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SIZE ANALYSIS OF PARTICULATES FOUND IN AIR AT HANFORD PLUTONIUM FABRICATION FACILITIES

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SIZE ANALYSIS OF PARTICULATES FOUND IN AIR AT HANFORD PLUTONIUM FABRICATION FACILITIES

INTRODUCTION

The Hanford Radiation Protection Operation is conducting a study on the characteristics of radioactively contaminated air at Hanford work locations. One of the main objectives in this study is to define the size of particular air contamination in plutonium handling facilities. In addition to characterizing the particulate contamination, it is desirable to know the normal background particle size distribution. This report summarizes the data collected on general particle size distribution and radioactive particle distribution at two Hanford plutonium fabrication facilities.

DISCUSSION

A Royco Particle Counter, Model PC 200-A, was used in obtaining the data on the general particle size distribution at various locations in two Hanford facilities. The aerosol of interest is circulated at 100 cubic centimeters per minute through the optical chamber of the Royco Counter. White light from a well regulated lamp is focused on the 1.98 cubic millimeter sensitive volume of the 4 cubic millimeter particle chamber. The light is deflected or reflected when it strikes a particle passing through the chamber producing a light pulse with an amplitude proportional to the square of the particle diameter. These pulses are observed by a photomultiplier tube and sorted according to size by the electronics of the counter. The results are displayed in 15 channels corresponding to defined particle diameter ranges (Slide 1).

The Royco Counter was operated in corridors and laboratory areas in the Plutonium Metallurgy Building (308 Building) and in offices, corridors and

laboratory areas of the Plutonium Fabrication Building (234-5 Building). The particle size distribution data collected at the Plutonium Fabrication Building is illustrated in Slide 2. The data were collected for counting coincidence losses and are reported as particles per cubic centimeter. It may be noted that the office areas where smoking is permitted have the greatest concentration of particles for each particle size. During the day, the corridors and laboratory areas exhibited similar particle distributions. The minimum particle concentrations were found in the corridors during the night hours when most of the building personnel were absent.

The data collected at the 308 Building are presented in Slide 3. There are no office areas in the operating part of this building. Smoking was permitted in the corridors. The particle size distributions found in the corridors are quite similar to the results found in the office areas in the 234-5 Building. Again, the particle concentrations in the corridors are lowest at night when most of the personnel are absent from the building. Theparticle size distribution curves for the laboratory areas are quite similar in both buildings.

A comparison of the particle size distribution in the 308 Building for laboratory areas, supply air (filtered by a roughing filter and water washed) and unfiltered outside air is shown in Slide 4. The supply air and the air in the laboratory areas exhibit similar particle size distribution curves although the air in the laboratory areas has an increased number of both the smaller and larger-sized particles than the supply air. For the untreated outside air, the particle concentration below one micron is significantly less and above one micron is significantly greater than the concentration in the treated air. This data was collected during a dusty weather condition, probably accounting for the high number of large particles. The large number of small particles

in the building may be partially due to presence of moisture molecules in the air.

The particle counter was used in an attempt to observe changes in the airborne particle size distributions during minor plutonium contamination incidents. No significant changes in particle size distributions were observed. A calculation of the theoretical microcurie concentration for various particle sizes was performed to determine what magnitude a contamination incident would have to reach to be observed above the normal particle background seen by the Royco Counter. The calculation was made assuming the particles were in the form of PuO_2 spheres with a density of 11.5 grams per cubic centimeter. Slide 5 shows the relationship of concentration in microcuries per particle to the particle size in microns. A single PuO_2 particle at the minimum particle size (0.32 micron) detectable with the Royco Counter is equivalent to 1×10^{-8} μc of plutonium and the maximum particle size (8.0 micron) is equivalent to 1.8×10^{-4} μc of plutonium. Assuming a contamination incident where the plutonium concentration was 1 MPC_A (2×10^{-12} μc/cc), the number of pure PuO_2 particles per cubic foot for particle sizes from 0.01 to 10 microns were calculated. The data from this calculation are illustrated in Slide 6. For the minimum and maximum-sized particles detectable by the instrument, only 5.4 pure PuO_2 particles per cubic foot and only 3.5×10^{-4} pure PuO_2 particles per cubic foot respectively would be present at 1 MPC_A. For the 100 cc/min sample rate, the Royco Counter would have to operate approximately 50 minutes in a 1 MPC_A atmosphere to count one 0.32 micron PuO_2 particle and about 8×10^5 minutes to count one 8.0 micron PuO_2 particle. Very obviously, these isolated particles would not be distinguishable from the normal background particle distribution.

Since the plutonium particles are apparently smaller than the range of the particle analyzer, it was concluded that analyses would have to be determined using electron microscope procedures. Initially, contaminated air from plutonium fabrication hoods was chosen for study since this would be the source of air contamination incidents and the levels would be high enough to permit the collection of suitable samples in reasonable time periods. Membrane filters are used in collecting the radioactive particulates because of the high efficiency of the filter and its easy dissolution in acetone or ethylene acetate. A positive displacement pump coupled to two membrane filter holders is used in collecting the air samples. The second filter is used as a backup since it was impractical to exhaust the sample air back into the hood. The sampling setup is connected to the hoods either by quick disconnect fire extinguisher outlets or through sealed bag openings.

Discs, 3/32 inch diameter, are punched out of the membrane filter for use in the electron microscope analyses. All air samples are counted to insure that the concentration on the small discs is less than 500 d/m. In order to prepare the sample, the membrane filter disc is placed on an S_iO substrate and the substrate is mounted on porous blocks in a petri dish. Acetone or ethylene acetate is placed in the bottom of the dish and the entire setup is covered with a watch glass. Over a period of several days, the membrane filter is dissolved leaving the particles in a relatively undisturbed position on the substrate. Electron micrographs are then taken of the particles.

The alpha concentrations observed with the hood air samples ranged from 6.5×10^{-10} μc/cc to 3.5×10^{-9} μc/cc.

Two of the hood samples were analyzed with the electron microscope. The data for hood samples was taken from electron microscope photographs magnified 20,000 times. The following equation was used to calculate the mean diameter:

$$\frac{\Sigma_i (N_i D_i)}{\Sigma N_i} = \text{mean diameter}$$

where N is the number particles of diameter D.

The data for these two filters are listed in Table I:

TABLE I

MEAN PARTICLE DIAMETERS

Sample No.	Total Part./specimen	Part./cc air	Part. Dia. Range	Mean Part. Diameters
1	165	1×10^5	0.025 u - 0.15 μ	0.046 μ
2	156	5×10^4	0.025 u x 0.1 μ	0.046 μ

The procedure should be improved in two ways before the previous data can be termed as conclusive. Alpha track labeling of these particles was not performed, therefore, the particles cannot be positively identified as plutonium. Particles as small as the ones found in these two specimens would have to be heavy metal to be opaque to the electron beam. It appears that there are particles smaller than the 0.025 μ sizes; however, an overall background density from the S_iO substrate prevents positive identification. Efforts are underway to improve or eliminate this background problem.

SUMMARY

Since the settling rate of particles is related to the particle diameter and mass, one would expect to see increasing numbers of particles in air as their aerodynamic diameter decreases. Air sample data analyzed by both light scattering and electron microscope techniques supports this theory. Consequently, for plutonium particles with their high density, the mean aerodynamic diameter is quite low compared to normally encountered dust particles. Improved electron microscope techniques may reveal airborne plutonium particles with a mean diameter significantly less than the 0.046 μ.

SLIDE 1

ROYCO COUNTER PARTICLE SIZE RANGES

Channel Number	Range-Microns
1	0.32 to 0.40
2	0.4 to 0.5
3	0.5 to 0.64
4	0.64 to 0.8
5	0.8 to 1.0
6	1.0 to 1.3
7	1.3 to 1.6
8	1.6 to 2.0
9	2.0 to 2.5
10	2.5 to 3.2
11	3.2 to 4.0
12	4.0 to 5.0
13	5.0 to 6.4
14	6.4 to 8.0
15	8.0 and Greater

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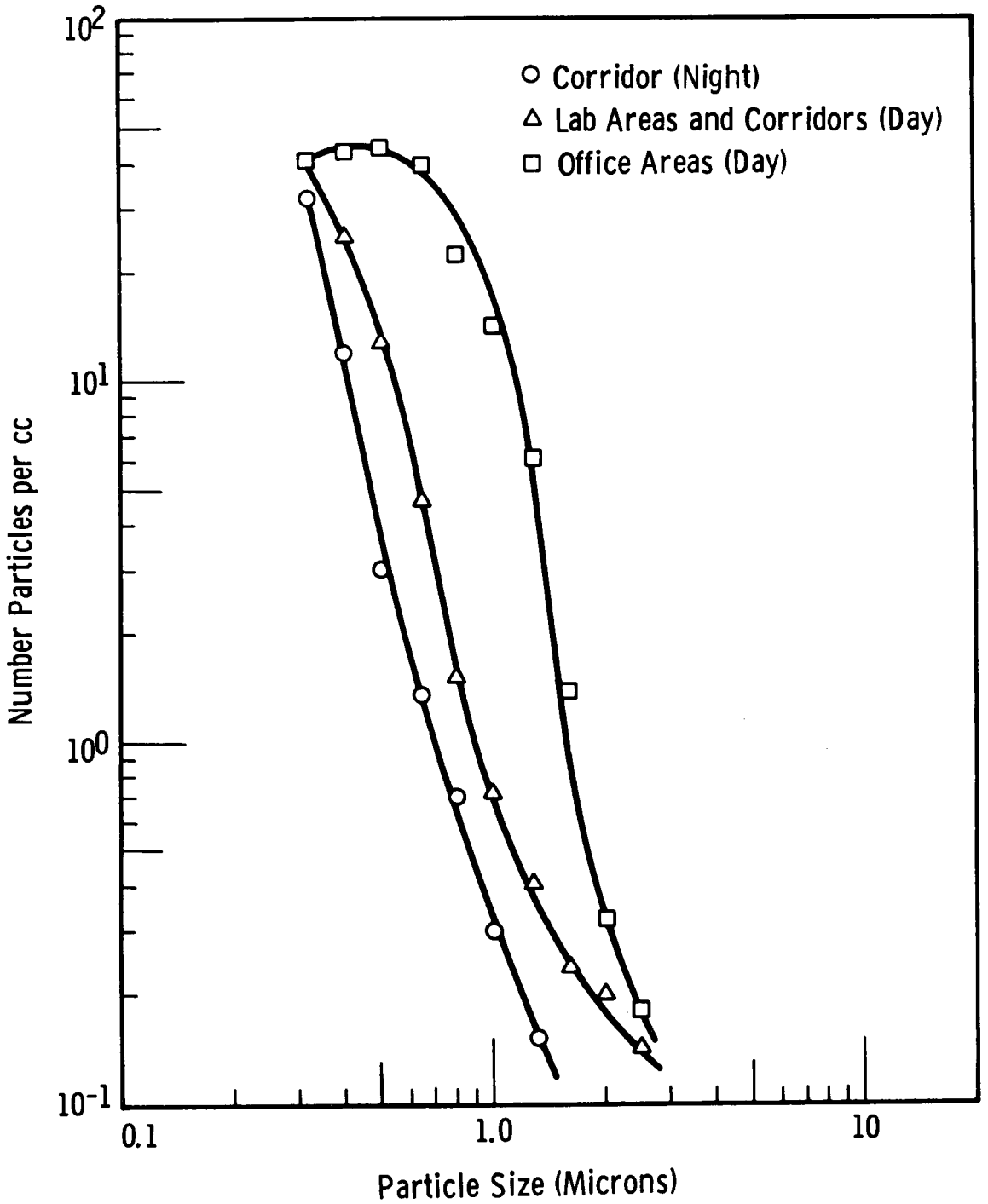
Slide 2

PARTICLE SIZE DISTRIBUTION 234-5 BUILDING



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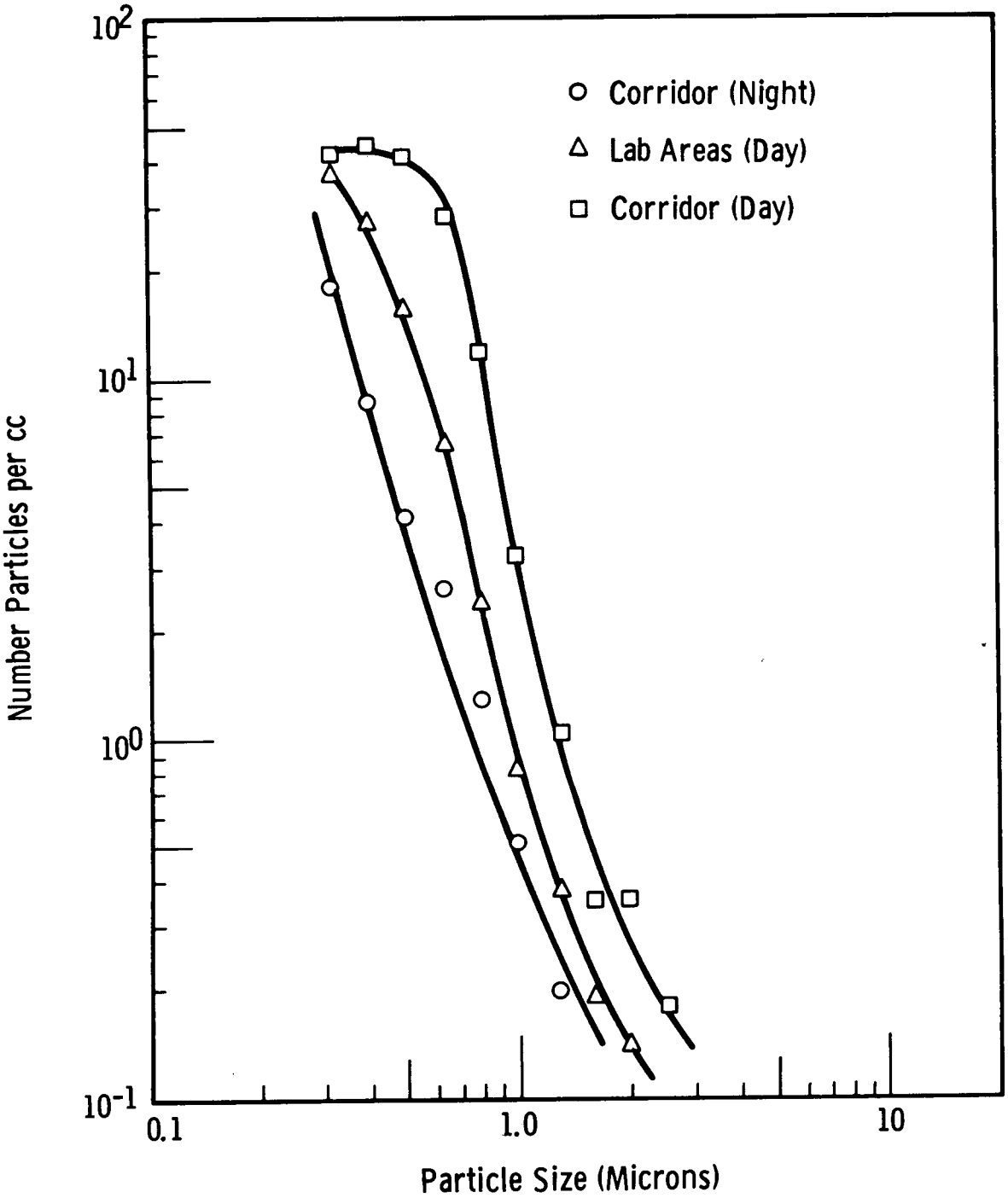
Slide 3

PARTICLE SIZE DISTRIBUTION 308 BUILDING



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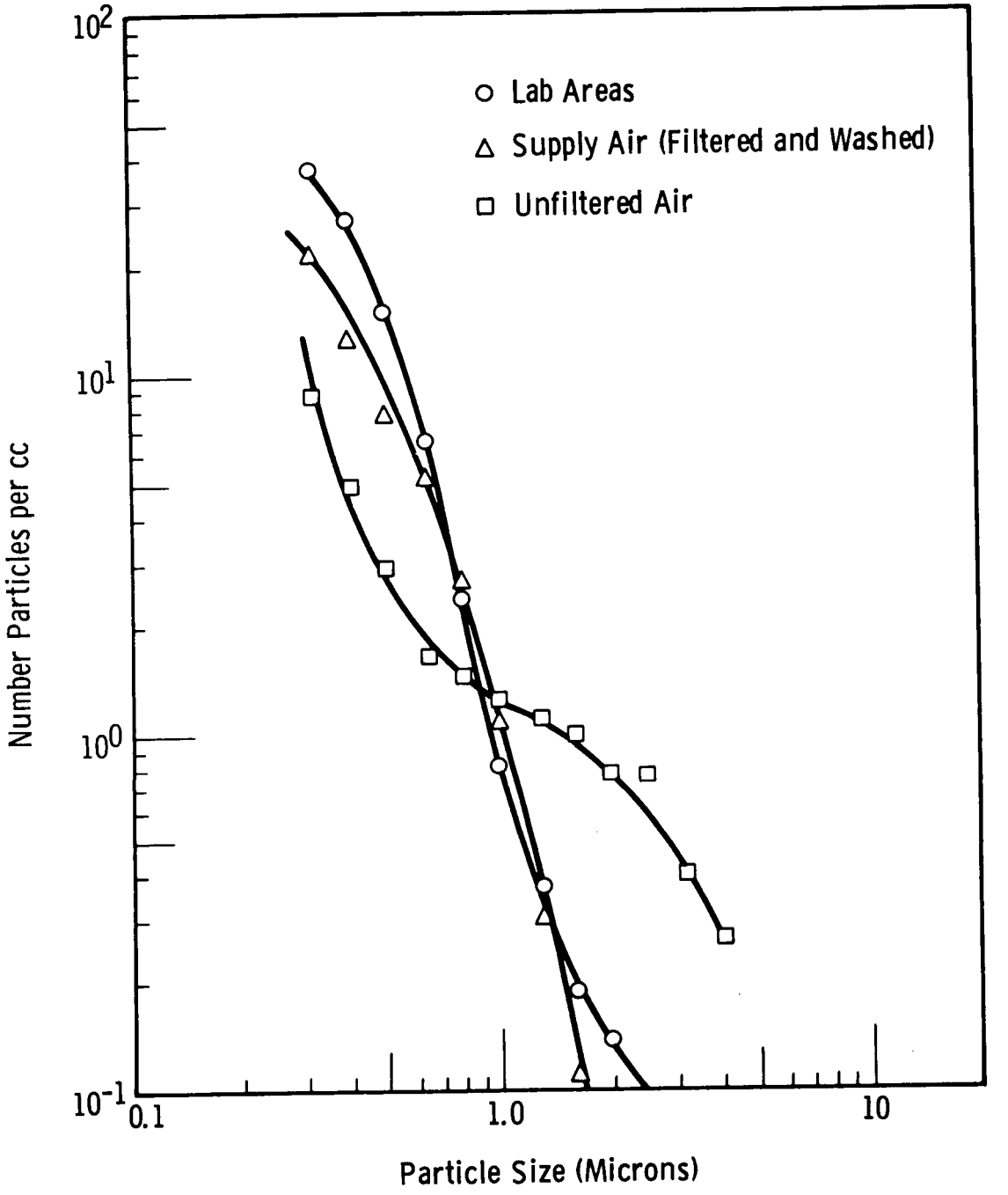
Slide 4

COMPARISON PARTICLE SIZE DISTRIBUTION TREATED AND UNTREATED AIR



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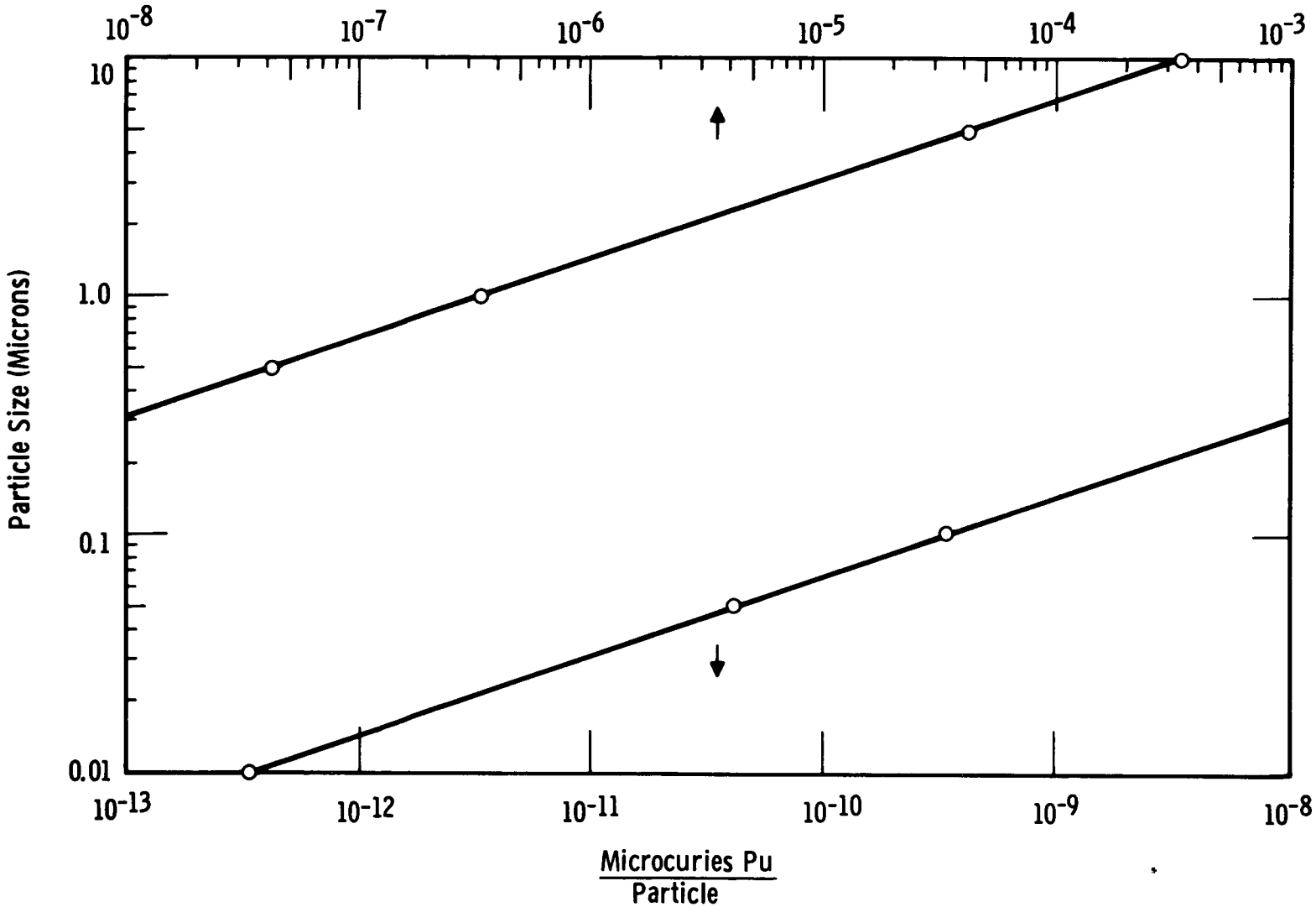
Slide 5

PuO_2 PARTICLE SIZE VS ACTIVITY/PARTICLE



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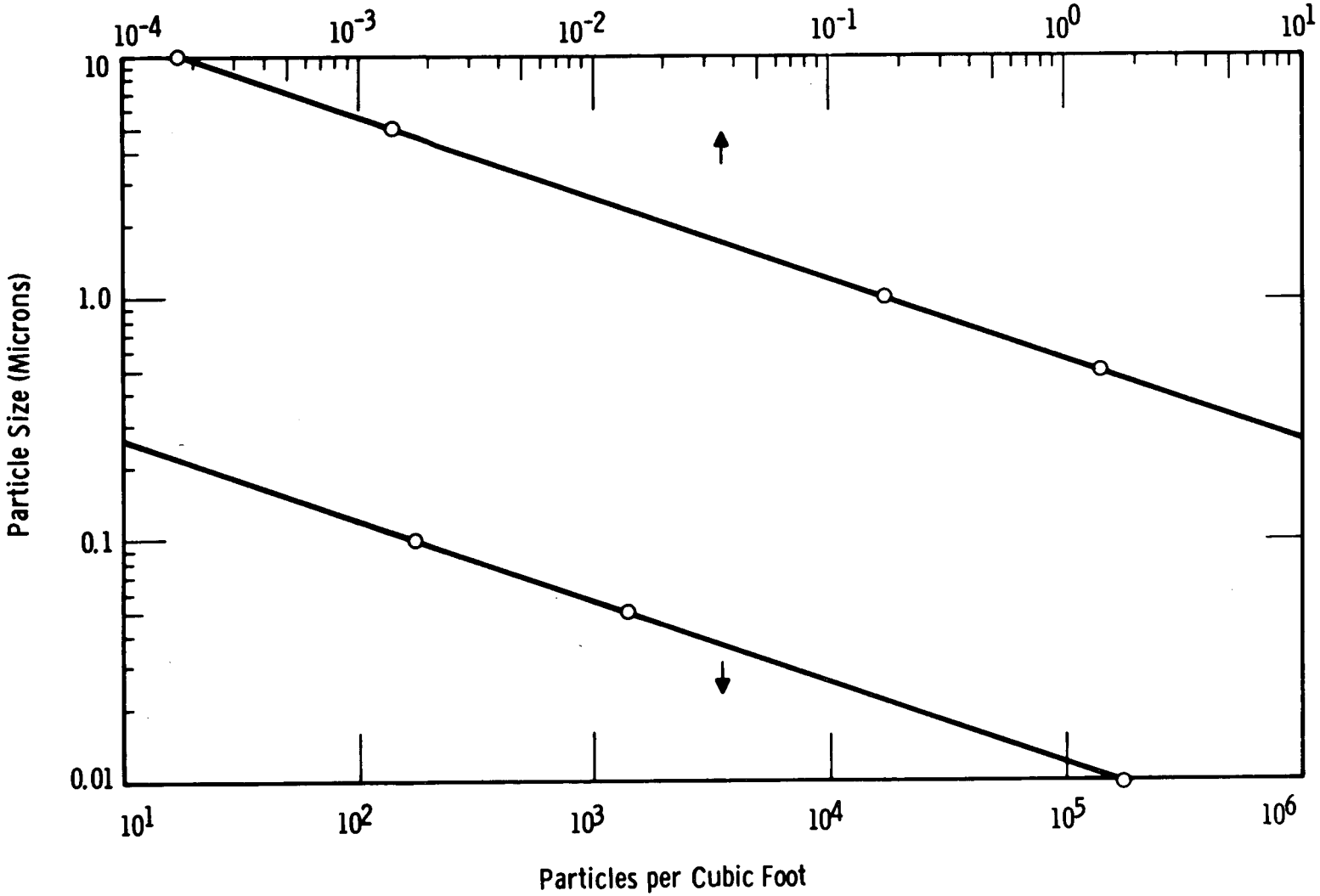
Slide 6

PuO_2 PARTICLE SIZE VS PARTICLES PER CUBIC FOOT FOR 1 MPC_A



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