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A PROGRESS REPORT

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Performance of Multiple HEPA Filters Against Plutonium Aerosols

for Period March 1 through June 30, 1972

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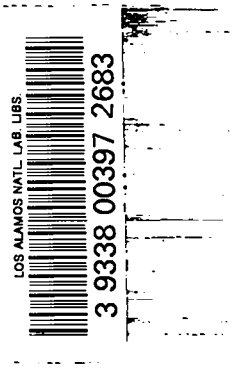

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Printed in the United St[illegible]. Available from
National Technical In[illegible] Service
U. S. Department of Co[illegible]rce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy $3.00; Microfiche $0.95

LA-5012-PR
A Progress Report
SPECIAL DISTRIBUTION
ISSUED: July 1972

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Performance of Multiple HEPA Filters Against Plutonium Aerosols

for Period March 1 through June 30, 1972

by

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INTRODUCTION

Increased concern regarding the potential release of radioactive particulates to the atmosphere has resulted in new stringent air cleaning requirements for facilities handling plutonium. This has included proposals to require decontamination factors of 10^9 to 10^{12}, which presently can be attained by use of multiple High Efficiency Particulate Aerosol (HEPA) filters, which singly can provide decontamination factors of 10^3 to 10^4. While extensive test data is available to substantiate this level of performance for individual HEPA filters, and filtration theory predicts multiple filter installations will provide the overall protection required, quantitative substantive data is not available. This is due to the limited sensitivity of tests using dioctyl phthalate (DOP) as a tracer aerosol. There is also concern that filter performance against laboratory test aerosols, such as DOP, is not fully representative of performance against plutonium aerosols to be removed by the air cleaner.

Because of these new air cleaning requirements, interest in realism, and the costs associated with unnecessary filter banks, a field and laboratory experimental program was initiated at the LASL. The field sampling program was designed to measure size characteristics and activity concentrations (source term) of plutonium aerosols generated by typical operations at several AEC facilities handling significant quantities of plutonium. Definition of these aerosols would provide a basis for generating similar aerosols in the laboratory to study the effectiveness of multiple stages of HEPA filtration.

This first progress report only covers four months since initiation of the program, therefore data reported is of a preliminary and limited nature, and complete statistical analysis is not yet available. However, general trends are indicated. To date, analytical procedures generally consist of gross alpha counting, with the assumption that Pu is the only particulate present. Alpha spectroscopy will be employed to verify this assumption.

I. SUMMARY

The field sampling program for activity concentrations (using membrane filters), and particle size analysis (using Andersen impactors), has been in progress since mid-March, 1972. Five sampling locations have been selected at the following three AEC facilities: Los Alamos Scientific Laboratory, Mound Laboratory, and Rocky Flats Plant. These five sampling locations will provide data to characterize the source aerosol resulting from 1) chemical production; 2) research and development activities; and 3) machining and fabrication. These sampling sites will include operations involving ^{238}Pu; ^{239}Pu; and a mixture of the two isotopes.

Initial data shows Pu concentrations ranging from 286 dpm/m^3 to 16.5 x 10^6 dpm/m^3. Aerodynamic size characteristics based on Andersen impactor sampling and analysis by a computer program which provides a log normal minimum chi-squares fit of these data indicate the following for two of the sampling locations:

aerodynamic activity median diameter (aamd) = .52 μm to 3.5 μm

geometric standard deviation (σ_g) = 1.5 to 6.6

Mean values of aamd and σ_g for each location are 1.70 μm and 2.33; 2.83 μm and 3.03, respectively. A third sampling location has an aamd too small to be adequately analyzed by Andersen impactors. Approximately 60% of the particulate activity at this sampling location is smaller than the smallest cutoff diameter of the Andersen impactor.

Potential errors due to particle rebound from impactor plates is being evaluated using twin impactor streams with one sampler having the impactor surfaces coated with a membrane filter. Preliminary results indicate that particle rebound is not a serious problem. Initial samples for electron microscopy have not been adequate for particle sizing due to low particle concentration and the background from nonradioactive particles. Electron microscopy has indicated minimal agglomeration of particles collected from one exhaust air stream.

A polydisperse DOP test system has been constructed to a) quality control test all HEPA filters to be evaluated using plutonium aerosols, and b) estimate HEPA filter efficiency for the second of two filters in series. The latter involves using a 25 cfm test system employing two HEPA filters in series, and generating a highly concentrated DOP aerosol upstream of the first filter to provide an aerosol concentration downstream of the first of about 0.4 to 1.0%. This then constitutes the upstream challange aerosol for the second HEPA filter. Using the minimum detectable limit for the LASL light scattering photometer, efficiency for this second filter is >99.9%.

Several methods of generating the plutonium aerosol are being investigated. Alpha counting capabilities are being expanded to accommodate samples from the laboratory experimental study.

II. FIELD SAMPLING

A. BACKGROUND

AEC plants participating in this first phase of the field sampling program fall within the jurisdiction of the AEC Albuquerque Operations Office. These include: Los Alamos Scientific Laboratory, Mound Laboratory, and Rocky Flats Plant. These three plants, represent many different production and research operations, utilizing both ^{238}Pu and ^{239}Pu, and are expected to provide suitable air sampling data for evaluating source terms under a variety of "worst normal" conditions. We have defined "worst normal" the highest range of activities generated as a result of routine operations. "Worst normal" would not include an accident situation, or rarely-performed maintenance operation, which causes an unusual aerosol release.

Generally only the source term, or particles incident on the exhaust HEPA filters at each location, is being measured, however, some air samples have been obtained (but not yet analyzed) downstream of those HEPA filters which are routinely tested using DOP. These data will define HEPA filter efficiency for air cleaning systems which have previously been in-place DOP tested.

Wide variations of activity concentration were expected, primarily due to differences in the amount of material handled at each location. Differences in size characteristics were also expected for two reasons: 1) plutonium handling procedures (mechanical and chemical) differ markedly at each site; and 2) aerosols incident on the main exhaust filters may undergo previous

stages of filtration at the glovebox. Therefore, results of this study will necessarily be reported as ranges of values to describe the source term and aerosol size characteristics.

Due to the nature of some operations performed in the plants involved, sampling locations and operational processes will be identified only in these general terms:

1) general research and development (r & d)

2) recovery (chemical operations)

3) production (mechanical operations)

It should suffice to report source term data without tying them to a specific location. If more detailed reports relating aerosol sources to types of operations are needed, these may be prepared with a limited distribution.

The field sampling program is expected to terminate at these three plants by mid-September. Sufficient data defining these source terms should be available early in the next quarter to permit initiation of the laboratory study.

B. EXPERIMENTAL APPARATUS AND TECHNIQUE

1. SIZE SEPARATION BY ANDERSEN IMPACTOR

The 8-stage Andersen impactor[1] was selected to be the primary sampler for particle size analysis. This device is simple, easy to operate in the field, and provides size separation over a fairly wide range of particle diameters. Size separation is based on inertial properties of the aerosol particles. Particles entering the impactor are accelerated through multiple jets and directed toward an impaction surface. Larger particles having

sufficient inertia leave the flow path and impact on the collecting surface. Smaller particles (particles with less inertia) continue on to succeeding stages in which smaller jets and higher velocities cause them to eventually deposit according to their aerodynamic size. Particles sufficiently small to clear all stages of the impactor are collected by a backup membrane filter (MF) which in effect becomes a ninth collection stage.

The backup MF and the eight impaction plates undergo radiometric analysis by alpha counting. The net count rate can be considered proportional to the mass of particles deposited on a plate if two assumptions are made: 1) absorption of alpha particles in the particle (self-absorption) is negligible, and 2) specific activity (dpm/gm) is constant. According to Anderson[2], a 5 MeV alpha particle has a range of at least 10 μm in PuO_2, compared to a physical diameter of 3-4 μm collected at the first stage of the impactor. An alpha particle emitted anywhere in a 4 μm PuO_2 particle should emerge from the particle with sufficient energy to be counted. Spectroscopy samples, discussed in more detail later, support this hypothesis for the particle size range of interest in this study. Constant specific activity of plutonium deposited on the impactor samples is a valid assumption if one Pu isotope is handled and if the form of Pu (density) does not change. One of the three facilities discharges a mixture of ^{238}Pu and ^{239}Pu in highly variable ratios. The complexity of this situation and some alternatives to the present methods of analysis will be discussed in Results and Discussion Section.

Impactor calibration information provided by the manufacturer has been utilized in determining size of particles deposited in impactor stages. Calibration of the Andersen impactor by Flesch et al[3] and May[4] verifies these calibration factors consistent with required precision and conditions of operation used in this study. Calibration factors of the manufacturer, and referenced studies are presented in Table I as aerodynamic diameters in terms of 50% effective cutoff diameters (ECD). The effective cutoff diameter (ECD) has been utilized by Mercer[5] and Parker and Buchholz[6] to describe the size range of particles which impact on a given stage of the impactor. Obviously the particles will not be all of one size but spread over a narrow aerodynamic size range described by a typical S-shape collection efficiency curve. Effective cutoff diameter defines the particle size which is collected with 50% efficiency on that stage. Because of the actual cutoff characteristics, some particles larger than the ECD will pass that stage and some particles smaller than the ECD will be collected on that stage. For the purposes of describing a size distribution utilizing impactor data, it is convenient and reasonably accurate[5] to assume step function cutoffs at the ECD's, and include in cumulative form (cumulative percent less than a given size) all particles collected on a given stage as smaller than the stage ECD. This technique requires analysis of the size distribution by assigning the ECD (noted in Table 1) of a stage to the next successive stage.

Cumulative percent and aerodynamic diameter, plotted

on log probability graph paper and analyzed by graphical methods[7], directly define aerodynamic activity median diameter (aamd) and geometric standard deviation (σ_g) of the size distribution. The aamd is closely related to aerodynamic mass median diameter (ammd). Results of size measurements will be reported in terms of these size parameters rather than converting to frequently used alternates such as microscopic count median diameter or mass median diameter. Aerodynamic diameter is the parameter of concern since it defines the aerodynamic properties of a particle (or cluster) which in turn defines the probability of filter penetration.

Evaluation of the Andersen impactor has indicated a potential error due to particle rebound; i.e. the particle does not adhere to the first plate it contacts[8,9]. The particle may then deposit on the next stage, or rebound from all succeeding stages and be collected by the backup filter. In either case the aamd would be reduced, and the σ_g would increase. Use of a "sticky" impaction surface would interfere with alpha counting procedures and was not acceptable. To evaluate the problem of Pu particle rebound, several types of membrane filter media have been applied to the surface of the impactor plates. Knuth[9] suggested that a glass fiber filter (MSA 1106B) provides the best retention and minimizes rebound. His tests, however, used polystyrene latex spheres, not Pu aerosols, and operated the impactor with much heavier particle loadings than encountered in this study. Limited qualitative tests at LASL has shown plutonium to adhere well to bare stainless steel plates; therefore, rebound may be of minor significance. In quantitative tests now under way, adjacent impactor sampling streams

have been installed at one sampling site. Samples have been obtained covering the plates of one impactor with Gelman vinyl metricel membrane filters and leaving the plates of the other impactor bare. By alternating the MF coating on each impactor each day, data indicating rebound (or no rebound) can be developed despite possible slight differences between the two sampling streams. Although some reduction in particle rebound may be attained by application of a MF plate coating, efficiency of the radiometric technique measuring plutonium concentration on each impactor stage may suffer to an unknown extent due to absorption of alpha particles imbedded in the filter media. The choice would then be to select a material sufficiently hard to resist burying of particles while reducing rebound. Vinyl metricel was selected as a material which might meet these requirements.

Alpha spectroscopy has provided information on the ratio of two plutonium isotopes present at one sampling site, and a qualitative indication of absorption of alpha particles by filter media applied to impactor plates as an anti-rebound agent. If energy peaks are well defined and consistent with the 5.5 MeV ^{238}Pu alpha and the 5.16 MeV ^{239}Pu alpha, little alpha absorption occurs in the media. If the energy peaks are degraded and show significantly lower alpha energy, then use of the filter media and the radiometric technique are not compatible. A limited number of samples for spectroscopy have been collected on the following filter media:

Gelman DM-800 vinyl metricel membrane

Whatman 41

Gelman Type E glass fiber

These data will be analyzed early in the next quarter. Preliminary analysis of Gelman vinyl metricel media showed very little absorption and indicate its suitability for use as a sampling filter, and impactor anti-rebound agent.

2. ACTIVITY CONCENTRATION MEASUREMENTS

A sampling stream parallel to the impactor stream, contains a 47 mm Gelman DM-800 (0.8 μm pore size) vinyl metricel membrane filter in a Gelman in-line filter holder. This sample defines total alpha activity concentration. An air sample is drawn through this filter at 1.0 cfm and should collect the same amount of activity as the adjacent impactor. The sampling streams are shown schematically in Figure 1. Alpha counting of the MF is similar to that for impactor plates. The vinyl material is also resistant to the acid mists found in the glovebox ventilation ducts.

Identical flow instrumentation and calibration techniques have been used on all streams. Bendix 0-2 cfm rotameters have been calibrated and corrected for altitude differences between sampling sites with a laboratory spirometer serving as the primary calibration standard.

3. SELECTION OF SAMPLING LOCATIONS

Reasonably accurate samples of particulate matter can be obtained from a rapidly flowing air stream, such as a ventilation duct, if the following rules are observed:

a) Use of small (less than 0.75" dia.) sharp-edged tubes facing the flow in the duct and drawing at a rate which makes velocity in the tube equal to velocity in the duct (isokinetic sampling)[10].

b) Place the probe at a location 5-10 duct diameters downstream from the last flow disturbance such as a bend or junction.

c) Keep the probe as short and straight as possible, smooth on the inside, and vertical in orientation[11].

d) Obtain a velocity profile by pitot tube across the duct at the sampling location and position the probe in a relatively constant velocity portion of the duct.

e) When compatible with a) above, set probe velocity at "flow of best transmission" which is described by a Reynolds number of 2800 in a region between laminar and turbulent flow[11].

These rules have been generally followed at all locations. Space limitations, short duct runs, liquid in bottoms of ducts, and high velocities have necessitated an alternate approach in some locations. Table 2 summarizes the sampling arrangement at each location and notes any exceptions to ideal sampling conditions. Flow in both impactor and MF sampling streams has been fixed at 1.0 cfm due to flow requirements of the Andersen impactor. Sampling probes have been sized for isokinetic sampling. In most cases duct velocity has been high enough to yield high Reynolds numbers inside the sampling probes. Sampling errors due departure from isokinetic sampling for ordinary reasons such as

erroneous sample flow settings or changes in ventilation flow are expected to be less than 1% for the small particle sizes encountered to date. Particles under 5 μm are sampled with greater than 99% efficiency if a departure from isokinetic conditions of ± 10% occurs[10]. It is believed that changes in actual flow conditions are less than 10%, and maximum particle size is on the order of 5 μm.

Sampling locations at each plant were selected to monitor the more contaminated air streams in each plant.

4. SAMPLE COLLECTION AND PREPARATION

Sample sets consist of 8 impactor plates (or the membrane filters used to cover them), the backup membrane filter (MF2), and the direct membrane filter sample (MF1). Samples are collected for varying lengths of time, depending on the activity in the duct. For convenience of handling, counting, and shipping, activity per sample is limited to about 10^5 dpm. The time required to collect a sample of this order of magnitude varies greatly for each sampling location. To arrive at the "worst normal" source term, sampling periods are selected at times when normal activities in the building are underway; i.e., not at lunch time or break time or near quitting time.

Samples are removed from impactor and MF holders at convenient locations, usually in a nearby fume hood or at a table in still air. To prepare each sample for shipment and counting, the sample is placed temporarily in a disposable petri dish for transport to the preparation area. The impactor plate or the MF

(with a blank backing plate) is then wrapped in a mylar film (.00025" thickness); identified; and placed in an interlocking plastic envelope. The samples are then shipped to LASL for counting and analysis. The shipping department of each plant ships each set as exempt quantities of radioactive material. To date, packaging and shipping has proceeded without any difficulties.

5. SAMPLE COUNTING AND DATA ANALYSIS

A gas flow proportional counter with a 9"x4" detector is presently used to count alpha activity on each sample. This counter is approximately 33% efficient for bare samples and 22% efficient for standard sources covered with a .00025" mylar film. Net count rate is determined for each sample and utilized as previously discussed in determining cumulative percent smaller than a stated aerodynamic particle size. Net count rates and the corresponding effective cutoff diameters (ECD) are submitted for computer analysis as a data set of 8 values to be fit by minimum chi-squares method[12] and plotted on a log probability grid. This analysis defines aerodynamic activity median diameter (aamd) and geometric standard deviation (σ_g) for the distribution and the deviation of each data point from the best fit line. The latter value can be utilized in a test to confirm or reject the assumption of log normality of the distribution. After a large number of samples are obtained and analyzed, a range of aamd and σ_g will be obtained to describe variation in the size characteristics of typical Pu aerosols.

6. ESP SAMPLING

Size information by microscopy has been sought to supplement information obtained by the Andersen impactor. The LASL point-to-plane electrostatic precipitator[13] was utilized to collect samples on electron microscope grids. Physical size, numbers, and appearance of the collected aerosol can be examined by electron-microscopy at high magnification. Sampling and resolution difficulty encountered in this process are discussed in Results and Discussion.

C. RESULTS AND DISCUSSION

1. WORST NORMAL CONCENTRATIONS

Analyses of plutonium samples taken during the first 3-1/2 months of the program show wide variations in activity concentrations. At one sampling location (00) concentrations were relatively low and were averaged over 7 or 8 hours. By contrast, another location occasionally yielded samples greater than 10^5 dpm in only one minute's sampling. Whether one minute samples taken routinely at this plant coincide with worst normal conditions is a matter of some concern and probably will only be answered by analysis of additional samples obtained during periods of high operational activity.

Daily data available for three of the five sampling locations initially proposed for this study are expressed as activity concentration versus time in Figures 2 through 6; (Location 00 - Fig. 2-4; Location 11 - Fig. 5; Location 14 - Fig. 6). The lower activity on most weekends at Location 00 is apparent; however,

very large variations in total activity (factors of 10 or more) also occur during the work week. Differences between these three locations appear primarily in the magnitude of activity concentration and in the frequency of peak values.

The arithmetic mean of all observations taken on normal working days (all three locations) and on weekends (Location 00 only), along with extremes, are presented in Table 3. Activity concentration (A_s), determined by summing activity on 8 impactor plates plus the backup filter MF2 is included for comparison with activity indicated by the membrane filter (A_1). Generally good agreement exists between the two sampling methods indicating no serious sampling error. Data comprising the results in Table 3 will be analyzed in several other ways to establish the worst normal source term as mean values with confidence limits or percentile values.

2. AEROSOL SIZE CHARACTERISTICS

Size characteristics of the Pu aerosols defined by the Andersen impactors with various plate coating and bare plates are summarized in Table 4, which also indicates the significance of the rebound problem. Arithmetic mean values of aerodynamic activity median diameter (aamd) and geometric standard deviation (σ_g) are listed along with their extremes. Wide variations are apparent, which is not too surprising considering the large number of different aerosol-producing operations conducted in these plants.

Somewhat surprising was the size distribution of the Pu aerosol at Location 11. Size characteristics of the aerosol

measured here are distinctive due to the high percentage of very small particles in the distribution. In the 14 impactor measurements made to date, the cumulative percent smaller than 0.43 μm (final stage ECD) has averaged 60%; that is, 60% of total activity measured in the impactor sampler passed through the impactor and was collected on the backup membrane filter. A distribution of this type is not readily characterized by the Andersen impactor. At this point in the program, an alternative method has not been devised to accurately measure the size characteristics at Location 11.

Figures 7, 8, and 9 are included as log probability representations of a typical plutonium size distribution, and the extremes. Figure 7 represents a real sample which closely approximates the mean aerosol at Location 00 (aamd = 1.70 μm and σ_g = 2.33). Figures 8 and 9 represent the two extremes of σ_g. In each figure a best fit line defined by the chi-squares method[12] is provided. The aamd is obtained by projecting the "50% smaller than" point on the best fit line to the diameter. The σ_g is the 84% value of the diameter divided by the aamd.

Examination of the data in Table 4 reveals no apparent particle rebound. Had the uncoated plates displayed a lower aamd and higher σ_g than coated plates, or larger amounts of activity on the backup filter, a rebound problem would be suggested. Preliminary results of rebound tests conducted with twin impactors has also been reviewed. Two impactors, one with plates coated and the other uncoated, were operated concurrently in adjacent

parallel streams close to the same point in a ventilation duct. They indicate very similar size characteristics and fail to show any indication of particle rebound, consistent with data from Table 4 noted above.

3. ESP SAMPLES

At one sampling location (00), a point-to-plane electrostatic precipitator (ESP) was utilized to obtain samples of the Pu aerosol for sizing by electron microscopy[12]. It appears that this technique will be of limited usefulness because 1) plutonium particle concentrations are low, necessitating long-term sampling and averaging of data which obscures short-term characteristics of the size distribution, and 2) the presence of many non-radioactive particles obscured the identity of Pu particles in electron micrographs. Useful information was derived from the ESP sampling, since they show no chain agglomerates. This indicates that the measured size data deals only with individual particles and not with unstable agglomerates.

4. SPECTROSCOPY

Spectroscopy of several samples has shown the presence at Location 00 of ^{238}Pu and ^{239}Pu in similar quantities. Spectroscopy at the other two locations has not been considered necessary since only one principle isotope is used at these facilities. However, the mixture of two isotopes complicates measurement of size characteristics because the ^{238}Pu and ^{239}Pu particles contributing to the total alpha activity on each impactor plate originate at different operations and probably are related

to different size distributions. Figure 11 represents a size distribution which is very likely a composite of two different aerosols with different size characteristics. The composite distribution does not display log normal size characteristics which are convenient in describing an aerosol size distribution.

Samples to investigate the makeup of composite distributions by spectrographic means have been obtained and are undergoing analysis. One set of samples, consisting of five MF samples taken on consecutive working days, has been obtained to provide ^{238}Pu to ^{239}Pu ratios. Another set of samples will provide ^{238}Pu to ^{239}Pu ratios for each impactor stage. This information may correlate with the composite distribution, showing a preponderance of one isotope at one extreme of the size spectrum.

5. FILTER EFFICIENCY DATA

Sampling downstream of exhaust filters with an impactor and membrane filter has been in progress at Location 00 since mid-March. This site is suitable for efficiency measurements because its exhaust HEPA filters are DOP tested in place to assure a 99.95% or greater efficiency. This in place test indicates that the filters and the filter housing are in satisfactory condition, and radiometric efficiency measurements should be applicable to the filter media performance, and not obscured by leakage. Daily downstream samples obtained concurrently with upstream samples yielded too little activity for accurate count statistics. Longer sampling times downstream were needed without losing the correlation of size characteristics upstream and downstream of the filter. This was

accomplished by operating the downstream samplers concurrently with the daily upstream samplers, but without changing the downstream sampler each day. The downstream sampler generally covered a one week period.

Penetration data and size characteristics of particles penetrating the filter are being analyzed.

III. LABORATORY STUDIES

The laboratory experimental phase of this study will use field sampling data as a basis for defining the experimental plutonium aerosols used to test multiple stages of HEPA filters.

A. QUALITY CONTROL HEPA FILTER TESTING

To have greater confidence that the HEPA filters used for multiple HEPA filtration of the plutonium aerosols, are of high quality and good physical condition, a quality control polydisperse DOP test system has been constructed. This system consists of an ATI 8-jet DOP aerosol generator, 3 inch duct with provision for installing two 8"x8"x3-1/16" HEPA filters in series, sampling ports upstream and downstream of the second HEPA filter, an orifice meter for monitoring the air flow, a LASL light scattering photometer[14], and a Lamb electric high vacuum blower (Figure 11). Air flow through the system can be adjusted for operation at the rated filter air flow rate (25 cfm) and at reduced air flow rates consistent with more severe test methods.

B. ESTIMATION OF HEPA FILTER EFFICIENCY FOR THE SECOND OF TWO FILTERS IN SERIES.

The quality control test system provides a means to estimate the DOP efficiency of a HEPA filter, which is preceded by a HEPA filter. The DOP aerosol generator is operated at 15 psig producing a highly concentrated (not measurable) aerosol upstream of the first filter, so that the aerosol concentration downstream of this filter ranges from 0.4 to 1.0%. This constitutes the upstream aerosol concentration for the second HEPA filter in the series. The LASL

light scattering photometer has several sensitivity range settings from 100% full scale to .01% full scale. The minimum detectable limit of the instrument is at least 5% of full scale on the .01% scale since its linearity and stability are good. Using this minimum detectable limit we can define a minimum efficiency of 99.9% to 99.95% for this second HEPA filter. This is based on the following:

Minimum detectable limit = 5% of full scale, or

.05 x .01% = .0005% = minimum detectable limit.

If upstream photometer reading = 1.0%, then

$\frac{.0005\%}{1.0\ \%}$ < .05% penetration, or

filter efficiency > 99.95%

If the upstream concentration is only 0.5%, then the filter efficiency could only be defined to be > 99.9%. Assumptions inherent in using this analysis are a) assurance of a minimum detectable limit of 5% of full scale at the .01% range and b) unimportance of loading of the first HEPA filter so that the source term to the second filter is affected. Photometer stability is adequate to satisfy a), and the first filter is changed frequently (for each test) to minimize any loading effect.

Table 5 shows data obtained for three HEPA filters tested in the above manner. Upstream concentration varied from 0.38 to 1.55% while downstream concentrations were measured as zero, but assumed to be < .05%. How much below .05% is not known, so that we can only say that the efficiency of the second HEPA filter is greater than a calculated value. In all cases the efficiency is greater than 99.9%

C. PLUTONIUM AEROSOL GENERATION

Several methods for the production of plutonium aerosol are being investigated. High plutonium concentrations are necessary to provide measurable activity after the second or third HEPA filter in series.

Burning of plutonium metal was initially considered the method of interest primarily because this has been done at LASL and could be a relatively simple procedure. However aerosol output may vary as a function of time and temperature, and aerosol size characteristics may be difficult to control[15].

Consideration was given to the use of an ultrasonic nebulizer because of its high output. This method produces a highly variable aerosol output with high wall losses and has been abandoned.

The Lovelace nebulizer[16] seems to meet the requirements for this study. It is a cheap, simple, air operated, constant output device, with the only drawback being its limited output of approximately 50 µl/min. To retain the good qualities of the Lovelace nebulizer, but increase its output, an alternate aerosol generation technique under consideration is the Re Tec nebulizer[17] which reportedly will dispense 260 µl/min. In addition, the Lovelace Foundation has developed a "Vortex" nebulizer which is capable of aerosolizing more than 1 ml/min.

The possibility of applying heat and vibration to a finely ground powder of PuO_2 is also under consideration. Control

of particle size depends on the degree of ball milling of the oxide. The rate of output for this procedure is unknown and would have to be investigated.

Nebulization methods for aerosol generation mentioned above rely on the availability of a suitable plutonium solution. Various chemical processes exist for the formulation of these solutions and should pose no great problem. With atomizer methods, the aerosol may have to be passed through at least one heating column or through a mixing chamber for drying and thermal degradation[18].

D. EXPERIMENTAL TEST SYSTEM

Plans for the laboratory experiment include a 2-module system for better containment and control of the airborne particulates.

The first glove box module (Figure 12) will contain the aerosol generator, heating columns, diluting air supply, upstream sampler number 1, HEPA filter number 1 and a sealing device which will allow the isolation of the glove box during HEPA filter changes and loading and unloading of the air samples. An airlock or passbox will be utilized to transfer the samples and change filters in and out of the glove box.

Module number 2 (Figure 13) will be made up of one or a series of hoods which will house HEPA filters #2 and #3, samplers #2 and #3, filter to be used as a sampler downstream of HEPA filter #3 and the vacuum pump to be connected to the house exhaust system.

An orifice meter will be placed downstream of the last filter for monitoring air flow rate.

Andersen impactors will be used to sample upstream of the first HEPA filter to verify the characteristics of the source plutonium aerosol.. These samplers will also be used downstream of the first two HEPA filters. Millipore filter samples will also be taken at each of those points, with an additional glass fiber filter across the entire output downstream of the third HEPA filter. Because such a small amount of aerosol is expected to reach that point, it is desirable to collect it all to provide better counting statistics. The Andersen impactor, which is identical to the samplers used to collect the field data, will give aerosol concentration as well as size information.

A new alpha counting system which will accommodate the 3-1/4" diameter samples obtained from the Andersen impactor, is being constructed. This counter will utilize Sylvania Type 130 scintillator paper in intimate contact with the sample plates. In this manner efficiency losses are minimized in that the alpha particles do not have to penetrate the mylar covering of the sample or traverse any distance before interaction with the scintillator. The counting system will be housed in a 30' x 10' trailer which will be placed behind and adjacent to the OHL building.

Planning and design phase of the experimental test system is almost complete and assembly will begin shortly.

IV. WORK FOR NEXT QUARTER

A. Activation of sampling locations 04 and 08 will occur early in July. No additional sampling locations are planned until analysis of data from present locations can be completed. Location 08 will be abandoned after 2 - 4 weeks additional sampling to provide a new more active location in the same building.

B. Alternate means of size analysis will be considered to more adequately describe the small aerosol at Location 11 since the Andersen impactor does not appear to be adequate for this aerosol.

C. Samples taken simultaneously upstream and downstream of Location 00 exhaust filters will be analyzed by spectroscopy to define isotopic mixture.

D. Approximately one additional week of sampling with twin impactors is planned to fully evaluate the particle rebound problem.

E. Analysis of size and activity concentration data in terms of mean values and confidence limits will be completed. Results will be reviewed for need to continue sampling at individual locations.

F. Concurrent upstream and downstream sampling data from Location 00 operations will be analyzed to provide an estimate of in-place filter efficiency against plutonium.

G. Size and space requirements for the laboratory experimental program will be finalized and arrangements made for installation.

H. A final decision will be made as to the choice of aerosol generator and associated equipment, and assembly of the multiple HEPA filter test system will be completed. Aerosol generation procedures to check particle size characteristics will be conducted. HEPA filters to be used will be quality control tested at LASL.

V. REFERENCES

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VI. ACKNOWLEDGMENT

The cooperation, guidance, and assistance of the following people in establishing the program is gratefully acknowledged:

W. D. Kittinger, Dow Chemical, Rocky Flats Plant
E. A. Putzier, Dow Chemical, Rocky Flats Plant
D. G. Carfagno, Monsanto Research, Mound Laboratories
W. H. Westendorf, Monsanto Research, Mound Laboratories
J. F. Bresson, AEC, Albuquerque Operations Office
W. L. Workman, H-1, LASL
W. D. McNeese, CMB-11, LASL

TABLE 1

ANDERSEN IMPACTOR CALIBRATION

Aerodynamic Effective Cutoff Diameters (Microns)
Sample Flow 1.0 cfm

Stage	2000 Inc	Flesch, et al[3]			May[4]
		Methylene Blue LM	Methylene Blue EM	Polystyrene Latex	
0	11.[a]				
1	7.				
2	4.7	6.2	5.35		5.5[b]
3	3.3	3.6	2.95		3.5
4	2.1	2.0	1.53	1.75	2.0
5	1.1		0.86	.92	1.1
6	.65			.54	
7	.43[a]				

a = Added Stage
b = Modified Stage
LM = Light Microscopy
EM = Electron Microscopy

TABLE 2

SAMPLING ARRANGEMENTS

Location Code	Isotope	Major Operations	Duct Dia.	Velocity fpm	Probe ID	Distance Free Run	Probe Orient.	Reynolds No. in Probe
00	238 & 239	r & d	36"	1850	.305"	1 D.D.[a]	Horiz.[b]	3760
04	238	r & d	24"	600	.555"	10 D.D.	Vert.	2730
08	238	prod., recovery	12"	900	.430"	10 D.D.	Horiz.[c]	3170
11	239	recovery	12"	2900	.242"	5 D.D.	Horiz.[c]	3575
14	239	production	24"	2200	.305"	10 D.D.	Vert.	4500

a Close to 90 bend; space limitation. D.D. = duct diameter

b Vertical duct run; vertical mounting not possible

c Liquid in duct; bottom entry not recommended

TABLE 3

ACTIVITY CONCENTRATIONS OF Pu AEROSOLS (dpm/m^3)

Description	Location*	# obs	A_1 (direct MF) Mean	Max	Min	A_s (Impactor + MF_2) Mean	Max	Min
Weekdays	00	65	11,388	34,590	286	9980	36,325	198
Weekends	00	15	988	4,579	51	1505	4,798	44
Weekdays	11	19	8.10×10^6	16.5×10^6	3.87×10^6	6.72×10^6	15.2×10^6	4.06×10^5
Weekdays	14	26	1.27×10^5	7.86×10^5	1,814	0.98×10^5	6.79×10^5	2,611

*Analyzed data from sampling Locations 04 and 08 not available

TABLE 4

ANDERSEN IMPACTOR

SIZE CHARACTERISTICS OF Pu AEROSOL

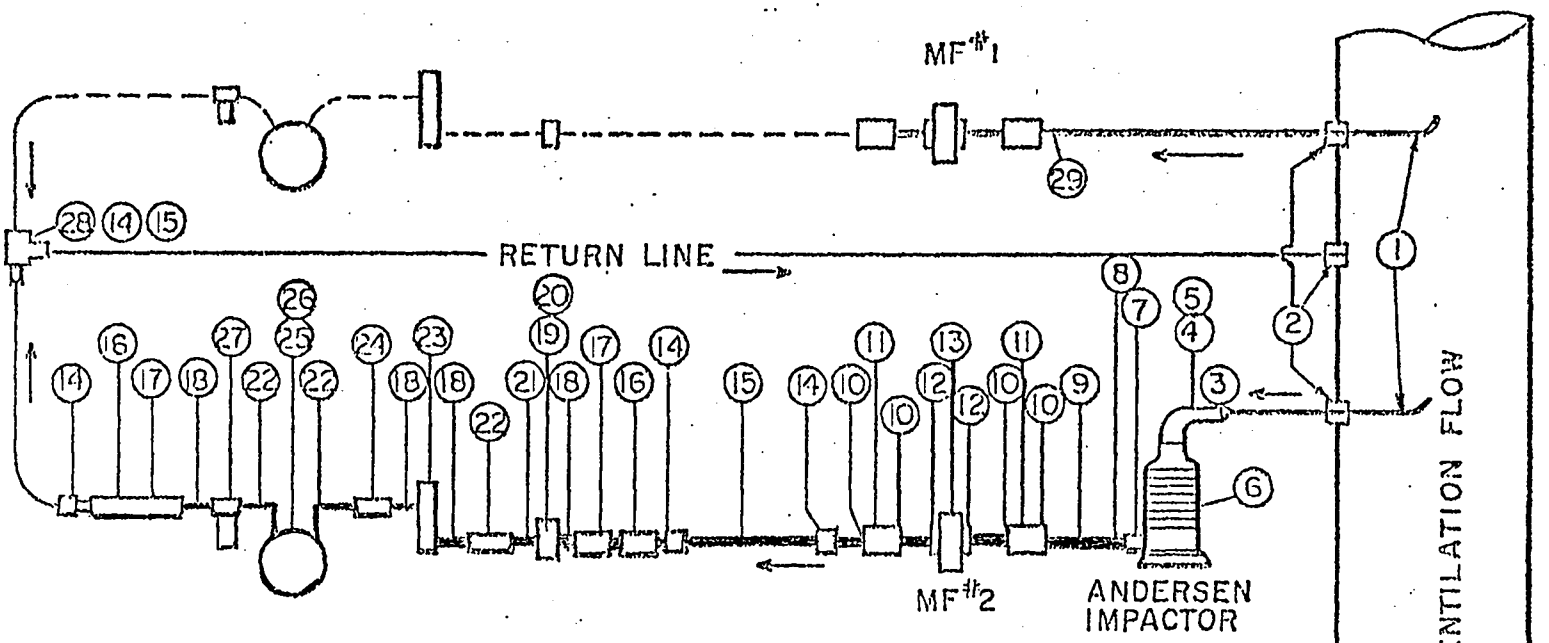
Description	Location	# obs	aamd(μm) mean	aamd(μm) max	aamd(μm) min	σ_g mean	σ_g max	σ_g min
Weekdays, no coating	00	63	1.70	3.30	.52	2.33	4.94	1.53
Weekends, no coating	00	12	1.92	3.32	1.03	2.37	3.20	2.03
All Samples, DM-800 coating	00	5	2.10	2.50	1.25	2.09	2.61	1.74
All Samples, AA coating	00	10	1.86	2.70	.92	2.38	3.37	1.83
Weekdays, no coating	14	6	2.83	3.53	1.96	3.03	6.40	1.98
Weekdays, AA coating	14	11	2.37	3.25	1.61	2.98	6.6	2.06
Weekdays, AA coating	11	14	*					

* 60% smaller than 0.43 μm; not amenable to analysis using Andersen impactor.

TABLE 5

Filter #	Flow Rate(cfm)	Photometer Reading Upstream(%)	Photometer Reading Downstream(%)	Filter Efficiency(%)*
PF002	25	.38	0.0	> 99.87
PF002	13	.52	0.0	> 99.90
PF003	25	.90	0.0	> 99.94
PF003	13	.62	0.0	> 99.92
PF004	25	.95	0.0	> 99.95
PF004	13	1.55	0.0	> 99:97

*Calculated assuming that a zero downstream photometer reading can conservatively be assumed to be less than 0.0005% (5% of full scale).



1. Probe - tube sized for isokinetic sampling (Al)
2. Bushing - 3/8"X1" (brass)
3. Reducer - 1-3/8"X3/8" (Al)
4. Elbow - 1-1/2" tubing, .065" wall (Al)
5. O-ring - 1.046" ID, 1.274" OD
6. Sampler - Andersen 8 stage impactor
7. Hose clamp - 25/32"
8. Tubing - Tygon 6" long, 3/8" ID, 1/2" OD
9. Hose fitting - 1/4"X3/8" male
10. Bushing - 1/2"X3/8"
11. Quick disconnect - vacuum 1/2" Snaptite PEC-8
12. Pipe nipple - 3/8"
13. Filter holder - Inline 47mm Gelman #1235
14. Hose fitting - 3/8"X1/2"
15. Hose - .500" ID X .750" rubber
16. Coupling - 3/8"
17. Bushing - 3/8"X1/4"
18. Pipe nipple - 1/4"X7/8" lg.
19. Filter holder - MSA DZ-78001
20. Cartridge - MSA Type H
21. Pipe nipple - 1/4"X2-1/2" lg.
22. Elbow - 1/4"
23. Flow meter - Bendix #8223 - 0.3 to 2 cfm
24. Coupling - 1/4"
25. Pump - Gast 0440A
26. Motor - GE 115 VAC, 5 AMP, 1/4 HP
27. Trap - Muffler
28. Tee - 3/8"X3/8" X3/8"
29. Bushing - 1/2"X1/8" - Brass - drill to fit

NOTES:
1. Sampling streams are identical from first quick disconnect (Item 11).
2. Male, half of quick disconnects are directed downstream.
3. Joints between Items 1 and 2 are epoxy potted unless otherwise specified.

FIGURE 1: Schematic Diagram and Parts List for Membrane Filter and Impactor Sampling Streams

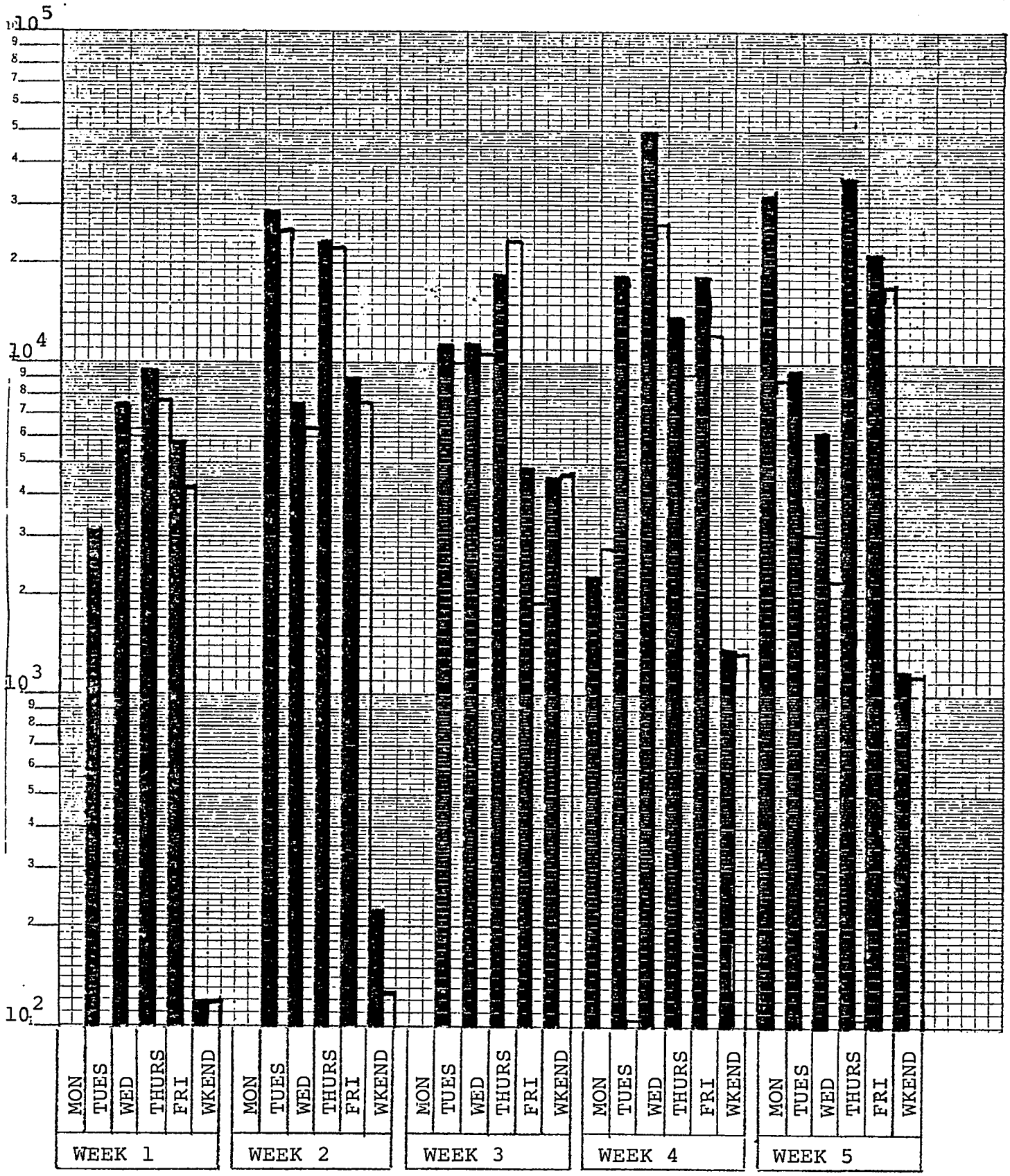


Figure 2: Activity Concentrations Incident On Exhaust Filters at Location 00. (Shaded bar = direct MF sampler; Unshaded bar = Impactor + Backup MF).

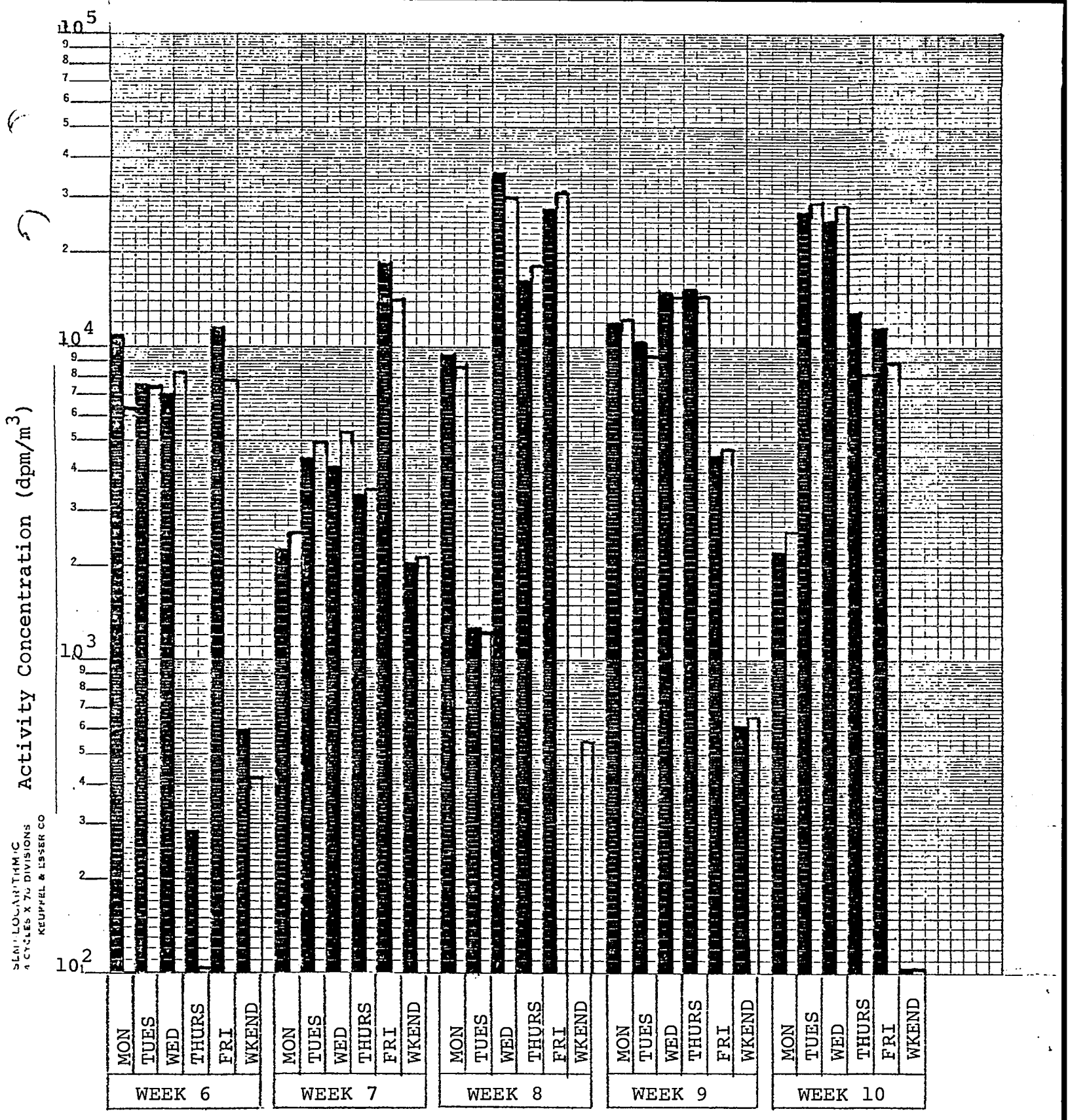


Figure 3: Activity Concentrations Incident On Exhaust Filters At Location 00. (Shaded bar = direct MF sampler; Unshaded bar = Impactor = Backup MF).

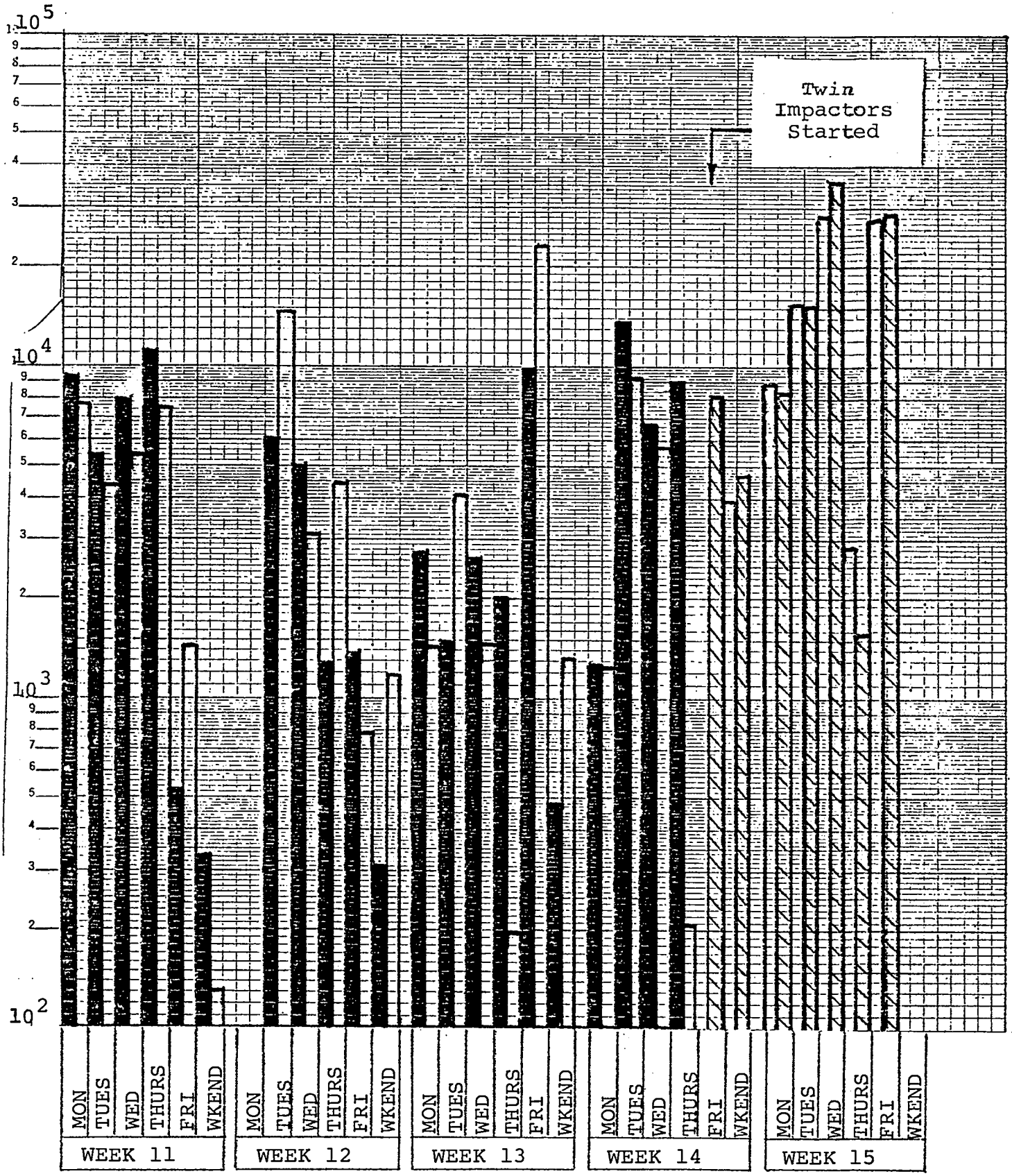


Figure 4: Activity Concentrations Incident On Exhaust Filters At Location 00. (Shaded bar = direct MF sampler; Unshaded bar = Impactor + Backup MF).

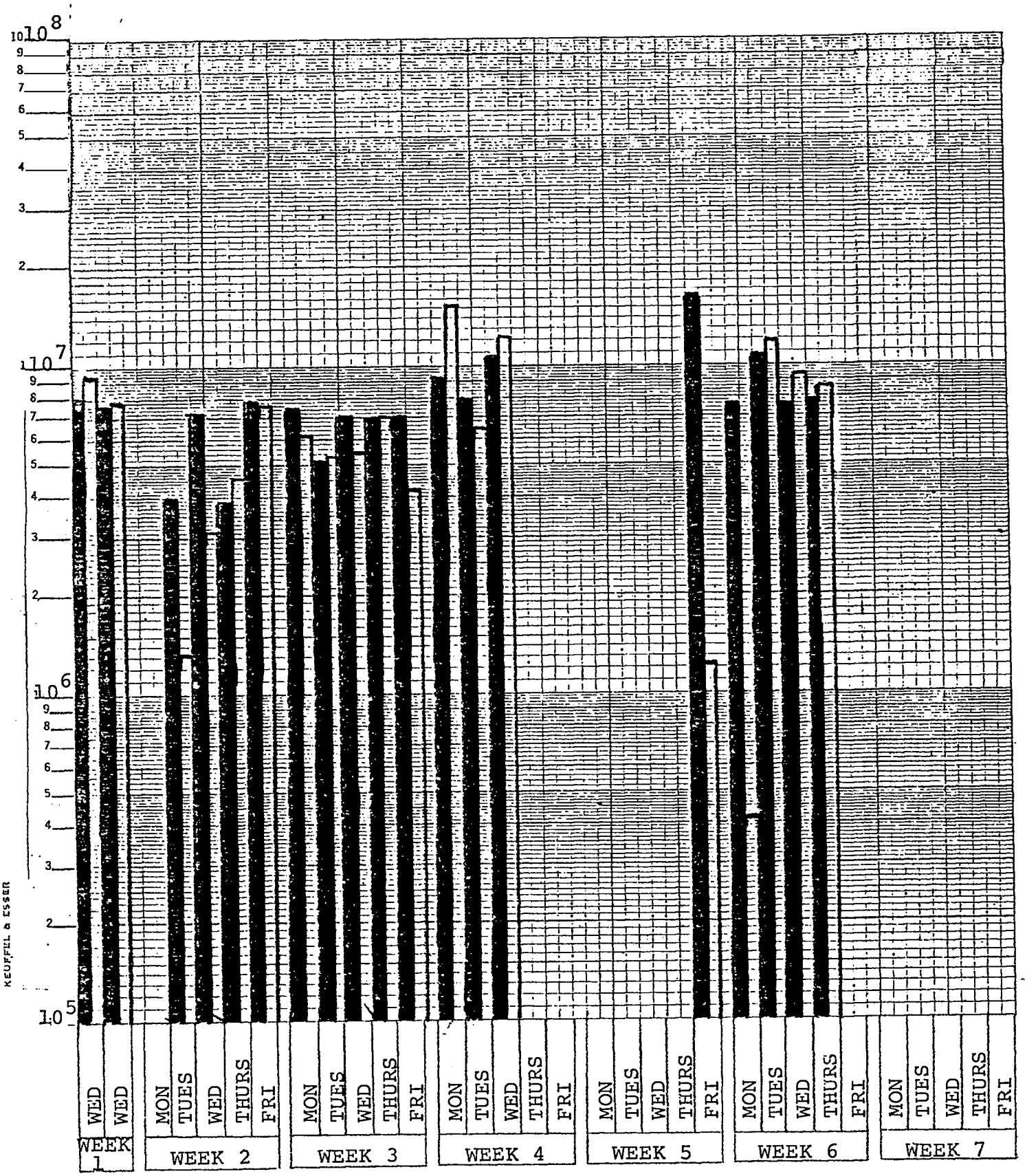


Figure 5: Activity Concentration Incident On Exhaust Filters at Location 11. (Shaded bar = direct MP sampler; Unshaded = Impactor + Backup MF).

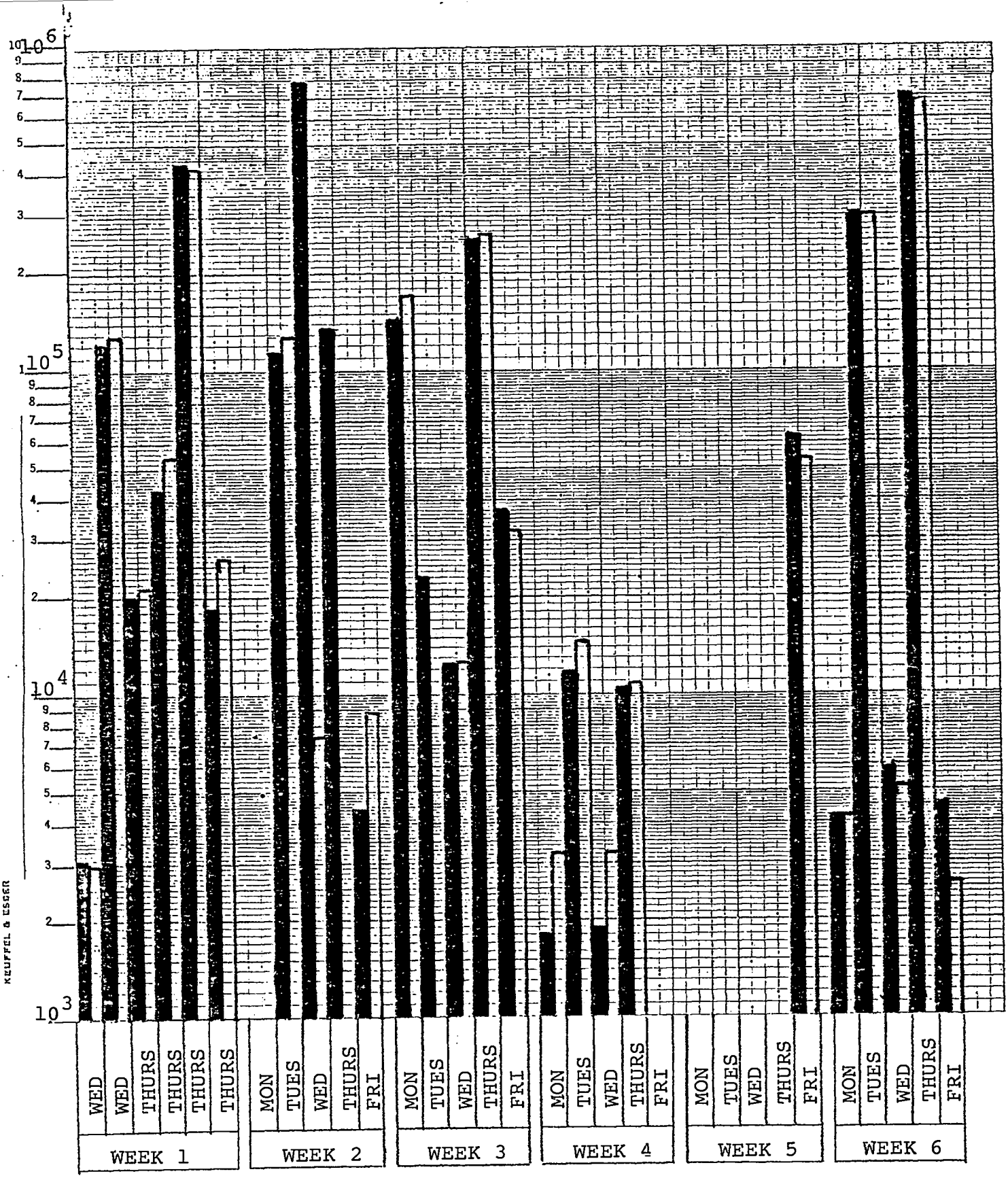


Figure 6: Activity Concentration Incident On Exhaust Filters At Location 14. (Shaded bar = direct MF sampler; Unshaded = Impactor + Backup MF).

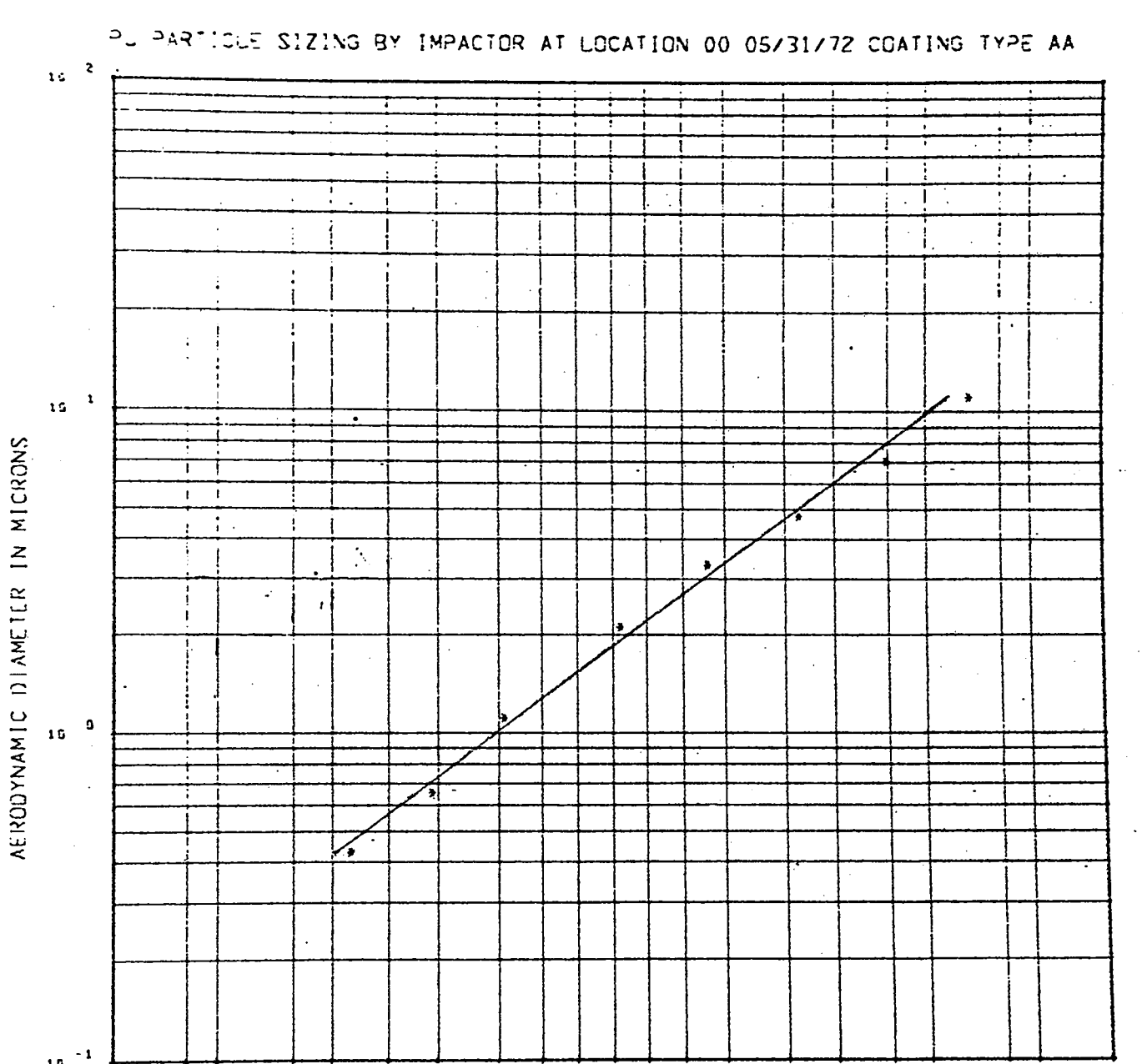


Figure 7: Size Characteristics of Pu Aerosol--
Representative of Mean aamd and σ_g.

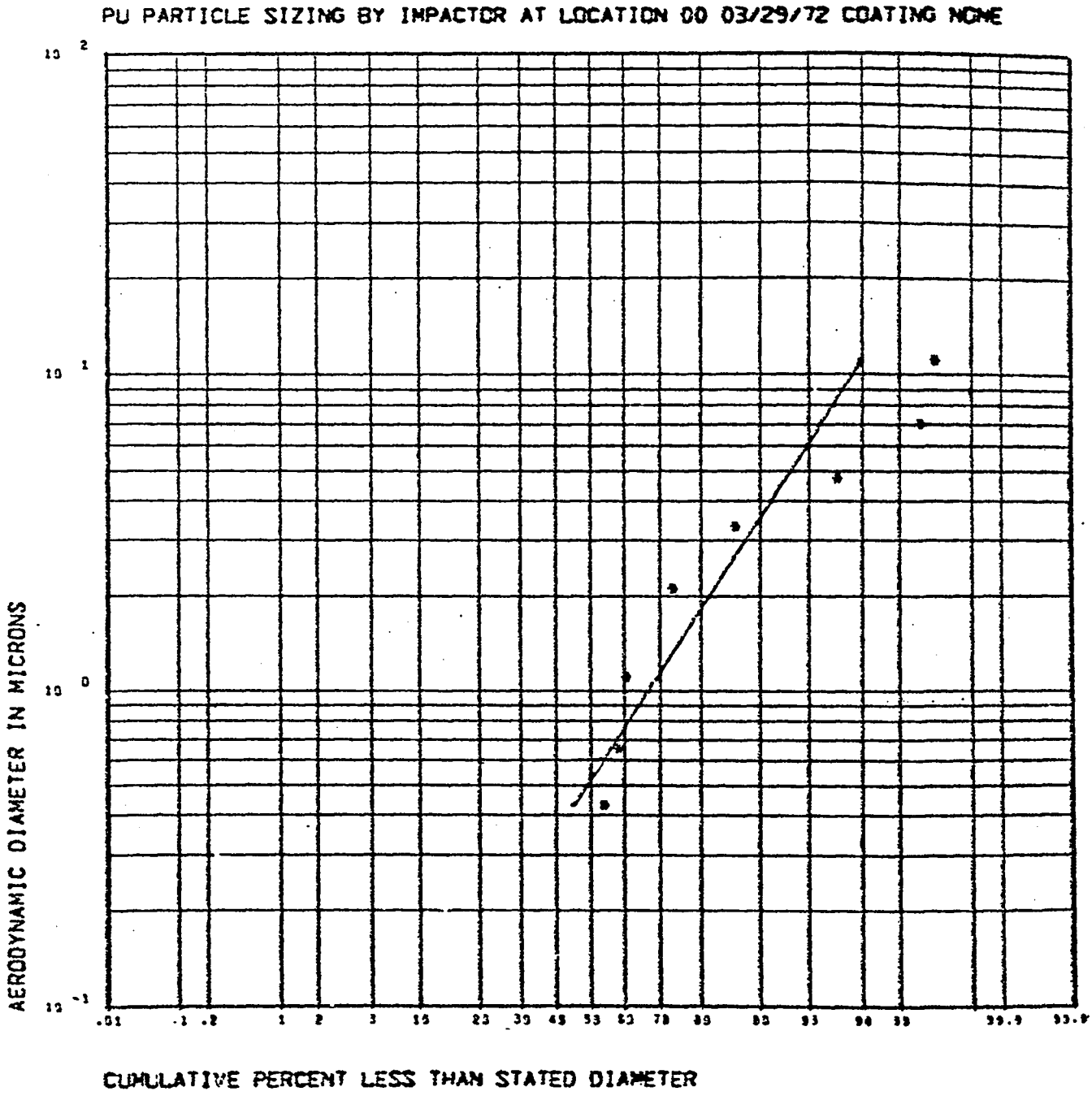


Figure 8: Size Characteristics of Pu Aerosol--
Representative of Distribution With High σ_g.

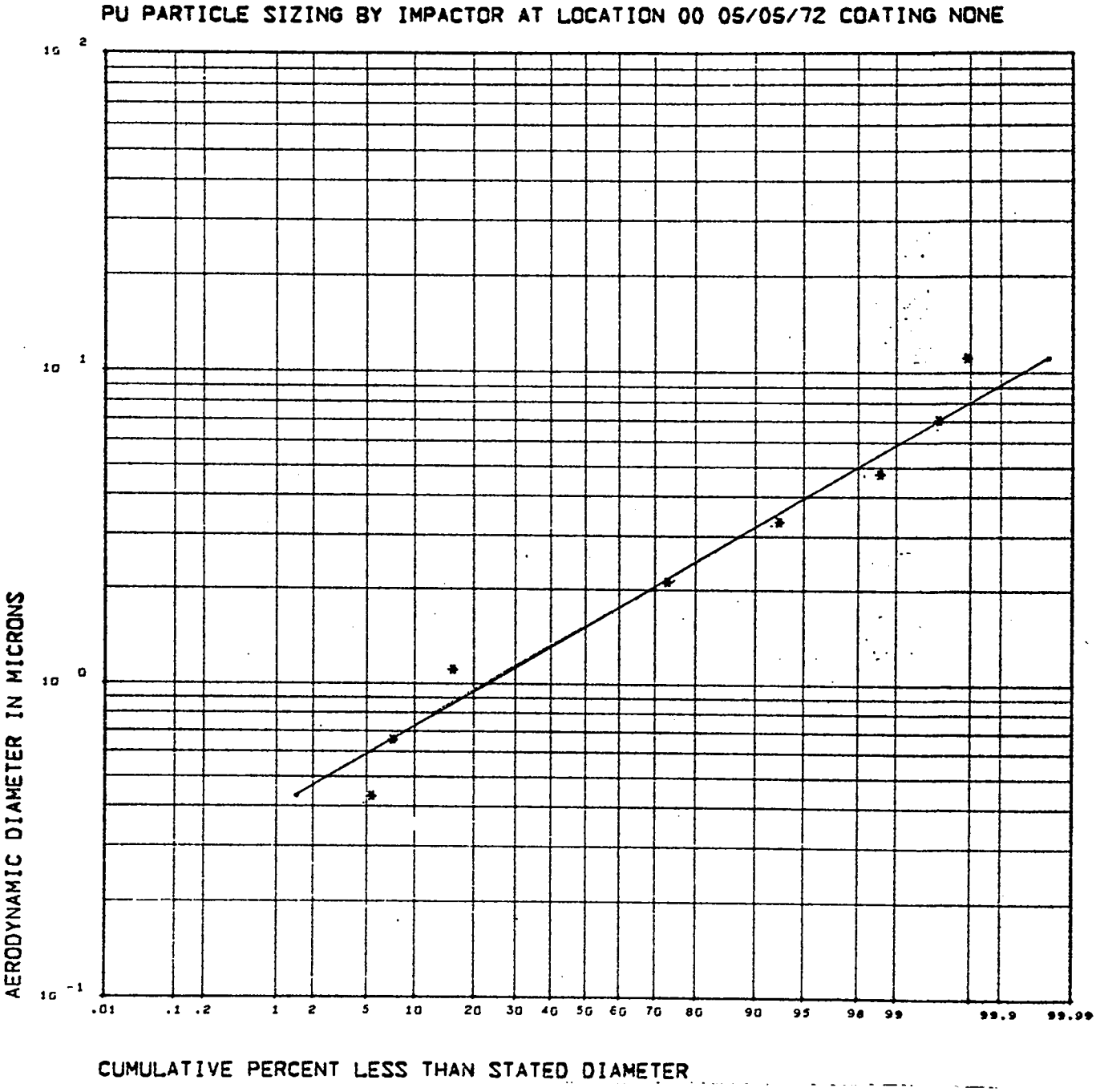


Figure 9: Size Characteristics of Pu Aerosol--
Representative of Distribution With Low σ_g.

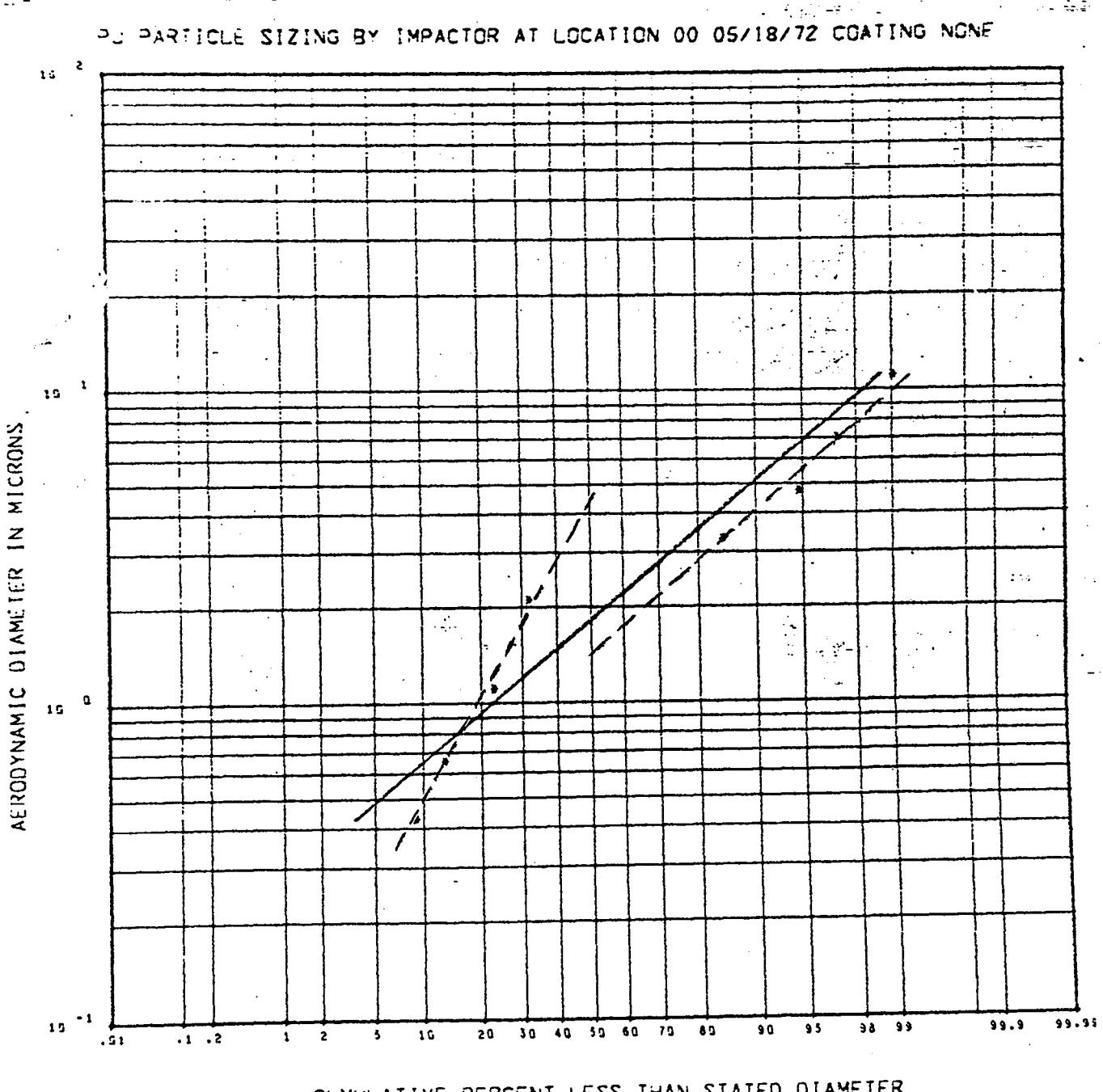
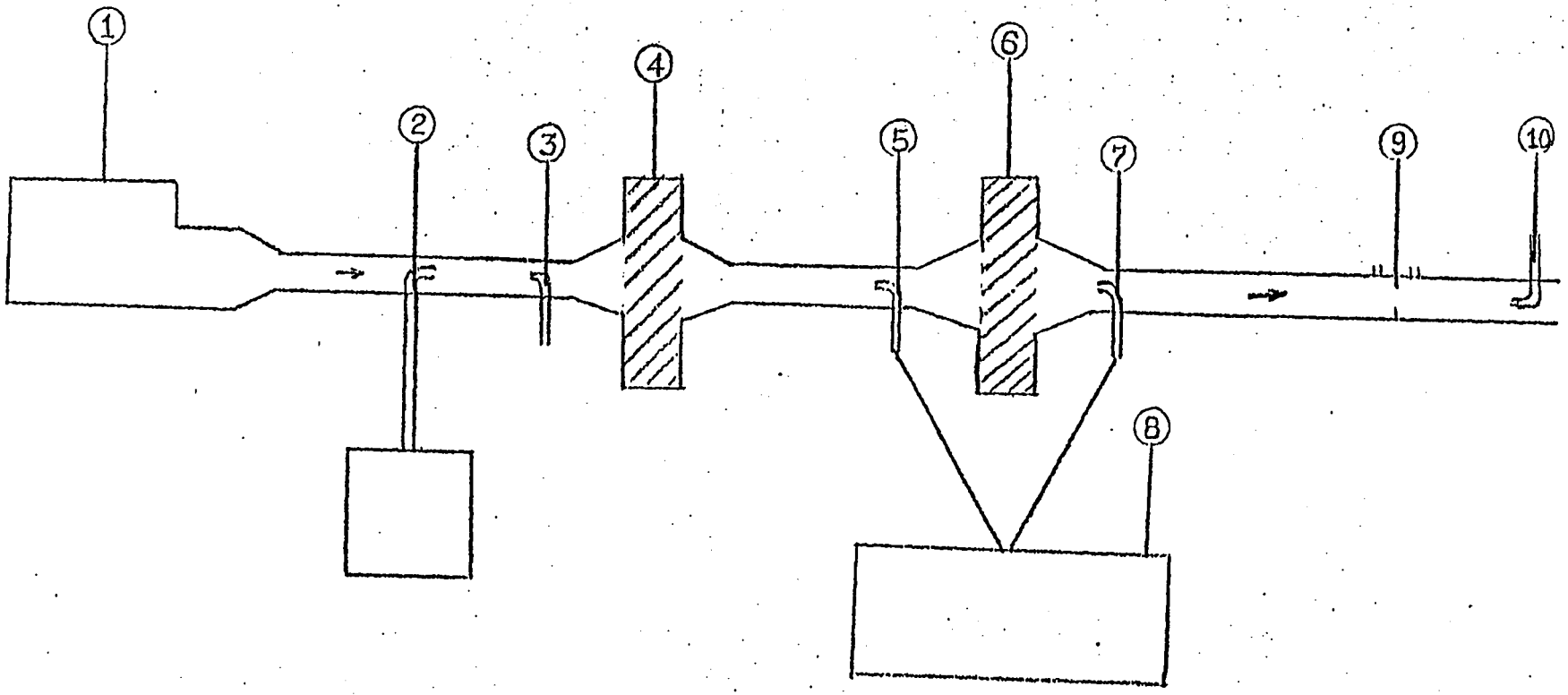


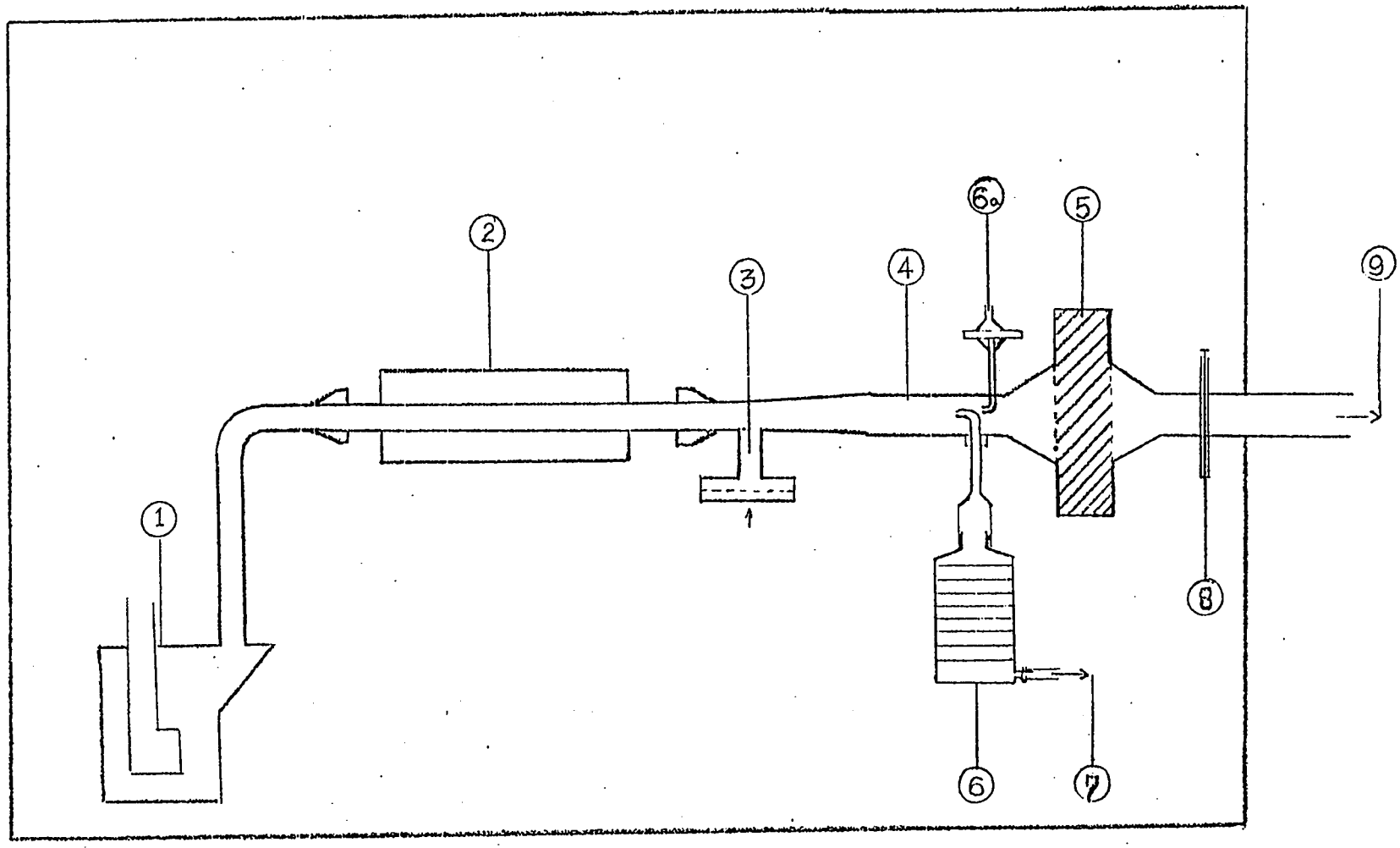
Figure 10: Size Characteristics of Pu Aerosol--
Representative of Composite Distribution.

Fig. 11 DOP Test System



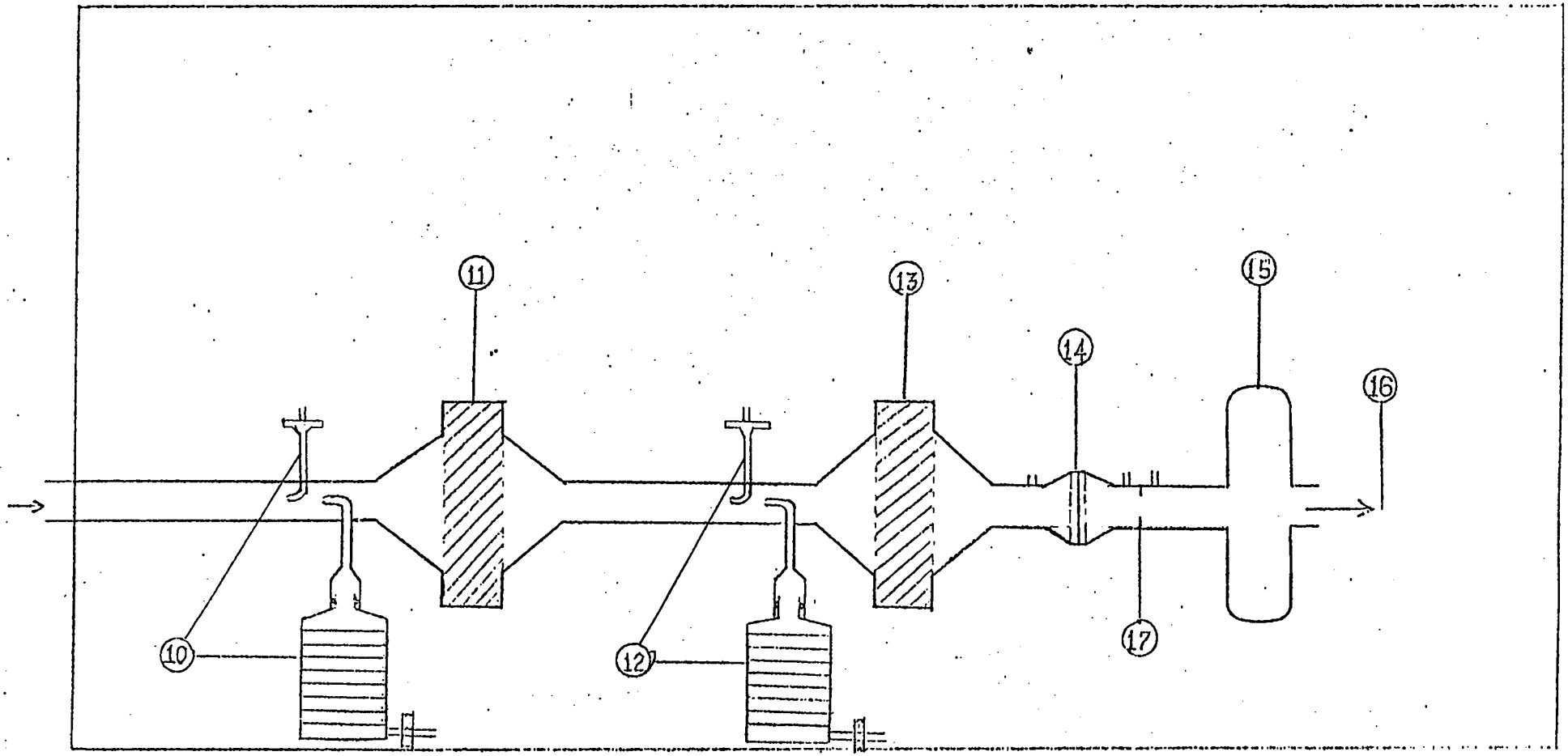
1. Blower
2. DOP Generator
3. Upstream Sampler #1
4. HEPA Filter #1
5. Upstream Sampler #2
6. HEPA Filter #2
7. Downstream Sampler
8. Light Scattering Photometer
9. Orifice Meter
10. Pitot Tube

Fig. 12 Glove Box



1. Aerosol Generator
2. Heating Column
3. Diluting Air Intake (filtered)
4. 2" Diameter Duct
5. HEPA Filter No. 1 (8" x 8" x 3 1/16)
6. Andersen Impactor (8-stage) + MF #1
7. To Vacuum Pump (May be inside or outside glove box).
8. Safety Seal
9. To adjacent Hood

Fig. 13. Hood



10. Andersen Impactor #2 + MF #2
11. HEPA Filter #2
12. Andersen Impactor #3 + MF #3
13. HEPA Filter #3
14. Glass Fiber Filter
15. Vacuum Pump
16. To House Exhaust
17. Orifice Meter