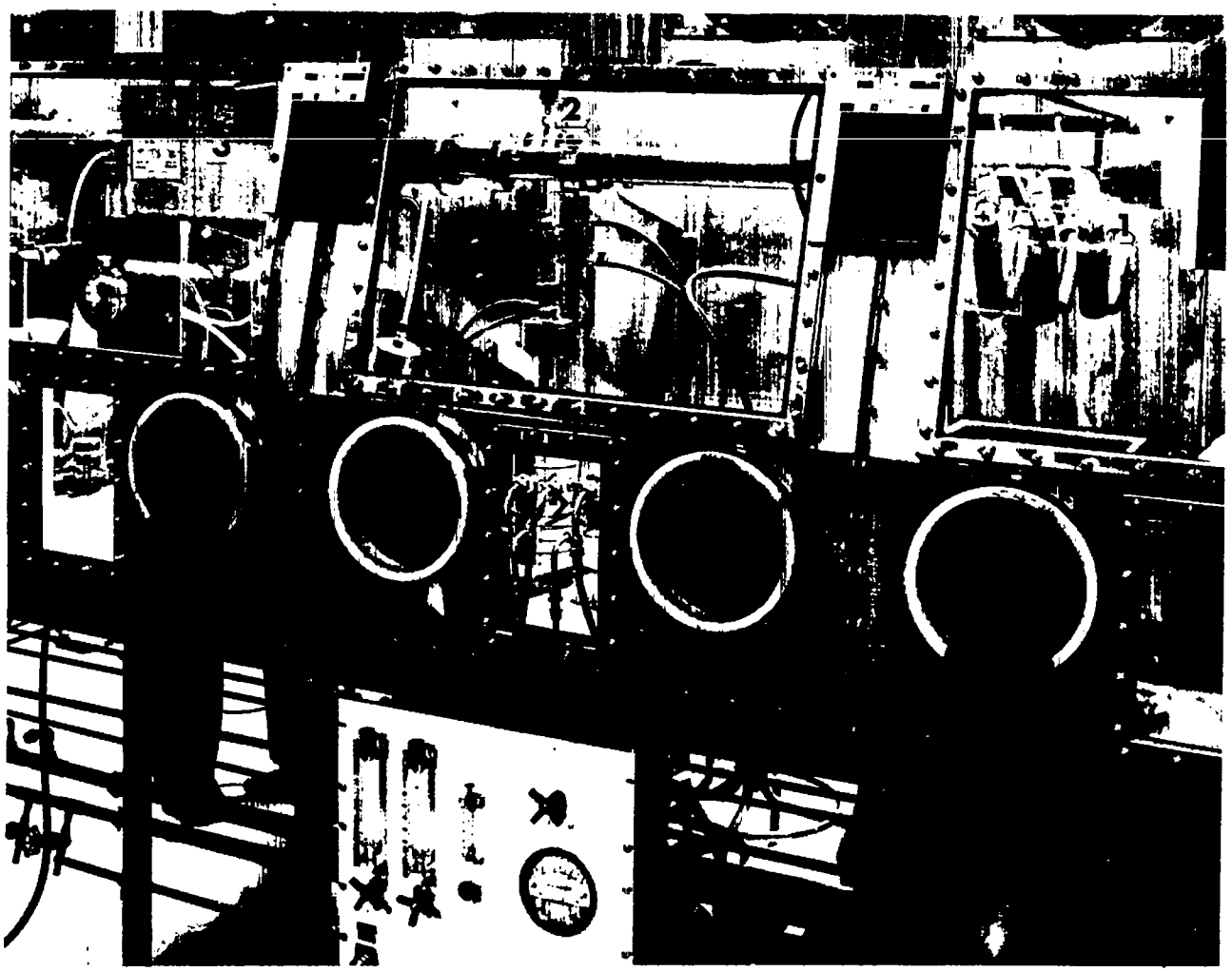

FIGURE 12: GLOVEBOX MODULE.

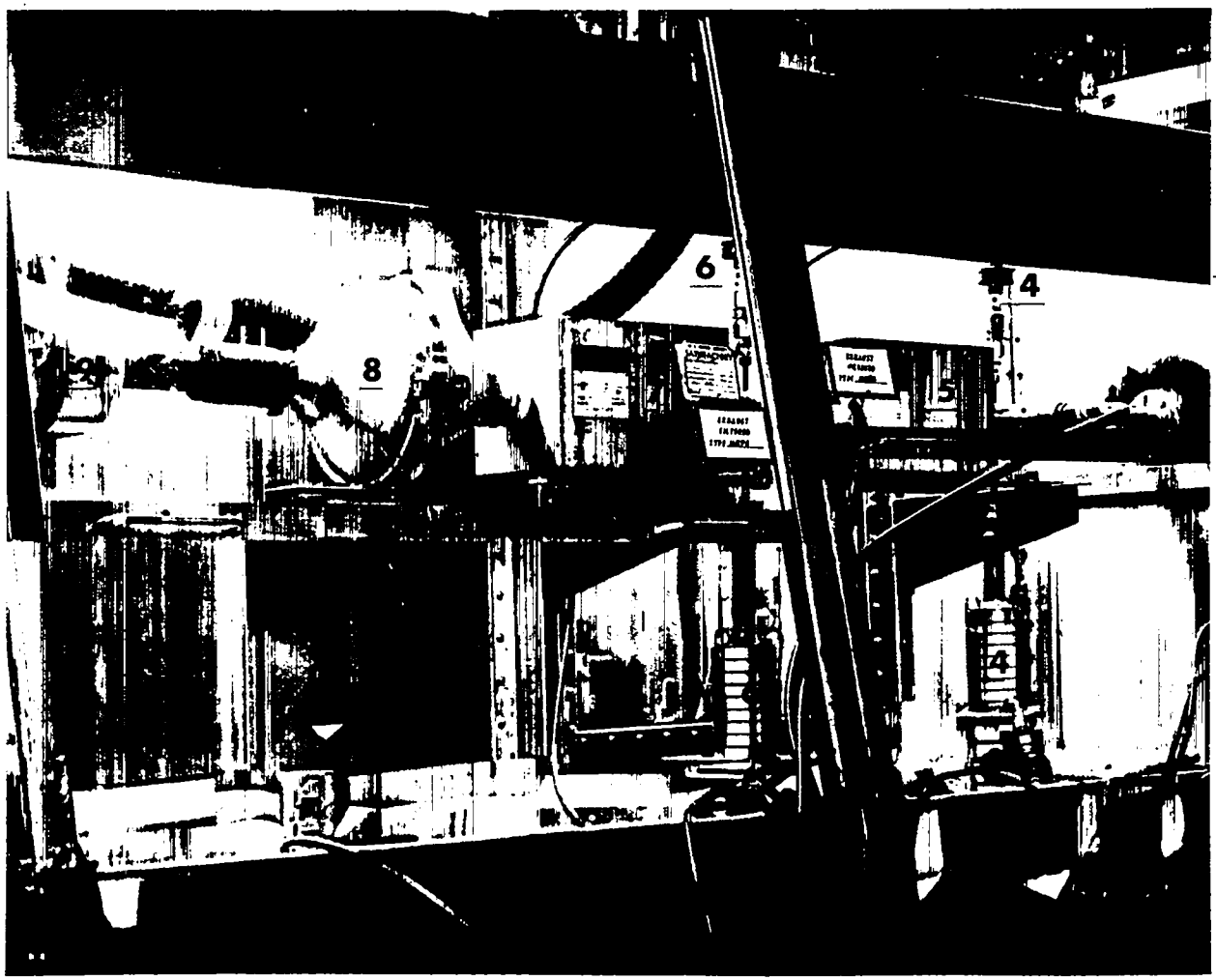

FIGURE 13: HOOD MODULE.