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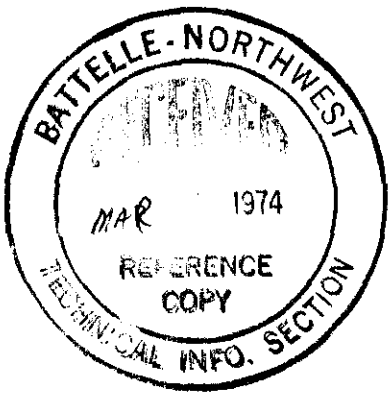

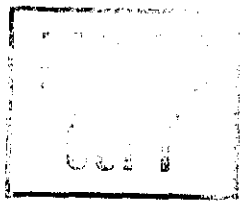

CHARACTERIZATION OF RADIOACTIVE PARTICLES IN THE 234-5Z BUILDING GASEOUS EFFLUENT

Battelle
Pacific Northwest Laboratories
Richland, Washington 99352

DECEMBER 1973

Prepared for the U.S. Atomic Energy Commission under Contract AT(45-1):1830

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CHARACTERIZATION OF RADIOACTIVE PARTICLES IN THE 234-5Z BUILDING GASEOUS EFFLUENT

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Particulate & Gaseous Waste Research Section
ATMOSPHERIC SCIENCES DEPARTMENT

DECEMBER 1973

Work Sponsored by ARHCO

Battelle, Pacific Northwest Laboratories
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ABSTRACT

Seven sets of large-volume air samples were taken from the main exhaust stack of the 234-5Z Building over a seven-month period. The concentration of alpha-emitters associated with particles was determined and the distribution of radioactivity as a function of particle size measured using a large-volume cascade impactor. The average concentration of alpha emitters over long periods of sampling was much less than the occupational MPC_a for ^{239}Pu. The acticity median aerodynamic diameter ranged from 3 to 9 μm. From 60 to 80 percent of the radioactive material was associated with particles less than 10 μm, aerodynamic equivalent diameter, which may be regarded as an upper limit for respirable particles. Detail characterization of the chemical and physical nature of these aerosols and the mechanisms for their generation require further study.

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CHARACTERIZATION OF RADIOACTIVE PARTICLES IN THE 234-5Z BUILDING GASEOUS EFFLUENT

INTRODUCTION

The Plutonium Finishing Plant at the Hanford site has a long history as a plutonium processing facility and has demonstrated that plutonium can be handled safely. The emission of radioactive materials is controlled well within established limits as evidenced by continuously monitoring of stack gases. But stewardship does not end with ascertaining that the radioactive material released is within established limits. An abiding concern for the possible effects of plutonium processing operations on the environs justifies a continuing effort to provide data that can be used to determine the fate of these radioactive particles after emission. Knowledge of the distribution of the activity in aerodynamic particle size fractions is necessary to determine the airborne behavior of emitted materials in the atmosphere, and their ultimate pathway to man.

A preliminary study was conducted earlier to indicate the concentrations and size distributions of radioactive particles in the 234-5Z Building exhaust system.[1] In this study filter and cascade impactor samples were taken of the 234-5Z Building stack gases and of various exhaust streams within the 234-5Z exhaust systems. The overall particle removal efficiency of the exhaust system filters was excellent. Few, if any, of the alpha-active particles emitted from the stack are recycled back into the ventilation intake. The small quantity of plutonium present appeared to be attached to larger, non-active particles. Because of the limited number of samples, (generally only a single sample at most locations) the findings were considered indicative rather than definitive.

OBJECTIVE

The present study was undertaken to determine the concentration and distribution within aerodynamic size fractions of plutonium-bearing particles emitted from the 234-5Z stack.

SUMMARY AND CONCLUSIONS

The quantity of alpha emitters as particles and their distribution into various aerodynamic size fractions was measured on seven sets of high volume samples extracted from the 234-5Z Building stack.* Sampling was initiated on October 12, 1972 and completed on May 31, 1973. Each set of samples was composed of an 8-inch by 10-inch glass fiber filter and collectors from a four-stage cascade impactor. Sample rate through the filter was 50 cfm and sample rate through the cascade impactor was 20 cfm. Total exhaust volume sampled ranged from 5.48×10^5 to 2.65×10^6 cu. ft. Plutonium content of individual samples was determined by dissolution of the glass fiber filter or impactor collectors and alpha counting.

The concentration of alpha activity measured was low -- less than the ^{239}Pu MPC_{air} for occupational exposure in all 14 samples. The values measured are higher than the concentrations measured in the preliminary study[1] and calculated from ARHCO Radiation Monitoring daily stack samples.[2] They do not appear to correlate with the nature of operations within the building during the sampling period.

As in the preliminary study, the activity appears to be distributed on larger particles than might be anticipated from the history of the exhaust gases--all gases are passed through from one to three stages of HEPA filters. The activity median aerodynamic diameters measured ranged from 3.3 to 9.0 μm with 60 to 80 percent of the activity associated with particles less than 10 μm aerodynamic equivalent diameter.

The downwind airborne concentration will be of the order of 2×10^{-13} to 2×10^{-14} μC_i/cu. cm., at the point of maximum concentration, under conservative assumptions -- a mean wind speed of 1 meter per sec. and a point rather than volume source. In as much as the exhaust is already less than the MPC_a for occupational exposure as it leaves the stack, downwind airborne concentrations should not constitute an inhalation hazard.

---

* We assume for the purpose of this study that all alpha-emitters can be regarded as ^{239}Pu. In some cases it became necessary to process samples chemically which may have removed americium. The assumption that all emitters are ^{239}Pu will not significantly change any conclusions reached from the study.

EXPERIMENTAL

234-5Z Building Ventilation and Exhaust System

The Building ventilation and exhaust system is composed of several subsystems. Air is supplied to all of the 234-5Z, 236-Z and 242-Z Buildings by eight fans located on the second floor of the 234-5Z Building. The 233-Z Building has its own air supply. Air from uncontaminated areas -- offices, shop areas, etc. -- that are separated from process areas by air locks is exhausted through the roof at several points with no filtration.

Exhaust from processing areas and equipment is filtered through from one to three banks of HEPA filters, combined in the 291-Z Fan House and released via the 234-5Z stack. The filtered exhaust system is diagrammed in Figure 1.

Sampling

Continuous high flow filter and cascade impactor samples were taken of the radioactive particles in the 234-5Z Building stack. Samples of the stack gases were extracted at the base of the 16 ft. diameter by 200 ft. steel lined-concrete stack (see Figure 2). The sampling arrangement used is shown in Figure 3.

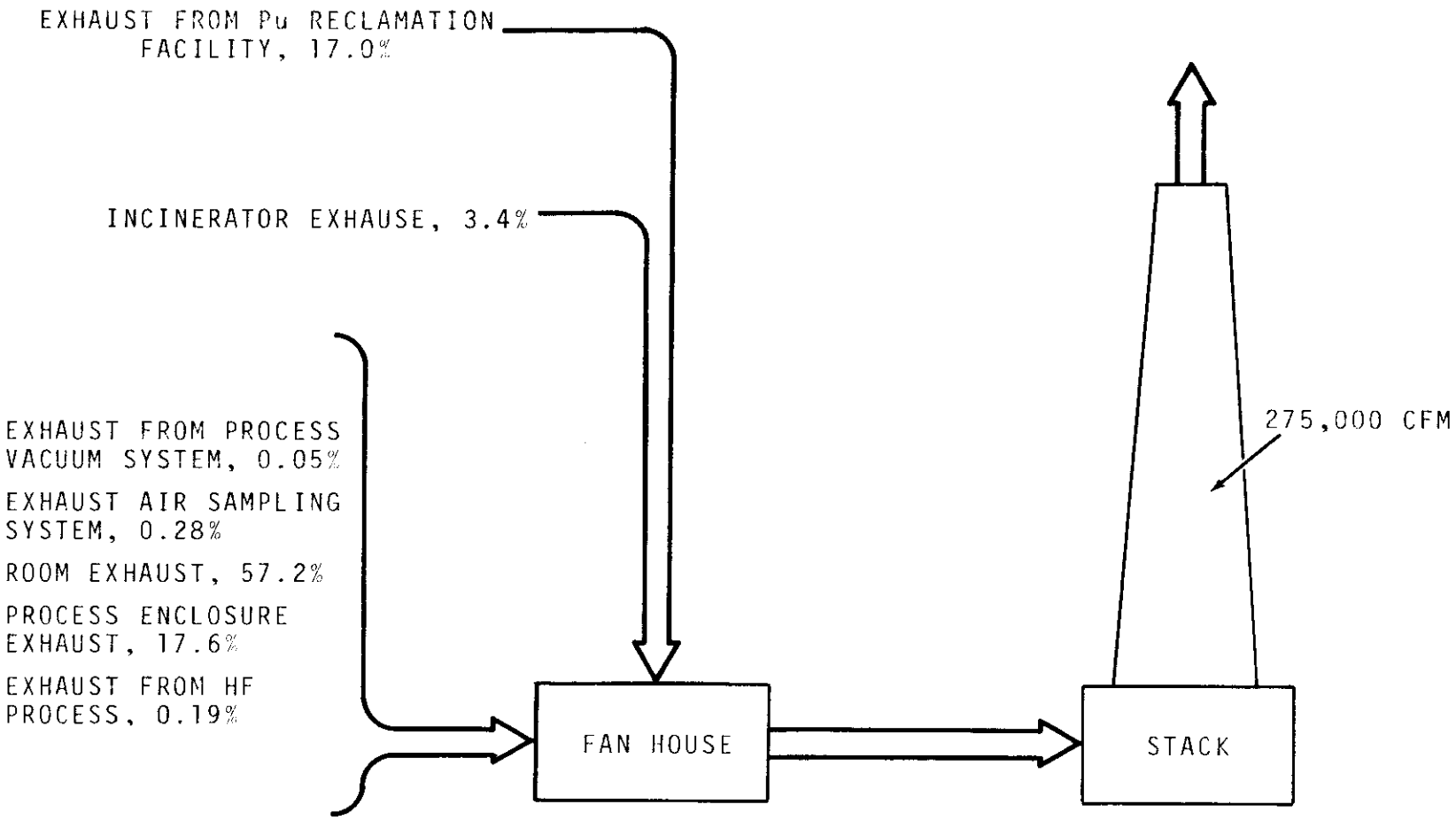
Samples for the 8 in. × 10 in. glass filter sampler were extracted via a 1 in. diameter sch. 10 pipe extending 5 ft. into the stack. To more nearly achieve isokinetic flow, the inlet to the probe was enlarged to 2.5 in. diameter. The opening of the probe was aligned with gas flow in the stack. Flow through the sampler ranged from 40.2 to 60.3 cu. ft./min., resulting in nominal sampling velocities in the range of 1180 to 1270 ft. per min. Nominal stack gas flow was approximately 1370 ft. per min. Sampling rates were monitored periodically by connecting a turbine type anemometer* between the sampler and blower. The filter sampler is shown in Figure 4. The anemometer arrangement is shown in Figure 5 attached to the High Vol cascade impactor. Exhaust from the blower was returned to the stack.

A similar arrangement was utilized for the high volume cascade impactor.**

---

* Sci-Med model Pk-3, Sci-Med, 13010 County Road 6, Minneapolis, Min. 55441

** A four stage hi-volume cascade impactor, 2000 Inc. model 65-000, P.O. Box 20803, Atlanta, Ga., 30320.



BASED ON ESTIMATED FLOW

FIGURE 1

PLUTONIUM FINISHING FACILITY GASEOUS EFFLUENT

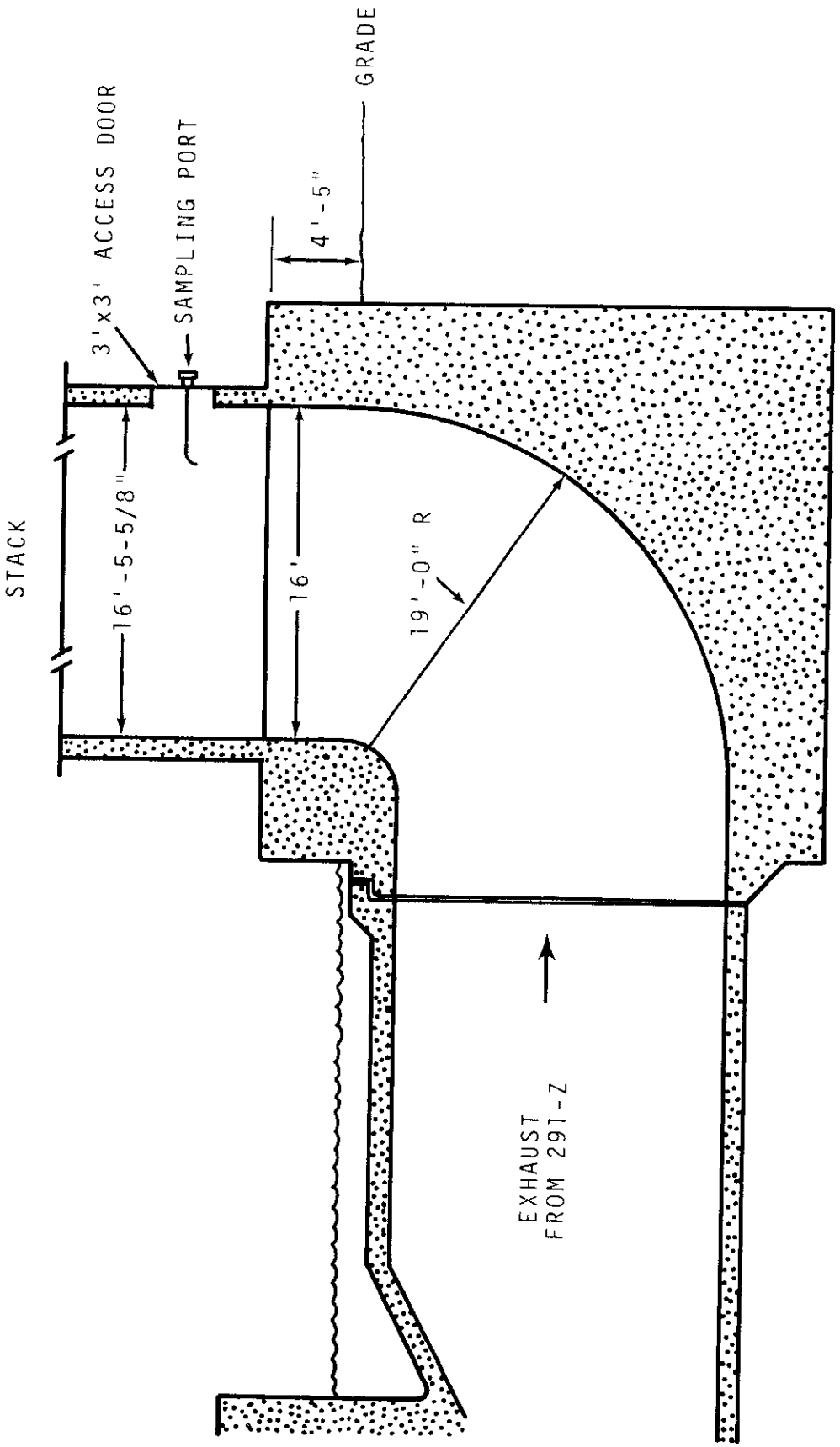


FIGURE 2

STACK SAMPLING LOCATION

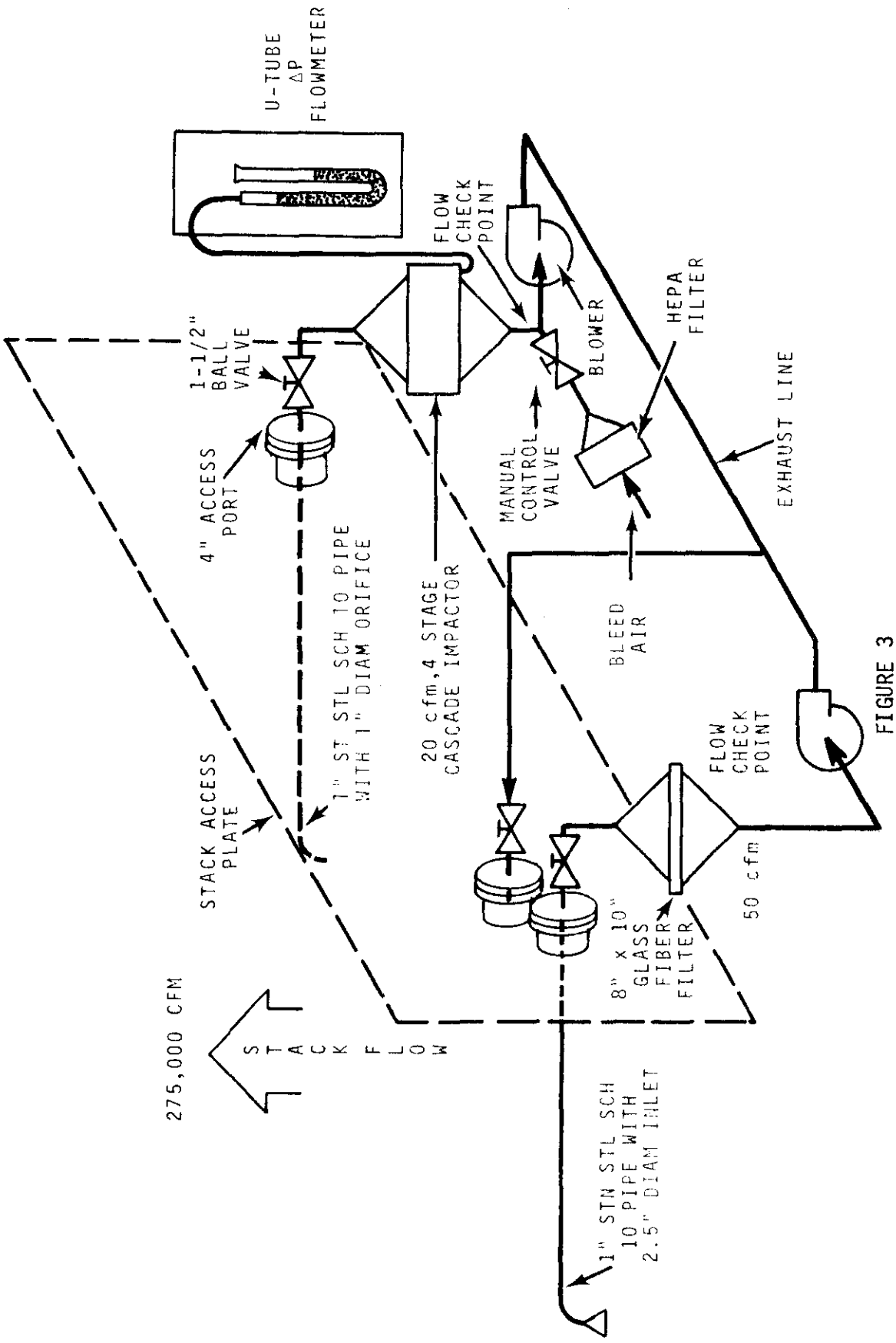


FIGURE 3

STACK SAMPLING SYSTEM

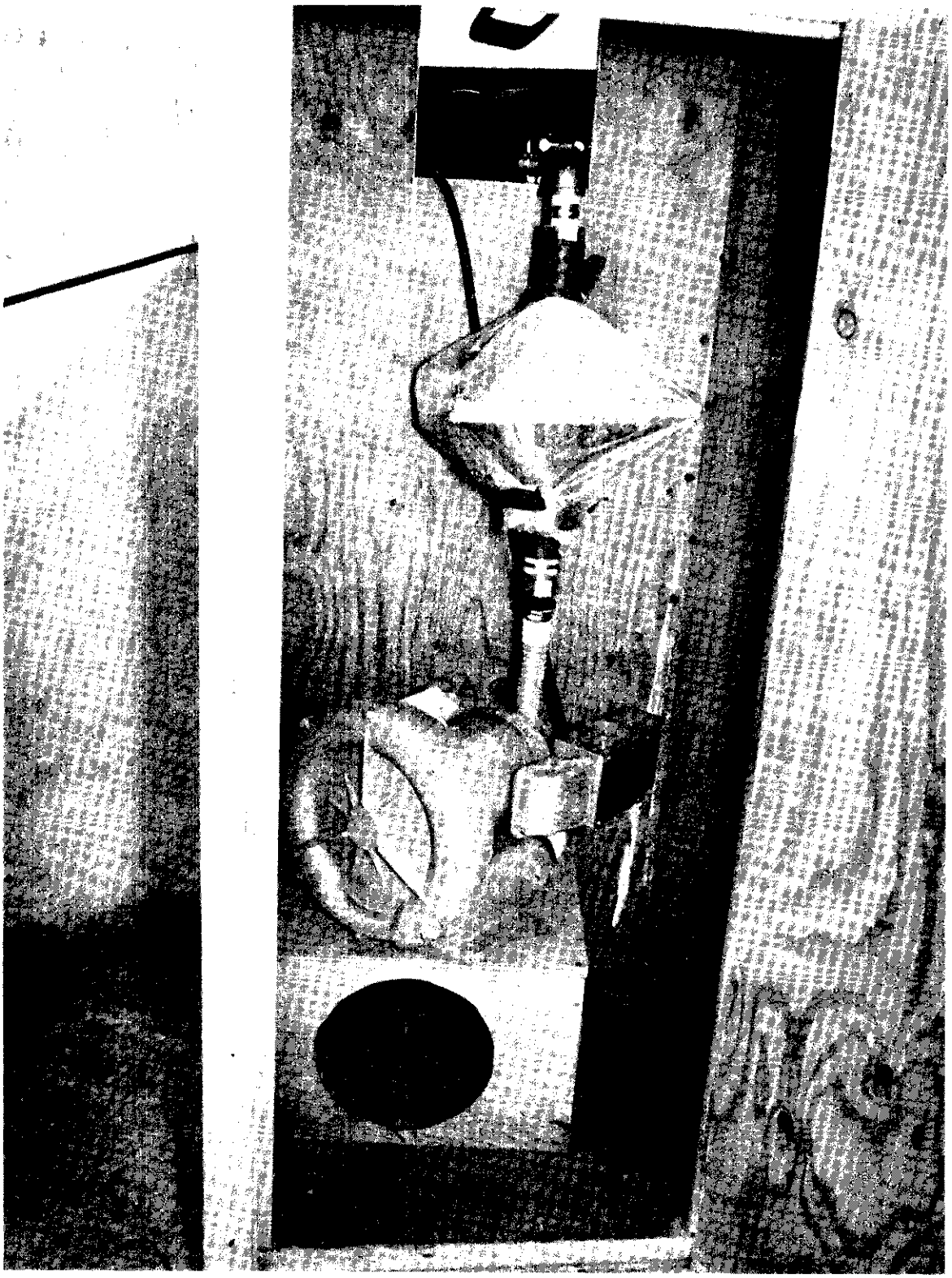


FIGURE 4

234-5Z STACK FILTER SAMPLER

Neg. No. PNL 732989-4

FIGURE 5

SAMPLING FLOW CHECK USING THE TURBINE-TYPE ANEMOMETER

Neg. No. PNL 732989-2

Samples were extracted from the stack using a 1 in. diameter, sch. 10 pipe extending 5 ft. into the stack. The 1 in. diameter opening was aligned with the stack flow. Sampling rates ranged from 17.8 to 22.0 cfm indicating nominal velocities at the probe orifice of approximately 3000 ft./min. Inlet velocity was thus about twice isokinetic, favoring somewhat the preferential sampling of small particles. The sampler is shown in Figure 6.

Sample Preparation and Analysis

All materials collected were placed into solution by mixed acid leach. The 8 in. by 10 in. glass fiber filter and sampler recovered on May 31, 1973, are shown in Figure 7, and are typical of the appearance of this type of sample. Due to the deflector in the inlet, the rust-colored deposit tended to collect around the edges.* The inlet was wiped with 6 N nitric acid soaked tissues which were added to the glass fiber filter sample. A double filter was used starting with the 4th sample.

The collection filters and high volume cascade impactor for the same period are shown in Figures 8a, 8b, 8c, 8d and 8e. Visible quantities of material are present on all filters but the quantity did not appear to be indicative of the alpha emitters present -- the plutonium equivalent detected was: top plate of impactor - 9600 dpm, 1st stage - 4000 dpm, 2nd stage - 1480 dpm, 3rd stage - 760 dpm and 4th stage - 460 dpm. Acid soaked wipes were used to remove the material deposited on the inlet transition piece and under side of the individual stage collector support plates. The wipes were added to the collector of the next stage.

All glass fiber filters were muffled for 1 hour at 550°F. Preliminary tests performed using this media indicated retention of the activity in an oily material during mixed acid leach. Heating for 1 hour at 550°F eliminated the oily residue. Thirty percent hydrogen peroxide was also added dropwise to the warm acid solution to destroy any residual organic. The solution was taken to dryness twice using hydrofluoric acid to remove silica and twice with nitric acide to remove residual fluorides which interfere with the ion exchange procedure required by the high salt content. Standard recoveries ranged from 85 to 90 percent, normal for this type of procedure.

---

* The deflector was required to prevent the jetting action of the sample stream from disturbing the filter media.

FIGURE 6

HI-VOLUME CASCADE IMPACTOR ON 234-5Z STACK

Neg. No. PNL 732989-1

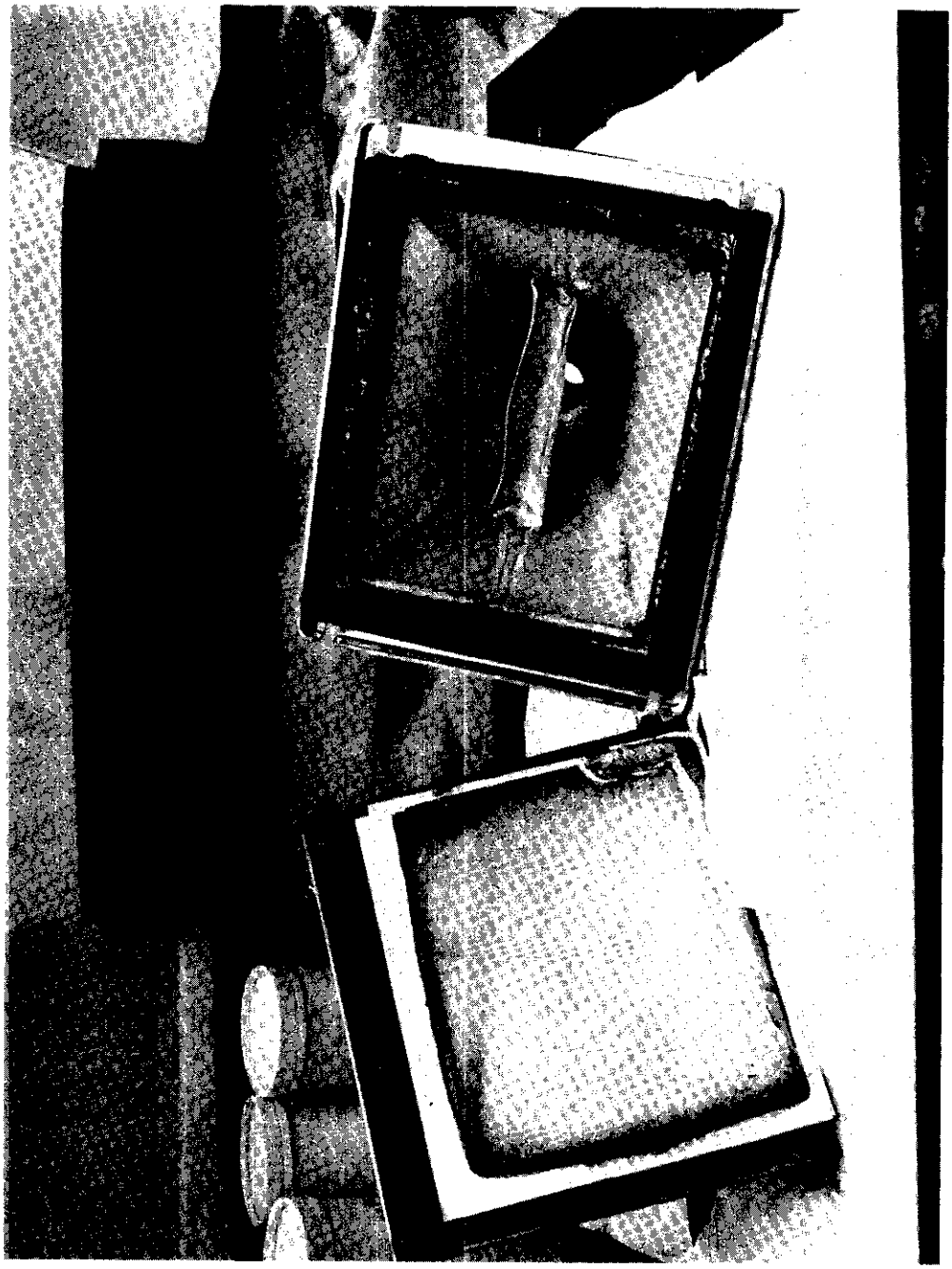


FIGURE 7

8" × 10" GLASS FIBER FILTER AND HOLDER FOR PERIOD
5/2/73 to 5/31/73

Neg. No. PNL 732989-9

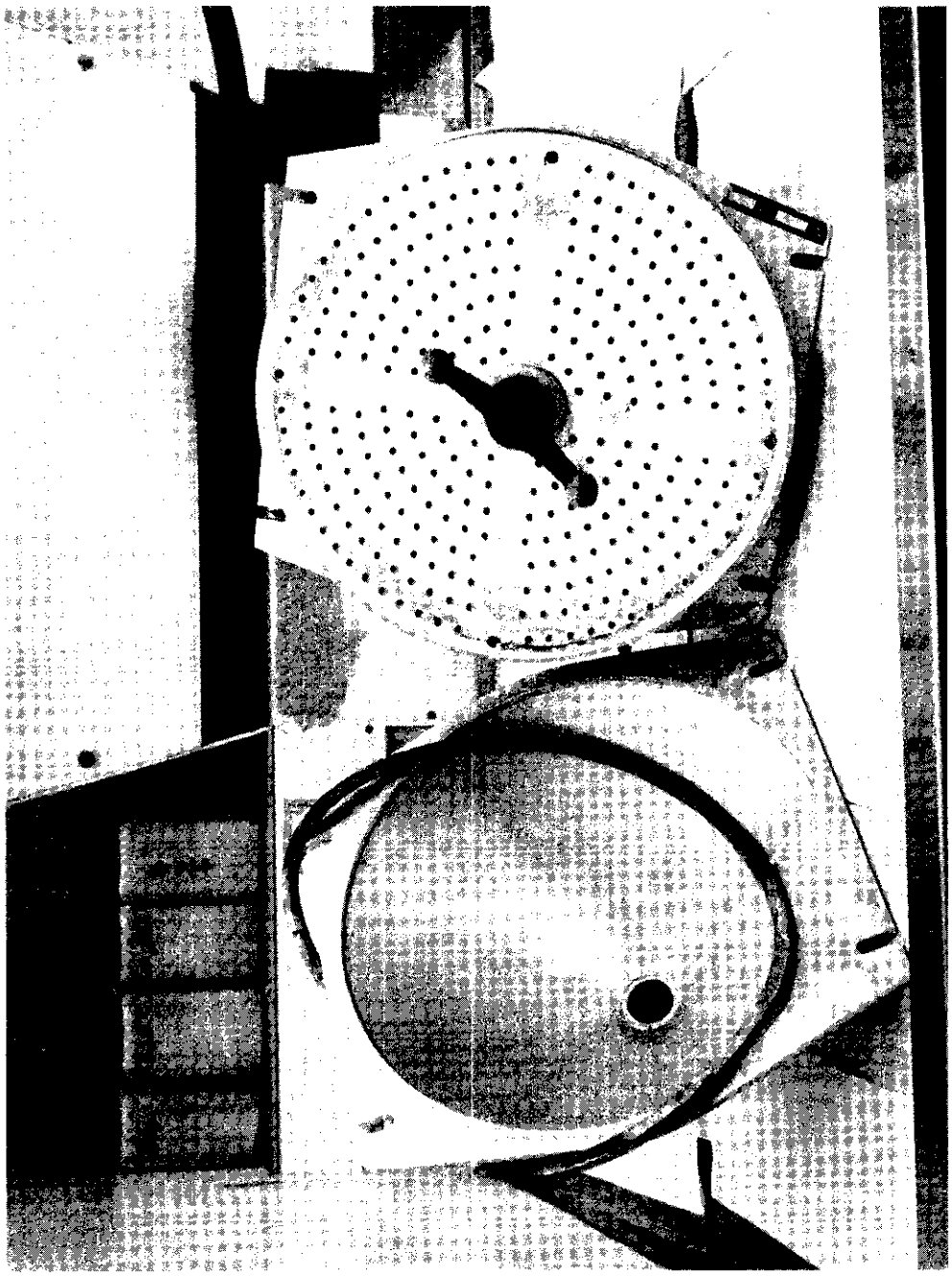


FIGURE 8a

HI-VOLUME CASCADE IMPACTOR AND INLET TRANSITION
FOR PERIOD 5/2/73 to 5/31/73

Neg. No. PNL 732989-3

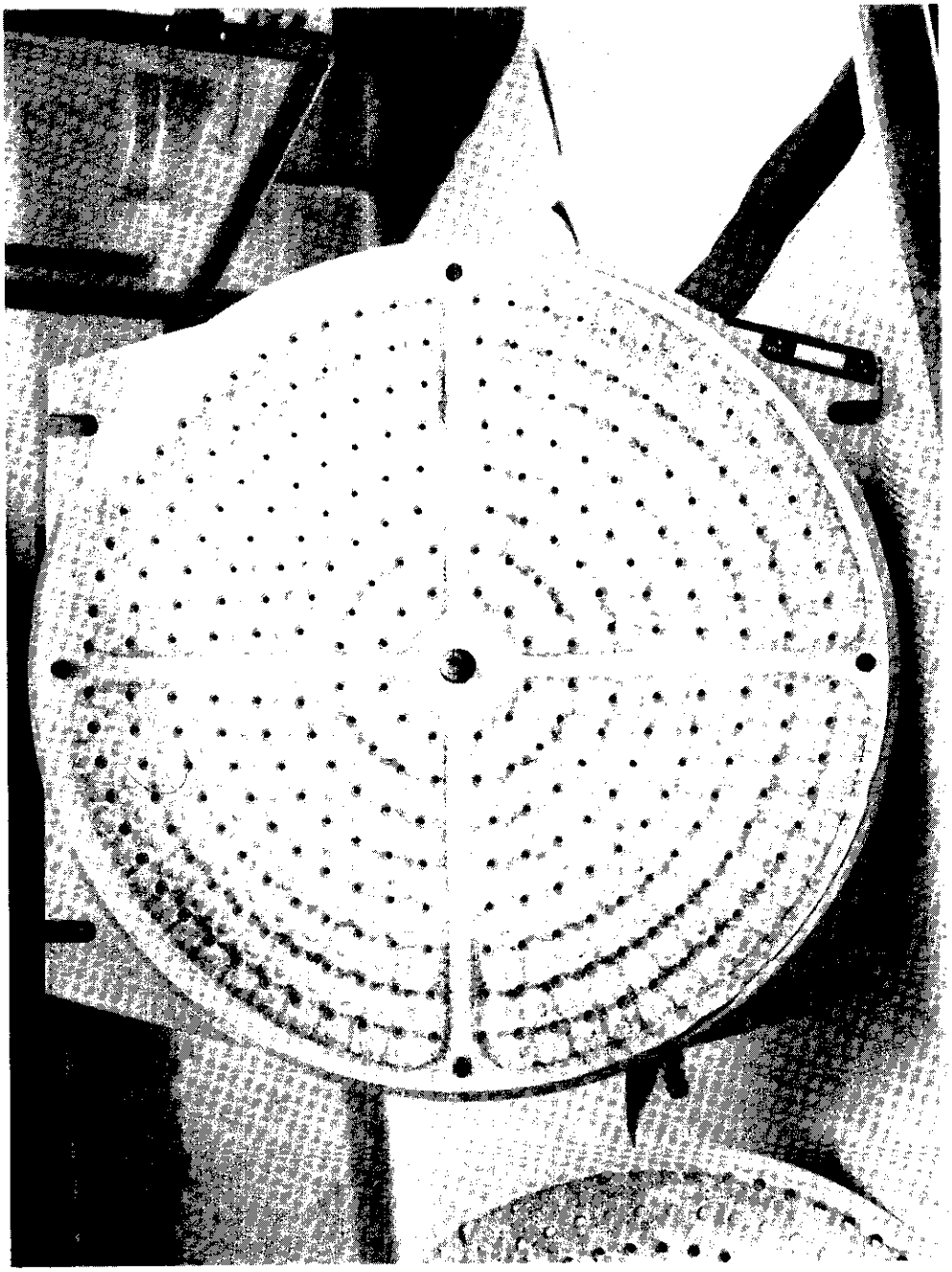


FIGURE 8b

1st STAGE COLLECTION FILTER FOR 5/2/73 to 5/31/73

Neg. No. PNL 732989-10

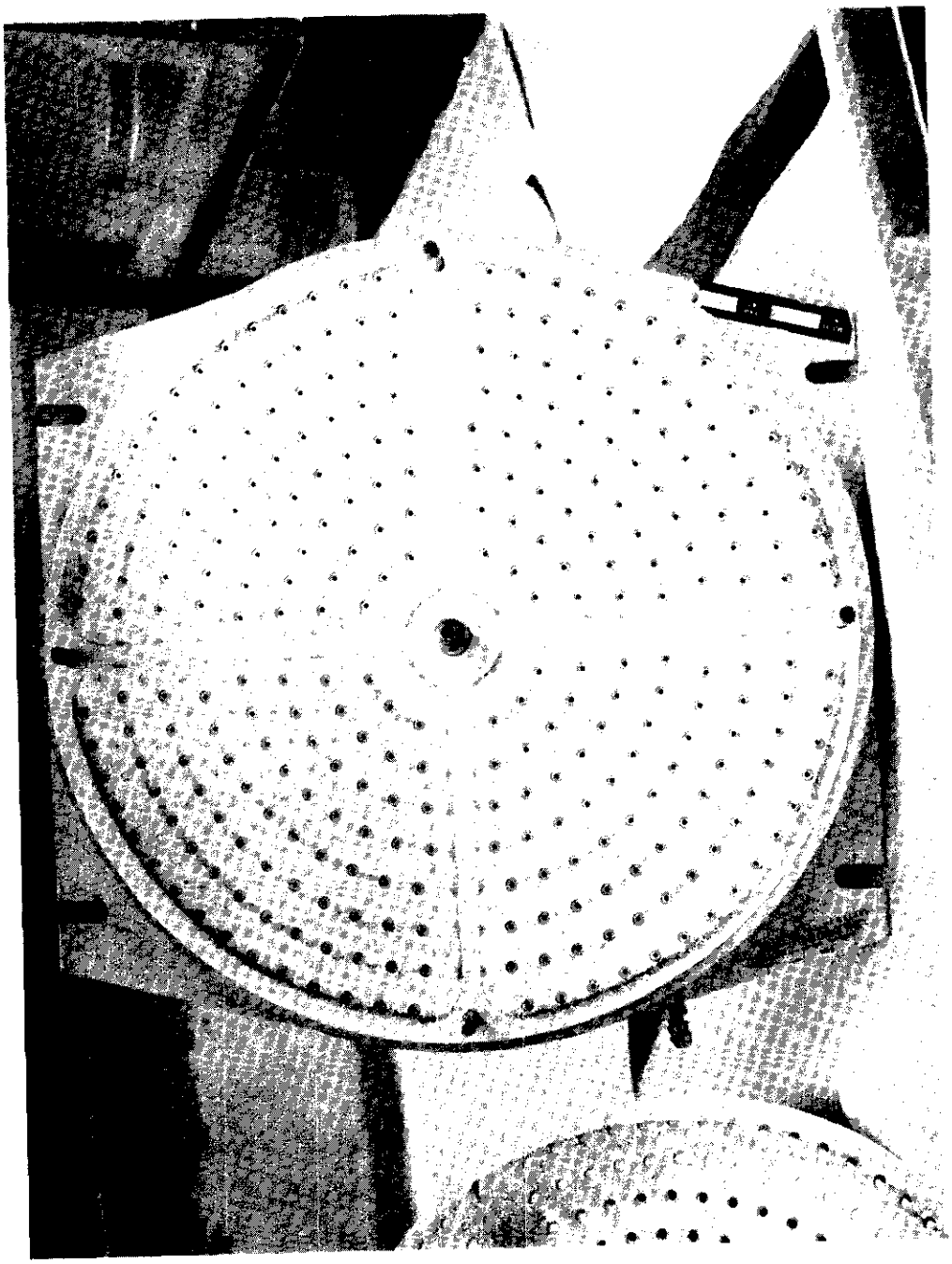


FIGURE 8c

2nd STAGE COLLECTION FILTER FOR 5/2/73 TO 5/31/73

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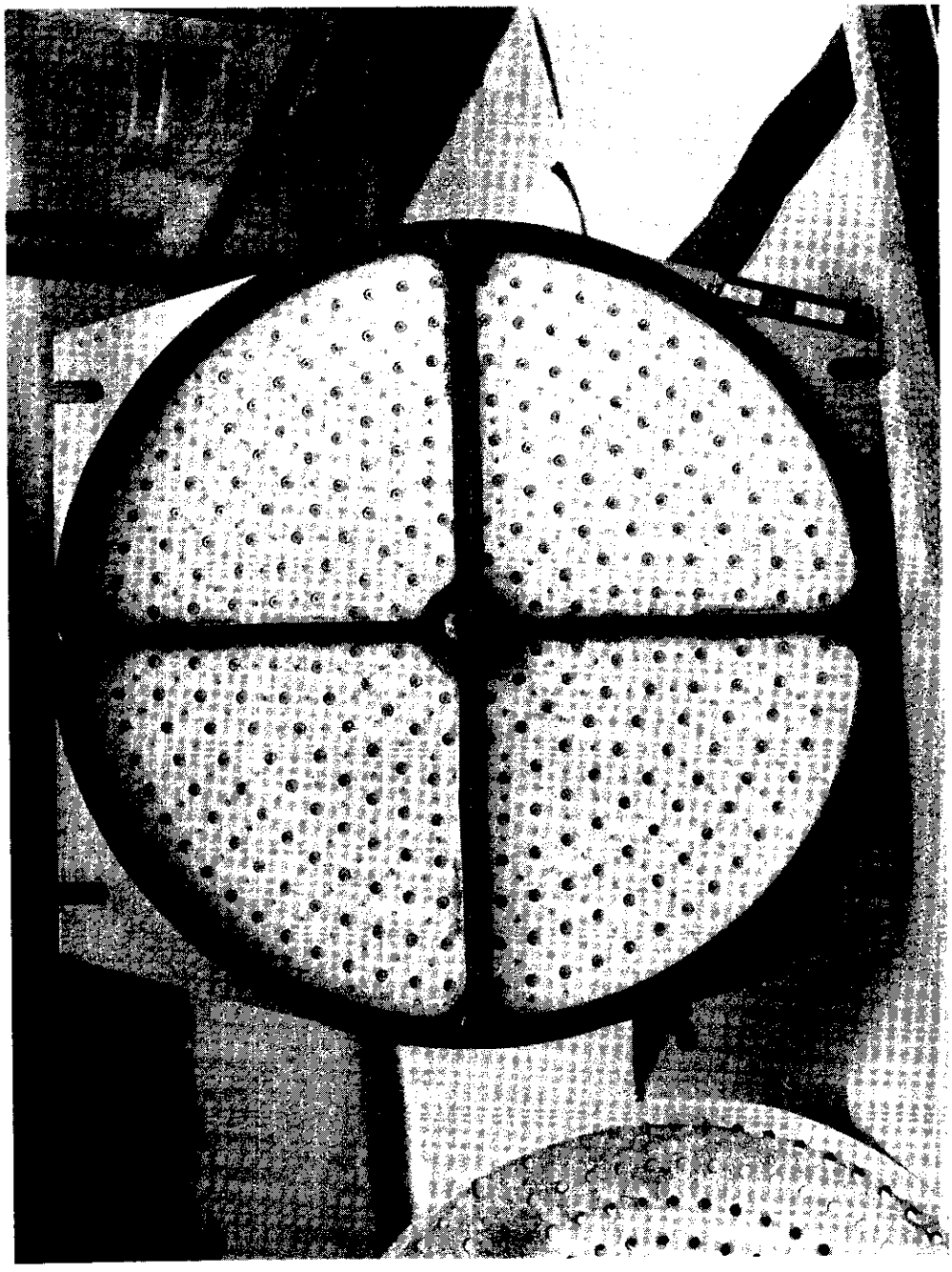

FIGURE 8d

3rd STAGE COLLECTION FILTER FOR 5/2/73 TO 5/31/73

Neg. No. PNL 732989-7

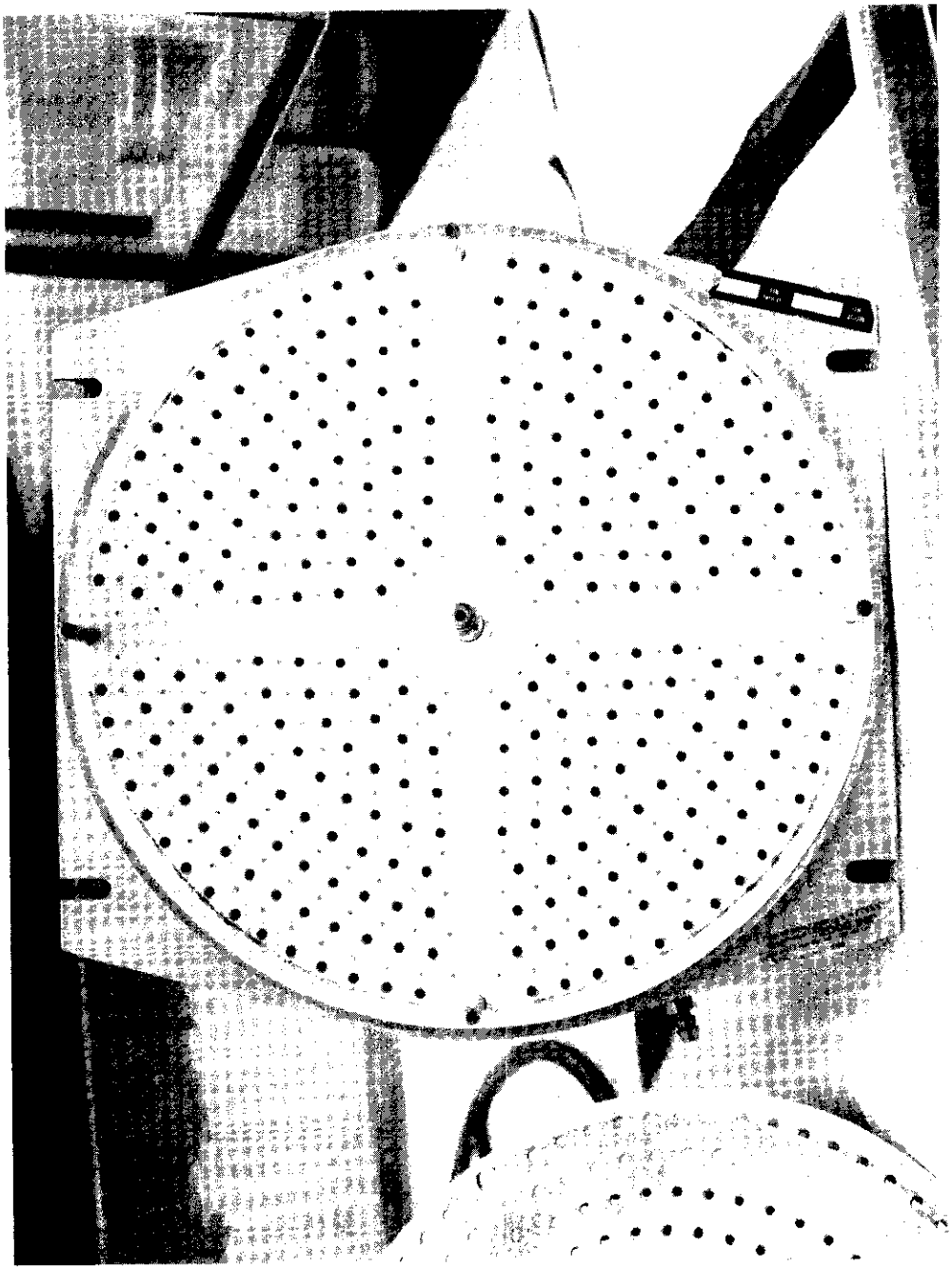

FIGURE 8e

4th STAGE COLLECTION FILTER FOR 5/2/73 to 5/31/73

Neg. No. PNL 732989-6

RESULTS

The quantity of alpha emitter radioactivity in the various samples collected is tabulated in Table I. All samples are appreciably above the lower detection levels of the analytical procedures. The alpha emitter radioactivity measured in the 8 in. × 10 in. glass fiber filter for the period Nov. 27, 1972 to Dec. 28, 1972, is low and is probably due to a torn filter. With the exception of the initial sample which is high (and could be due to an accumulation of materials in the probe prior to initiation of sampling), the remaining activity levels measured are relatively consistent and some correlation exists between activity concentrations measured by the filter results and calculated by summation of all activity in the cascade impactor.

The activity distribution into aerodynamic size fractions indicates a rather coarse aerosol. The activity median diameter (A.M.D.) measured ranged from 3.3 to 9.0 μm aerodynamic equivalent diameter (A.E.D.), see Table II and Figure 9. A significant portion of the activity entering the impactor (28 to 56 percent) was deposited in the inlet transition piece and on the inlet plate. Since the larger particles tend to deposit out more readily at these low velocities, the actual distribution may be even more coarse than measured. Extrapolation of the size distribution curves plotted indicates from 60 to 80 percent of the radioactivity in the gaseous effluents is associated with particles less than 10 μm A.E.D.

DISCUSSION

The airborne concentration of activity in the 234-5Z Building gaseous effluent over the period October 12, 1972 to May 31, 1973, is low. The values measured from 0.013 to 0.041 dpm per cu. ft. with an average of 0.034 dpm per cu. ft. for the sampling period (see Table III). All values are less than the MPC_{air} 40 hour week occupational exposure -- 2×10^{-12} μCi per cu. cm. ($\times$ 28320 cu. cm. per cu. ft. $\times$ 2.2×10^{6} dpm per μCi = 0.13 dpm per cu. ft.). These values indicate an activity concentration from 3 to 10 times greater during this period than measured during the preliminary study.[1]

The activity measured in the stack does not appear to exhibit a close correlation to plant operations. Processing lines were in operation for short periods during 5 of the 7 sampling intervals. The alpha activity concentra-

TABLE I

CHARACTERIZATION OF RADIOACTIVE PARTICLES IN THE 324-50 BUILDING GASEOUS EFFLUENTS

Collection Interval →	10-12-72 to 11-27-72	11-27-72 to 12-29-72	12-21-72 to 1-16-73	1-16-73 to 3-6-73	3-6-73 to 4-3-73	4-3-73 to 5-1-73	5-2-73 to 5-31-73
FILTER -- 8in. x 10 in. glass fiber							
Collection period, hr	786.7	716.2	671.1	932.9	427.9	660.7	651.8
Average sampling rate, cu. ft. per min.	56.2	54.5	52.5	49.5	47.7	51.9	44.7
Total volume sampled, cu.ft.	2.65×10^6	2.34×10^6	2.11×10^6	2.72×10^6	1.22×10^6	2.06×10^6	1.75×10^6
Alpha emitter activity, dpm	2.6×10^5	2.5×10^4	5.1×10^4	6.0×10^4	5.0×10^4	4.5×10^4	3.3×10^4
CASCADE IMPACTOR -- 12-1/4 in. glass fiber paper collectors and backup filter							
Collection period, hrs.	786.7	716.3	671.1	933.0	599.6	477.8	694.6
Average sampling rate, cu. ft. per min.	20.0	20.0	20.0	20.0	20.0	20.4	20.0
Total volume sampled, cu.ft.	9.42×10^5	8.61×10^5	8.06×10^5	1.11×10^6	7.20×10^5	5.84×10^5	8.33×10^5
Alpha emitter activity,							
Top of impactor, dpm	2.8×10^4	4800	7080	5048	11580	7230	9600
1st stage, dpm	1.6×10^4	3700	4150	4474	5200	2290	4000
2nd stage, dpm	1.5×10^4	1600	1045	2918	3060	1510	1480
3rd stage, dpm	5500	680	676	2176	1120	250	760
4th stage, dpm	1500	200	2012	970	520	130	460
filter, dpm	1500	490	1944	2176	300	160	440
Total alpha emitter activity, impactor, dpm	67500	71470	16907	17762	21780	4340	16700
Total alpha emitter activity, in impactor, dpm	39500	6670	9827	12714	10200	2050	7340

TABLE II

ACTIVITY DISTRIBUTIONS OF ALPHA EMITTERS IN 234-5Z BUILDING GASEOUS EFFLUENTS

COLLECTION INTERVAL →	10-12-72 to 11-27-72	11-27-72 to 12-28-72	12-28-72 to 1-6-73	1-16-73 to 3-6-73	3-6-73 to 4-3-73	4-3-73 to 5-1-73	5-2-73 to 5-31-73
Collection Period, hrs.	786.7	716.3	671.1	933.0	599.6	477.8	694.6
Total Alpha activity measured in cascade impactor, dpm	67500	11400	16907	17762	21780	4340	16700
Alpha Activity on stage collectors and filter, dpm	39500	6670	9827	12714	10200	2050	7240
Percent activity							
*1st stage > 7	40.5	55.5	42.2	35.2	51.0	52.7	56.0
*2nd stage 7.0 μm	38.0	24.0	10.6	23.0	30.0	34.8	20.7
*3rd stage 3.3 μm	13.9	10.2	6.9	17.1	11.0	5.8	10.6
*4th stage 20 μm	3.8	3.0	20.5	7.6	5.1	3.0	6.5
*Filter < 1.1 μm	3.8	7.3	19.8	17.1	2.9	3.7	6.2
Activity Median Diameter, μm	7.0	9.0	3.3	4.0	7.0	7.3	8.0
Percent Activity less than 10 μm	83	63	61	80	63	68	57
Percent of total activity collected in cascade impactor (other than that on the inlet transition section and the first stage orifice plate.)	58.5	58.5	58.1	71.6	46.8	47.2	44.0

* Percent activity on stage collectors and filter.

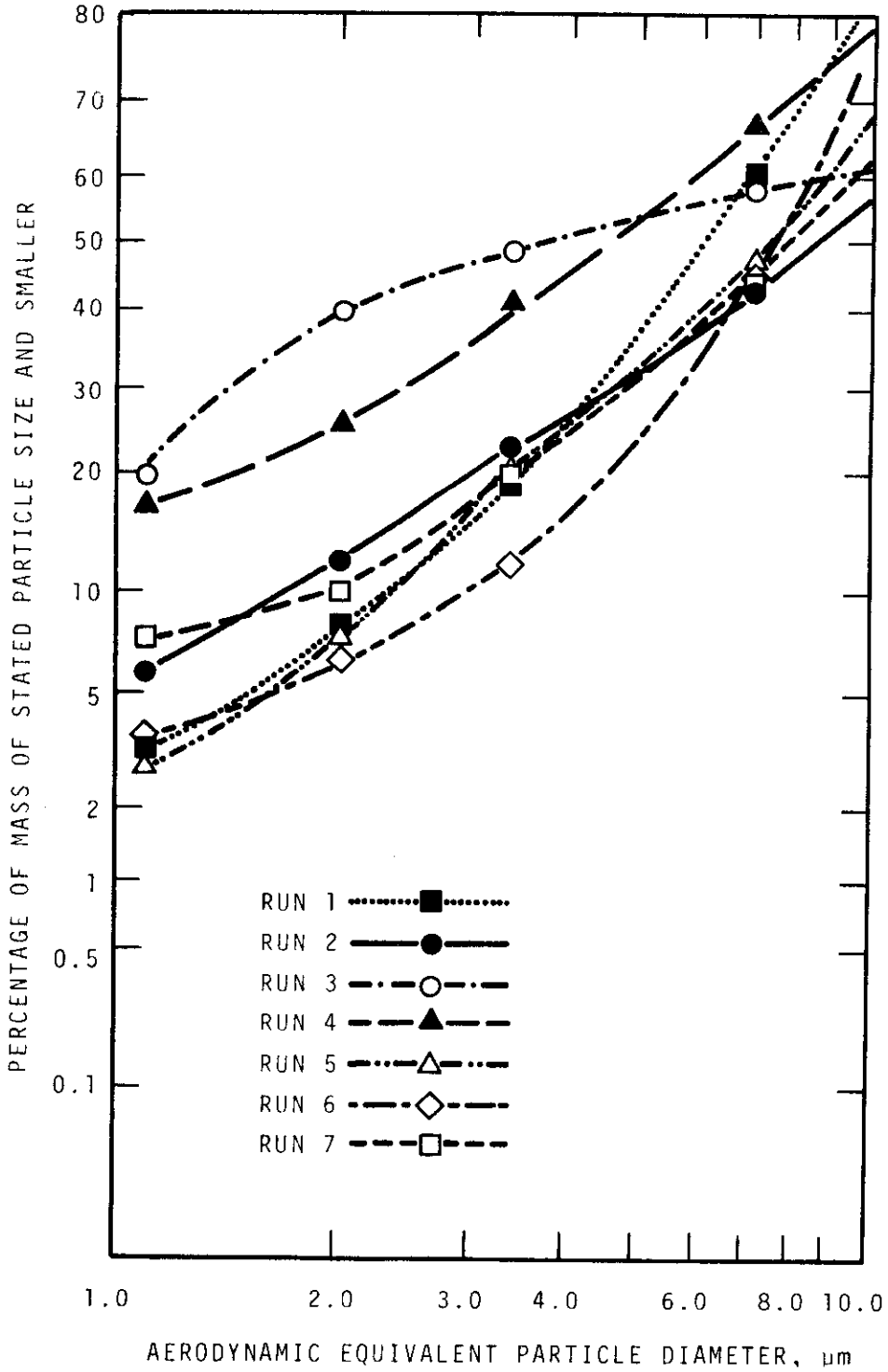

FIGURE 9

ACTIVITY DISTRIBUTION OF ALPHA EMITTERS IN 234-5Z BUILDING GASEOUS EFFLUENTS 10/12/72 TO 5/31/73

TABLE III

ALPHA ACTIVITY CONCENTRATION AND ASSOCIATED PARTICLE SIZE DISTRIBUTION EMITTED FROM 234-5Z BUILDING STACK DURING PERIOD 10/12/72 TO 5/31/73

Sampling Period	Glass Fiber Filter-50 cfm		Cascade Impactor - 20 cfm		Mass Median Diameter (μm A.E.D.*)
	Volume Sampled (cu ft)	Activity (dpm/cu ft)	Volume Sampled (cu ft)	Activity (dpm/cu ft)	
10-12-72 to 11-27-72	2.65×10^6	0.098	9.42×10^5	0.072	7.0
11-27-72 to 12-28-72	2.34×10^6	0.001	8.61×10^5	0.013	9
12-28-72 to 1-16-73	2.11×10^6	0.024	8.06×10^5	0.021	3.3
1-16-73 to 3-7-73	2.52×10^6	0.024	1.11×10^6	0.016	4.0
3-63-73 to 4-3-73	1.25×10^6	0.041	7.20×10^5	0.030	7.0
4-3-73 to 5-1-73	2.09×10^6	0.022	5.84×10^5	0.040	7.3
5-1-73 to 5/31/73	1.75×10^6	0.019	8.33×10^6	0.020	8.0

* Aerodynamic Equivalent Diameter -- having aerodynamic characteristics equivalent to a sphere of unit density of the stated size.

trations both increased and decreased during these periods. Except for routine monthly inventories the Reclamation Facility was in continuous operation during the entire time period. The incinerator operation was sporadic with more operational periods through January than from February through May. The high emission values in the initial sampling period does coincide with contamination problems in the same time period in the incinerator operation but increased activity during March and April can not be correlated to operations in that facility.

Based on the alpha activity concentrations measured and a stack flow of 2.75×10^5 cu. ft. per min., the amount of activity emitted from the stack was calculated. The values range from 80 to 542 μCi per month. These values are from 2 to 20 times greater than the values reported by ARHCO Radiation Monitoring from their stack sampling measurements over the same periods. The calculated quantity emitted calculated from the filter and cascade impactor samples and the Radiation Monitoring values are shown in Figure 10.

Various explanations are possible. The samples used in this study were extracted from a turbulent, well mixed portion of the stack. The sample for the filter was taken under near isokinetic conditions. Since the sampling line was smaller than the orifice, the particles were accelerated through the sampling line enhancing the tendency for turbulent deposition, but also favoring reentrainment. Although sampling lines were used to extract materials, the lines were a maximum of 8 feet. Thus the values determined are felt to be a reasonable approximation of the activity concentration in the 234-5Z stack.

The distance downwind of the average maximum concentration and the maximum ground level air concentration for a continuous elevated release can be calculated.[3] Assuming an effective release height of 200 feet (approximately 60 m) and release rates of 2.7×10^{-5} and 2×10^{-4} μCi per sec., the maximum air concentration in the range of 3×10^{-14} to 2×10^{-13} μCi per cu. cm., are found from 300 to 6000 m downwind assuming a mean wind speed of 2.5 mph. The air concentration calculation is orders of magnitude high in as much as the actual material is already diluted by 4.6×10^3 cu. ft. of air. The higher value is below the MPC_a for the general population.

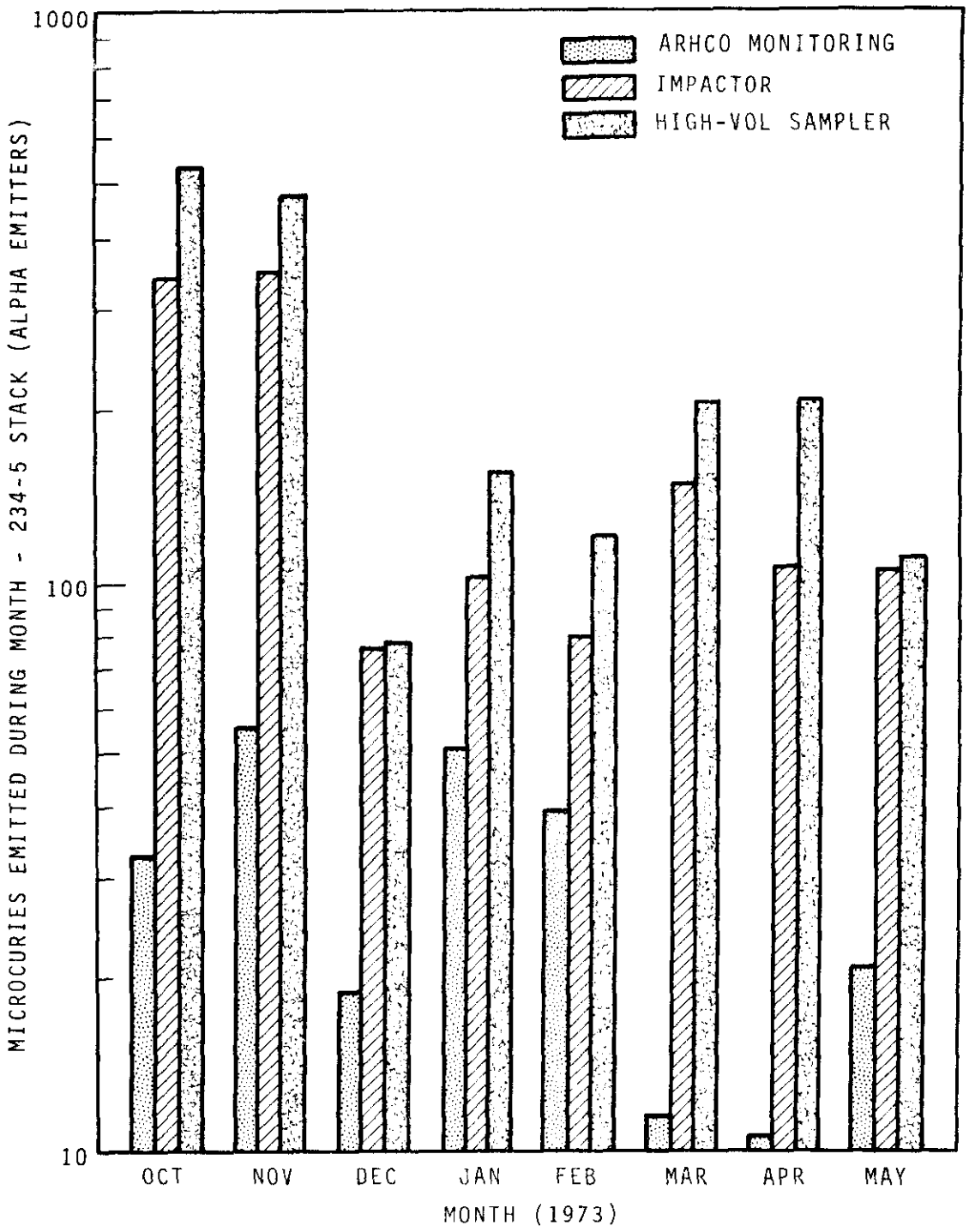

FIGURE 10

ALPHA-PARTICLE-EMITTERS IN 234-5 STACK GAS AS DETERMINED BY 3 DIFFERENT SAMPLERS

REFERENCES

[1] J. Mishima and L.C. Schwendiman. Characterization of Radioactive Particles in the 234-5Z Building Ventilation System -- Interim Report, BNWL-B-105, Battelle-Northwest, Richland, Wash., 99352 (June 1971).

[2] F.A. Perkins, Private Communications, ARHCO, July 20 , 1973.

[3] D.H. Slade, Ed., Meteorology and Atomic Energy 1968, U.S.A.E.C., Div. of Technical Information, July 1968.

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