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CHARACTERIZATION OF RADIOACTIVE PARTICLES IN THE 234-5Z BUILDING VENTILATION SYSTEMS -- INTERIM REPORT

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INTRODUCTION

The Plutonium Finishing Plant at the Hanford site has a long history of safe operation, and radioactive material release is controlled well within the established limits. But an abiding public concern of possible effects of plutonium processing operations on the atmospheric environs justifies a continuing effort to reduce potentials for release to the lowest practicable level. Routine stack monitoring is performed as part of the radiological responsibility but does not provide all the data necessary to determine the fate of particles after emission. Knowledge of the distribution of the activity in aerodynamic particle size fractions is necessary to determine the airborne behavior of released material in the atmosphere. The contribution of the various components of the exhaust system to the total emission may be helpful in delineating areas of potential gain or identifying potential trouble points within the system. A body of information has been published on the amounts and size distribution of plutonium particles in various types of installations and in accident situations,[1-11] but little information is available on the nature of particles within the ventilation system of plutonium processing plants.

OBJECTIVES

- The current study was undertaken to determine the distributions of aerodynamic sizes of plutonium-containing particles present in the 234-5Z gaseous effluents. The total concentration of plutonium as particulates was also to be determined. By direct use of the aerodynamic characteristics for classification, such factors as particle density and shape, agglomeration, etc., need not be of concern since the property of interest is measured. Obtaining size distribution data in terms of aerodynamic characteristics will generate information which can be directly used in existing dispersion and deposition models that can forecast spatial distribution and predict any build-up on the terrain.
- A second objective was to determine the amount and distribution of radioactivity associated with particles contributed by various components of the 234-5Z Bldg. exhaust system.

SUMMARY

Filter and cascade impactor samples were taken of the 234-5Z Building stack gases and of various exhaust streams within the 234-5Z ventilation system to characterize the amount and distribution by aerodynamic characteristics of particles with their associated radioactivity. Stack samples were extracted at the base of the stack. Exhaust stream samples were taken at various locations downstream of final filtration. Sampling periods ranged from 7 to 63 days and high volume samples (flows up to 72 cfm) taken of streams where extremely low activity concentrations were anticipated. Volumes of gases sampled ranged from 10^4 to 4.4×10^6 cubic feet. During the collection of these samples various conditions were disclosed that may have a bearing on the quantity of airborne plutonium detected. These observations were forwarded to the operating units concerned. Activity detected in the stack gases showed rather poor agreement with daily stack monitoring results integrated over the same period. Two components of the exhaust system -- the exhaust from the 232Z (Incinerator) Building and the 26-inch (Process) vacuum system exhaust -- may be contributing a significant amount of the total activity emitted. Another system with significant potential to contribute to the loading, the HF system exhaust, was not on-the-line during the sampling period and could not be evaluated. Air samples taken in the inlet plenum of the supply to the building showed low activity concentrations -- 6.4×10^{-8} dpm/cu.ft. to 6.7×10^{-7} dpm/cu.ft. -- indicating little, if any plutonium is recycled from the stack. The activity distribution into aerodynamic fractions was approximately log-normally distributed and surprisingly coarse -- with one exception, Median Activity Diameter (M.A.D.) ranged from 1.6 to $\sim$ 20 micron Aerodynamically Equivalent Diameter (A.E.D.).

Because these measurements were the first of their kind ever taken within the facility and because the 234-5Z Building operations during the sampling period were not regarded as fully representative, the results to date are regarded as indicative rather than definitive. A few general conclusions can be drawn from the limited data obtained.

- Although the exact level of activity in various exhaust streams could not be well defined with the data obtained, the overall efficiency of the exhaust system appears good. The low quantity of alpha particle emitters leaving the stack and low activity concentrations in the exhaust streams indicates satisfactory performance of the system.

- Little, if any, of the alpha-emitters emitted via the stack is being recycled back into the ventilation system.
- The plutonium present appears to be attached to larger, non-active particles.
- The source and nature of the fairly large quantity of inert material in certain ducts should be investigated before additional data on radioactive particles can be obtained. Additional work to define the activity release during a variety of processing conditions will better define stack activity emissions.

EXPERIMENTAL

234-5Z BUILDING VENTILATION AND EXHAUST SYSTEM

The 234-5Z Building ventilation and exhaust system is composed of several subsystems. The exhaust system is diagrammed in Figure 1.

Building Ventilation Supply Air

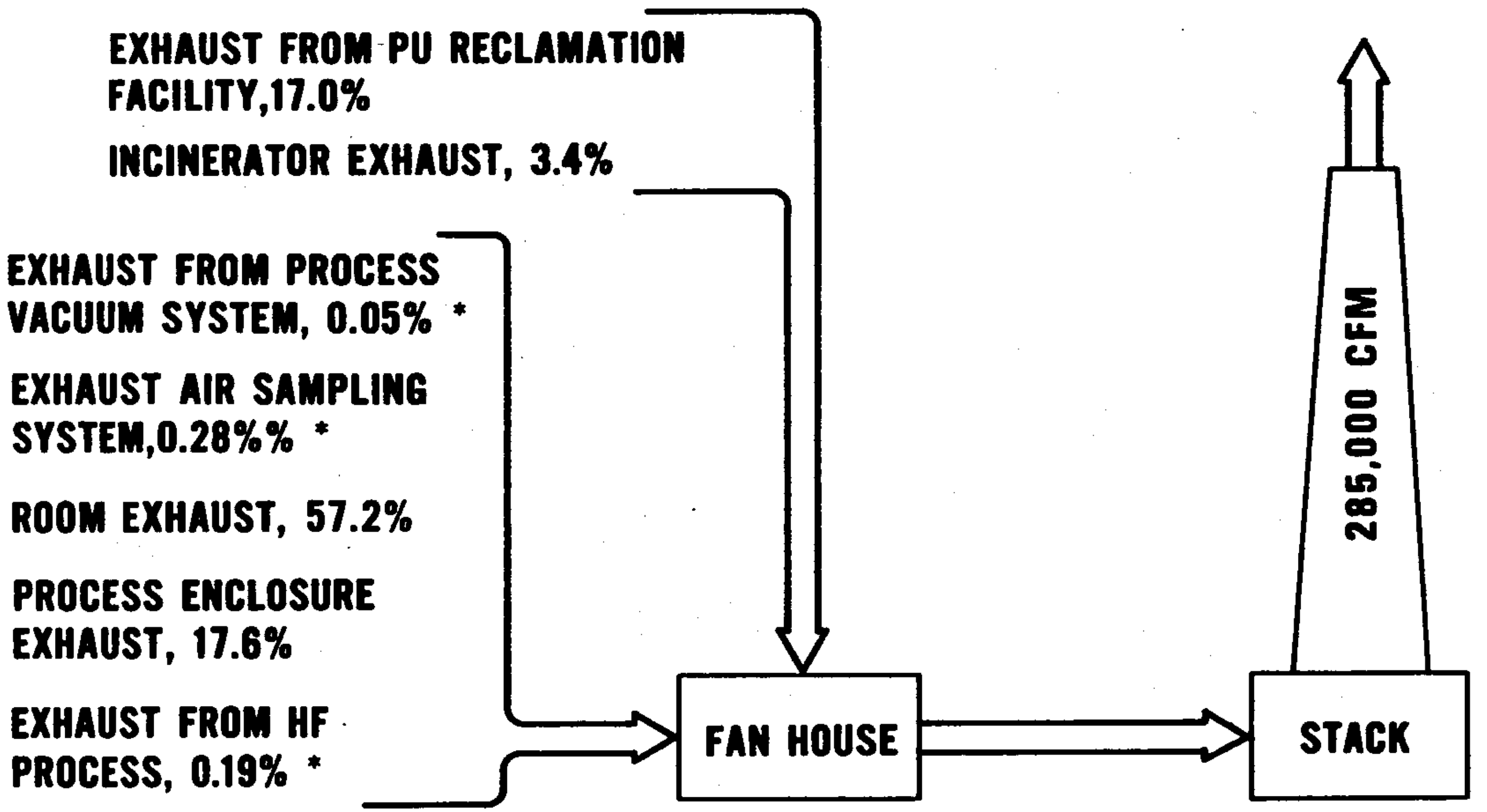
Air is supplied to all of the 234-5Z, 236Z (Plutonium Reclammation) and 242Z (Americium Recovery) Building by eight fans located on the second floor of the 234-5Z Building. The 232Z (Incinerator) Building has its own air supply.

Exhaust

Zone 1 -- Air from uncontaminated areas -- offices, shop areas, etc., separated from process areas by air locks -- is exhausted through the roof at several points with no filtration.

Zone 2/3 -- Air from all working areas in the process section of the 234-5Z and 242-B Buildings is manifolded and passed through large banks of HEPA filters. Generally three or four of seven banks are used at any given time. The exhaust flow passing through the system is approximately 163,000 cfm and contributes approximately 57.2 percent to the exhaust gases emitted from the stack.

Zone 4 -- Exhaust from process enclosures and hoods is filtered at the enclosed outlet, after the exhaust from several enclosures is manifolded, and, by a large HEPA filter bank when all exhaust air is combined. Thus Zone 4



* BASED ON ESTIMATED FLOW

FIGURE 1

PLUTONIUM FINISHING FACILITY GASEOUS EFFLUENT exhaust is filtered three times prior to release. Total flow in this system is 46,500 cfm and contributes some 18 percent to the exhaust gases emitted from the stack.

Exhaust from 236Z (Plutonium Reclamation) Building -- Combined exhaust from process enclosures and process rooms after double and single filtration respectively joins 234-5Z Building exhaust at 291Z Fan House. Exhaust flow is 46,500 cfm and contributes 17.0 percent of exhaust gases.

Exhaust from 232Z (Incinerator) Building -- Combined room and enclosure exhaust after triple filtration joins exhaust plenum in 291Z Fan House. Flow level is at 9000 cfm which is about 3 percent of exhaust gases emitted from stack.

10-Inch (Air Sampling) Vacuum System -- Exhaust primarily from air sampling heads in the process areas passed through a HEPA filter. Thus, this stream is made up of process room air (already normally less than detection limits) filtered twice -- once at inlet and subsequently by the system filter. Approximate flow is 750 cfm, comprising 0.3 percent of stack flow.

HF System -- Exhaust from dry processing is passed through three stages of absolute filters. This exhaust has been the source of some stack emission on occasions due to the corrosive nature of gas on filters. Some 500 cfm passes through system making up 0.2 percent of gases emitted from the stack.

26-Inch (Process) Vacuum System -- The vacuum exhaust from process and laboratory vacuum systems contains much vapor, principally water, and passes through two sets of demisters plus a HEPA filter. Liquid from the demister is recycled through the system. Flow through this system is small, only 140 fpm which represents less than 0.1 percent of volume emitted from the stack.

Stack

All exhausts from the 234-5Z Building are released via a 16-foot diameter by 200-foot tall metal-lined, concrete stack located south of the 291Z Fan House. Gas flow is approximately 285,000 cfm.

SAMPLING SITES AND EQUIPMENT

Filter and Andersen impactor samples were taken at various sites in the ventilation and exhaust system to determine the activity concentration and distribution in various aerodynamic diameter fractions. Existing facilities were used whenever possible. Since much of the air had been filtered two to three times prior to sampling, a very fine size distribution was anticipated and requirements for isokinetic sampling were not rigorously imposed. Samples were taken at the base of the stack, in the inlet plenum of one of the air supply fans, and in the duct work of most of the exhaust subsystems downstream of the final filtration (the sampling arrangements are described in detail in the subsequent paragraphs). Two exhaust subsystems -- the Zone 2/3 Exhaust and 10-inch Vacuum System -- were not sampled.

The Zone 2/3 Exhaust is room air from the processing areas and is routinely sampled by Radiation Monitoring. The measured airborne concentration is beneath detectible limits in the vast majority of cases. Even when the airborne activity is above detectible limits, the high concentrations are limited to a small area and, thus, any airborne activity is diluted and filtered before a sample would be taken in this sampling scheme. Because of the difficulties of obtaining a sample and the anticipated low activity, a decision was made not to sample this exhaust stream.

The 10-Inch Vacuum System is primarily used to draw samples of airborne activity from Zone 2/3 room air. The air is filtered twice prior to its entry into the Main Exahust Plenum -- by the sample filter and a HEPA filter in the 10-Inch Vacuum System exhaust duct. Since the Zone 2/3 room air is already below detectible limits, filtering the air twice prior to sampling would insure very low activity levels. The 10-Inch Vacuum System contributes a very small fraction (0.3 percent) to the total exhaust from the building. For these reasons, no attempt was made to obtain a sample from this source.

Stack

Two 4-inch flanged nozzles were installed in the 3 ft. × 3 ft. access at the base of the 16 ft. diameter × 200 ft. tall stack. Nominal air velocity in the stack is about 1250 ft. per minute. Filter samples were extracted by a probe of 1-inch diameter, schedule 10 pipe extending 5 ft. into the stack. A 90° elbow with a tapered opening was butt welded to the end of the pipe and the opening faced into the exhaust stream. The sample stream passed through a 1-inch ball valve into a 4-inch glass fiber filter holder directly coupled to a two stage centrifugal blower. The blower exhaust was released through a HEPA filter. Sampling rate used was 61 cfm. Sampling data are listed in Table I.

Impactor samples were obtained via a 4 ft. long probe of 0.5 inch stainless steel ODT with a 90° bend at the end. The orifice of the probe faced into the flow. The sample stream passed through a 1-inch diameter stainless steel ball valve into an eight-stage Andersen impactor, followed by a 2-inch diameter glass fiber filter. One cfm was drawn through the impactor by maintaining a 34-inch W.G. negative pressure on the downstream side of the instrument by a sliding vane, rotary, vacuum pump.

236Z Building Exhaust

Two 4-inch flanged nozzles were installed in a 5 ft. × 5 ft. duct just upstream of its entry into the exhaust plenum in the 291Z Fan House. Filter samples were taken by a 1-inch diameter stainless steel ODT with a 90° bend at the end. The opening of the probe was faced into the exhaust flow. The air passed through a one-inch diameter stainless steel ball valve to a 4-inch glass fiber filter. Air was drawn through the equipment by a multistage centrifugal blower at a flow of 50 cfm. The probe and valving arrangement used to obtain impactor samples were identical to that described for stack sample. A negative pressure of 36 inches W.G. was required to maintain a one cfm through this impactor. Exhausts from both samples were returned to the 232Z Building exhaust duct via an existing valved 1/2-inch half coupling.

232Z Building Exhaust

Both the filter and impactor samples were drawn from existing valved 3/4-inch half couplings in the duct wall near its entry into the exhaust plenum. Sample holders and pumps were the same as described in the previous section.

TABLE I

SAMPLING DATA

Location	Sample Type	Flow cfm	Date	Total Sample cf	Duration Of Run	Observations
STACK	Filter - 1	61	10/5 to 11/23	3.42×10^6	38 d, 22 hr	dark grey
	Impactor - 1	1	12/2 to 1/11	51840	36 d	considerable condensation
EXHAUST, 236Z BLDG.	Filter - 2	49.5	10/16 to 11/23	2.65×10^6	38 d	much reddish-brown, granular paste
	Impactor - 1	1	10/16 to 11/23	54220	38 d	rubber cement hardened, Al oxidized
EXHAUST, 232Z BLDG.	Filter - 2	49.5	12/2 to 1/11	2.85×10^6	39 d, 23.5 hr	small amount of ash
	Impactor - 1	1	12/2 to 1/11	57570	39 d, 23.5 hr	
26 IN. VACUUM SYSTEM	Filter - 3	1	12/2 to 12/9	10080	7 d	
	Filter - 3	1	12/15 to 1/11	39090	27 d, 3.5 hr	filter wrinkled
	Impactor - 2	1	10/15 to 11/23	56160	39 d	loaded with glassy, hard material
	Impactor - 2	1	12/2 to 1/11	49170	34 d, 3.5 hr	loaded with glassy, hard material
309 FILTER ROOM	Filter - 1	67	9/24 to 10/13	unknown	19 d, 11.5 hr	filter torn within 3 days
	Filter - 4	49	12/8 to 12/28	2.54×10^6	36 d	lost in analysis
	Filter - 4	49	12/29 to 3/2	4.44×10^6	62 d, 23.5 hr	
	Impactor - 2	1	9/24 to 10/13	28050	19 d, 11.5 hr	
	Impactor - 3	1	12/8to 12/28	28800	36 d	
HF SYSTEM	Filter - 5	1	12/2 to 1/7	51840	36 d	filter wrinkled
	Impactor - 4	1	12/2 to 1/7	51840	36 d	
INTAKE-AIR SUPPLY	Filter -- 1	72	9/24 to 10/13	1.34×10^6	12 d, 21 hr	Staplex sampler burned out
	Filter - 4	52.6	11/23 to 12/8	1.14×10^6	15 d	
	Filter - 4	52.6	12/8 to 12/28	2.54×10^6	34 d, 0.5 hr	

Filter 1 4" glass fiber filter + Staplex Sampler
Filter 2 4" glass fiber filter + Rotron pump
Filter 3 2" glass fiber filter + sliding vane pump
Filter 4 8" x 10" glass fiber filter + Gelman "Hurricane", low speed
Filter 5 2" PVC membrane filter + sliding vane pump

Impactor 1 8 stage Andersen cascade impactor with 2" glass fiber filter (impactor plates coated with rubber cement + sliding vane pump, 1 cfm)
Impactor 2 6 stage Andersen cascade impactor with 2" glass fiber filter (impactor plates coated with rubber cement) + sliding vane pump, 1 cfm
Impactor 3 6 stage Andersen cascade impactor with 2" glass fiber filter (impactor plates coated with petroleum jelly) + sliding vane pump
Impactor 4 6 stage Andersen cascade impactor with 2" PVC membrane filter (impactor plates coated with rubber cement) + sliding vane pump

26-Inch Vacuum System

A "Tee" was attached to an existing 1-inch half coupling on the 6-inch exhaust line from the vacuum pump to the main exhaust plenum. Each end of the "Tee" was sealed by a 1-inch ball valve. A 2-inch glass fiber filter and a six stage Andersen cascade impactor were attached to the valves. Flow through the filter was measured at 1 cfm by a flowmeter and a 1 cfm flow through the impactor was maintained by holding the downstream pressure at a negative 24-inches W.G. Air was drawn through the sampling equipment by a sliding vane, rotary, vacuum pump. Exhaust was discharged to an existing, nearby outlet to the 10-inch vacuum system.

H.F. System

One-cfm samples were extracted from the downstream portion of the Lucite enclosure housing the final filtration bank. Both samples were extracted by means of aluminum 1/2-inch ODT probes sealed in 8-inch plastic glove port covers sealed to existing glove ports. Polyvinyl chloride filters were used to determine the mass concentration of the stream and as the final stage in a six stage Andersen impactor. Exhaust gases were drawn through both samplers by a single sliding vane, rotary, vacuum pump with a divided inlet. Exhaust from the vacuum pump is returned to the HF system downstream of the sampling probes.

Zone 4 Exhaust -- 309 Filter Room

Filter and impactor samples were obtained by placing the sampling units in the plenum downstream of the filter bank in the 309 filter room. The initial filter sample was taken using a pre-calibrated STAPLEX two-stage centrifugal sampler with a 4-inch diameter glass fiber filter. Difficulties noted in a later section resulted in finally using an 8-inch × 10-inch glass fiber filter on a different type of centrifugal sampler. The size distribution of the particles airborne was determined by a six-stage Andersen impactor attached to a sliding vane, rotary, vacuum pump. A 1-cfm flow was maintained by holding a negative pressure of 12 inches W.G. on the downstream side of the impactor.

Air-Intake

Because of the very low concentrations anticipated for this location, only mass concentration (filter) samples were taken. The initial sample was taken using a 4-inch glass fiber filter and a pre-calibrated multistage centrifugal pump. The pump was out of service in a short period and further samples were taken with an 8-inch by 10-inch glass fiber filter with a pre-calibrated centrifugal pump of another design.

OPERATIONAL EXPERIENCE

Difficulties were encountered during sampling because of the demanding conditions imposed during the study, particularly the long periods of continuous unattended operation. Pumps which were capable of both high volumes and long sampling periods were not readily obtained.

A 4-inch glass fiber filter supported on an open support (three 1/8-inch flat bars in the shape of a star, furnished with the unit) was initially tried in three locations -- stack, 309 filter room, and air inlet. The bars did not provide enough support for the glass fiber filters and the filters tore. Closer mesh supports were fabricated from stainless steel perforated plate. But all the blowers were subsequently removed from service due to failures. The sampler in the air inlet overheated after approximately 13 days service. The unit on the stack functioned for the initial sampling period (39 days) but the bearing froze and the motor would not restart. Because of the difficulties associated with retrieving samples in this location, the sampler in the 309 filter room was allowed to operate 20 days with a torn filter. The equipment was replaced upon retrieval because of the additional hazards that could arise if problems were encountered in the equipment in such an inaccessible location. Mass concentration samples in the 309 filter room and air inlet plenum were obtained with an 8-inch by 10-inch glass fiber filter and another type of centrifugal blower operating at a low speed. These have proven satisfactory in these locations. Since a useable sample was obtained from the stack, the sampler was not replaced. A new sampler will be required before additional sampling can be performed.

All sliding vane, rotary vacuum pumps showed a tendency to clog upon extended use. This type of pump requires a constant addition of light machine oil into the pumping chamber. Heat and excess oil cause the accumulation of tar-like residues on the vanes causing the vanes to stick. The problem is magnified in this work due to the inaccessibility of the pumps and lack of round-the-clock coverage.

The sampling arrangements in most locations also require modification. All impactors and filter holders should be equipped with quick-connect couplings to minimize handling times. The space and heights of some sample sites is inconvenient and could be viewed as hazardous -- 232Z/236Z Bldg. Exhaust and 26-inch vacuum system. These require rearrangement and additional service. Finally, many impactors were operated in a horizontal position which is undesirable. Additional piping must be installed for proper operation of the impactors in an extended study.

Conditions were observed at a few sampling sites which may indicate some problem spots in the exhaust system and challenge the validity of the samples obtained. They are:

- A large amount of moist, reddish-brown granular material was found in the sample of the 236Z Bldg. exhaust. The entire inlet section and filter were coated with some 1/4-inch of this material. The individual grains appeared quite coarse (greater than a millimeter in diameter) and the material appears much too large to have passed through filters. Thus, the material must be generated downstream of the filter bank. The exhaust plenum in the 236Z Bldg. downstream of the filters is made of concrete. But the duct from the building to the 291Z Fan House is painted metal (black iron/mild steel) and is above ground. During the winter (the season in which the study was conducted), moisture in the warm building air would be condensed by the cold outdoor temperature. Heat transfer through the metal duct would be good. Since the duct is horizontal, moisture would accumulate in the bottom of the duct and be broken away from the surface by the flexing created by variations in the outdoor temperature. Since mass flux is toward a cooler surface, small airborne particles may also accumulate on surfaces to be dislodged with rust particles at a later time. If activity-bearing rust particles are accumulating at some point in the system, the accumulated material can possibly be a source for subsequent spread of contamination. The presence of rust is also indicative of corrosion which may ultimately cause failure of the duct.
- Presence of highly active, coarse, red and grey particles in the 232Z Bldg. exhaust sample. A small quantity of this material was found on the glass fiber filter. The outlet from which this sample was extracted is on a vertical wall on a sloping section of duct, not the optimal conditions for obtaining samples of coarse particles. Thus, the sample may only be

a small portion of this type of material passing down the duct. Ash from the incineration operation may have accumulated in the duct and past the filter during changing or via penetrations. The material may accumulate in dead air pockets and only transferred during turbulent conditions associated with rapid air changes. Room air filters are recessed in the floor and present potential pockets for the accumulation of material.

- All the interior surfaces and plates from the 26-inch vacuum system cascade impactor samples were coated with a glassy, colorless coat. The material had the appearance of the residue from hard water.
- Presence of significant quantities of activity bearing rust on the floor of the 309 filter room. Each entry into the space downstream of the filters was well monitored and, in almost all cases, the presence of activity on clothing (shoe covers and gloves) was associated with the color of rust. Since entry and retrieval of equipment is not a trivial task, the sampling arrangement was reduced to bare essentials and placed on the floor. Entries were made weekly to oil the sliding vane, rotary, pump used for the impactor sample. These entries potentially entrained the rust on the floor.[12,13]

SAMPLE PREPARATION

Dissolution of particulate material collected was performed in plastic-lined, low background hoods in the 3706 Bldg. using 9 N HNO_3-0.1 N HF. Glass fiber filters were carefully placed into premarked glass beakers and enough mixed acid added to cover the filter. After the beaker had been warmed for 1 hour, the liquid was decanted to a second beaker, more acid added to the filter and rewarmed. The liquids were combined and filtered. The liquid volume was reduced by evaporation, cooled, filtered through analytical grade paper and brought to a known volume in a volumetric flask. The plutonium in some samples was subsequently separated by solvent extraction to minimize the effects of salts in the aqueous solution on alpha counting instruments.

All glass plates used to collect samples in the cascade impactors were coated with a thin layer of rubber cement prior to use to provide a sticky surface to retain the particles. After sampling, the rubber cement layer was carefully removed, placed in a porcelain crucible and gently heated with a burner until all the carbon was vaporized. A small quantity of mixed acids was added and the crucible warmed. The liquid was allowed to cool, filtered and the sample made up to a known volume.

Liquid samples were submitted to the Technical Analysis Section for determination of alpha activity by scintillation counting.

RESULTS

The alpha activity concentrations detected in all exhaust streams was quite low -- greater than 70 percent of the samples analyzed were below the MPC_{air} for a 168 hour week. The activity measured in individual samples and the precision of the measurement (when significant) are listed in Table II, "Analytical Results". The quantity of alpha activity emitted by the stack measured during this study indicated a rather poor agreement with emission values calculated from the daily stack sample taken by the Radiation Monitoring unit over the same period (see Table III). High alpha activity concentrations (greater than MPC_{air} for a 40 hour week) were measured in some of the samples from two of the exhaust streams -- 232Z Building, and 26-inch Vacuum System. The particles bearing the activity appear to be surprisingly coarse for material that has passed through multiple stages of filtration and indicates attachment of activity to inert particles (see Table IV, "Activity Distribution of Aerodynamic Fraction"). Results for individual sampling sites is discussed below.

STACK

Single filter and cascade impactor samples were taken during the last few months of calendar year 1970. Total volumes of exhaust sample were 3.42×10^6 and 5.18×10^4 cu. ft. respectively. A sufficient quantity of material had been deposited on the filter during the course of sampling to discolor the filter (see Table 1, Sampling Data). Considerable amounts of moisture condensed in the cascade impactor due to the great differences in temperature between indoor and outdoors at this time of year. The quantity of activity measured on the filter sample indicates an activity concentration during the time of sampling of 10^{-4} dpm per cu. ft. (equivalent to a single PuO_2 sphere less than 0.1μ in diameter per cu. ft. -- see Table V, Activity Anticipated in PuO_2 Particles of Various Diameters), and a total emission of about one percent of that obtained by the summation of Daily Stack Samples for the same period taken routinely

TABLE II

ANALYTICAL RESULTS

Sample Designation	Sample	Sampling Period	Contamination Level* (dpm)	Activity Level Detected (dpm)	Activity Concentration In Air (dpm/ft^3)
S-1	4" glass fiber filter	38 d, 22 hr	nda	350	10^{-4}
S-2	0 stage cascade impactor		"	2.4 ± 1.2	
S-3	1 stage cascade impactor		"	4.1	
S-4	2 stage cascade impactor		"	5.0	
S-5	3 stage cascade impactor		"	4.8	
S-6	4 stage cascade impactor	36 d	"	2.7 ± 0.57	6.4×10^{-3}
S-7	5 stage cascade impactor		"	2.7 ± 0.86	
S-8	6 stage cascade impactor		"	6.7	
S-9	7 stage cascade impactor		"	1.1 ± 0.12	
S-10	filter cascade impactor		"	3.6	
PR-1	4" glass fiber filter	38 d	†	1.0 ± 0.8	4.0×10^{-7}
PR-2	0 stage cascade impactor		nda	0.77 ± 0.11	
PR-3	1 stage cascade impactor		"	1.0 ± 0.2	
PR-4	2 stage cascade impactor		"	0.44 ± 0.10	
PR-5	3 stage cascade impactor		"	0.30 ± 0.10	
PR-6	4 stage cascade impactor	38 d	"	0.70 ± 0.12	10^{-4}
PR-7	5 stage cascade impactor		"	0.79 ± 0.11	
PR-8	6 stage cascade impactor		"	0.48 ± 0.10	
PR-9	7 stage cascade impactor		"	0.54 ± 0.11	
PR-10	filter cascade impactor		"	0.40 ± 0.10	
IN-1	4" glass fiber filter	40 d	5000	1.0×10^5	0.035
IN-2	0 stage cascade impactor		nda	220	
IN-3	1 stage cascade impactor		"	190	
IN-4	2 stage cascade impactor		"	240	
IN-5	3 stage cascade impactor		†	150	
IN-6	4 stage cascade impactor		nda	120	0.018
IN-7	5 stage cascade impactor	40 d	"	46	
IN-8	6 stage cascade impactor		"	40	
IN-9	7 stage cascade impactor		"	13	
IN-10	filter cascade impactor		"	20	
V-1	2" glass fiber filter	7 d	†	2.8×10^4	2.8
V-2	filter cascade impactor		unknown	1100	
V-3	1 stage cascade impactor		"	660	
V-4	2 stage cascade impactor		"	980	
V-5	3 stage cascade impactor	34 d	"	980	0.23
V-6	4 stage cascade impactor		"	2100	
V-7	5 stage cascade impactor		"	3600	
V-8	6 stage cascade impactor		"	3000	
V-9	2" glass fiber filter	34 d, 3.5 hr	nda	400	0.01
V-10	1 stage cascade impactor		500	150	
V-11	2 stage cascade impactor		500	38 ± 4.2	
V-12	3 stage cascade impactor		†	32	
V-13	4 stage cascade impactor	34 d, 3.5 hr	nda	65	0.11
V-14	5 stage cascade impactor		"	270	
V-15	6 stage cascade impactor		"	36	
V-16	filter		5000	52	
E-1	4" glass fiber filter	19 d, 11.5 hr	†	57	
E-2	filter cascade impactor		nda	3	
E-3	1 stage cascade impactor		"	14	
E-4	2 stage cascade impactor		"	1.9	
E-5	3 stage cascade impactor	19 d, 11.5 hr	"	13	1.2×10^{-3}
E-6	4 stage cascade impactor		"	0.39 ± 0.09	
E-7	5 stage cascade impactor		"	0.25 ± 0.09	
E-8	6 stage cascade impactor		"	0.33 ± 0.09	
E-9	8" × 10" glass fiber filter	36 d	"	lost in anal.	
E-10	filter cascade impactor		"	1.6	
E-11	1 stage cascade impactor		"	14	
E-12	2 stage cascade impactor		"	3.3	
E-13	3 stage cascade impactor	36 d	"	0.85 ± 0.11	7.5×10^{-4}
E-14	4 stage cascade impactor		"	0.90 ± 0.11	
E-15	5 stage cascade impactor		"	0.40 ± 0.09	
E-16	6 stage cascade impactor		"	0.58 ± 0.13	
E-17	8" × 10" glass fiber filter	62 d, 23.5 hr	2000 to 5000	20	1.2×10^{-3}
HF-1	2" PVC membrane filter	36 d	nda	60	0.0012
HF-2	1 stage cascade impactor		500 to 1000	930	
HF-3	2 stage cascade impactor		2000	1400	
HF-4	3 stage cascade impactor		1000	450	
HF-5	4 stage cascade impactor	36 d	500	170	0.059
HF-6	5 stage cascade impactor		nda	38	
HF-7	6 stage cascade impactor		"	18	
HF-8	filter cascade impactor		500	50	
AI-1	4" glass fiber filter	12 d, 21 hr	nda	0.37 ± 0.09	2.8×10^{-7}
AI-2	8" × 10" glass fiber filter	15 d	"	0.073 ± 0.065	6.4×10^{-8}
AI-3	8" × 10" glass fiber filter	34 d, 0.5 hr	"	.1.8	6.7×10^{-7}

* Probe area SCINTRAN
† Possible trace quantity
nda No detectable activity

TABLE III

ALPHA PARTICLE EMISSION RATE OF SAMPLES TAKEN

Location	Date	dpm - ft^3	ft^3 - day	dpm - Day	Stack* dpm - day	% Act.
STACK -- filter	10/15 to 11/13	10^{-4}	4.10×10^8	4.10×10^4	2.85×10^6	
impactor	12/2 to 1/11	6.4×10^{-3}		2.62×10^6	1.59×10^5	
236Z EXH -- filter	10/16 to 1/23	4×10^{-7}	6.62×10^7	2.6×10	2.86×10^6	9×10^{-4}
impactor		10^{-4}		6.62×10^3		.2
232Z EXH -- filter	12/2 to 1/11	.035	1.3×10^7	4.55×10^5	1.59×10^5	28.6
impactor		.018		2.3×10^5		14.5
26" VAC -- filter	12/2 to 12/24	2.8		5.6×10^5	1.52×10^6	36.8
	12/15 to 1/11	.01	2.0×10^5	2×10^3	1.4×10^6	.1
impactor	10/15 to 11/23	.23		4.6×10^4	2.85×10^6	1.6
	12/15 to 1/11	.11		2.2×10^4	1.40×10^6	1.6
309 FILTER - filter	12/29 to 3/2	4.5×10^{-6}		3.01×10^2	2.21×10^5	.01
impactor	9/24 to 10/13	.001	6.7×10^7	6.7×10^4	7.56×10^5	8.9
	12/8 to 12/28	7.5×10^{-4}		5.02×10^4	7.4×10^5	6.7
HF -- filter	12/2 to 1/17	.0012	7.2×10^5	8.64×10^2	1.86×10^6	.05
impactor		.059		4.25×10^4		2.3

*Radiation Monitoring daily stack samples averaged over appropriate period.

TABLE IV

ACTIVITY DISTRIBUTION BY AERODYNAMICALLY EQUIVALENT DIAMETER PERCENT ACTIVITY ASSOCIATED WITH PARTICLES OF LESS THAN STATED DIAMETER

Impactor Stage	Aerodynamic Fraction (Stage cut-off)	Stack	Exhaust From 236Z Bldg.	Exhaust From 232Z Bldg.	26-Inch Vacuum System -- 1	26-Inch Vacuum System -- 2	309 Filter Room -- 1	309 Filter Room -- 2	HF System
Filter	*	11	7	1.9	8.9	90	9	7	2
0	.3	14	17	3.2					
7	1.	34	36	7.0	33	90	10	10	3
6	2.	42	41	11.4	62	95	11	12	3
5	3.3	50	54	22.9	79	96	12	16	9
4	5.5	65	60	37.3	87	97	51	20	24
3	9.2	80	68	60.5	95	92	57	35	70
2	†	93	86	78.5					
1	>30								
MEDIAN ACTIVITY DIAMETER		3	3	7.9	1.6	>0.3	9.5	∿20	7.4

A.E.D. -- Aerodynamically Equivalent Diameter, equal to sphere of stated size of unit density.

* Less than 0.1 micron A.E.D. in 8-stage Andersen cascade impactor and less than 0.3 micron A.E.D. in 6-stage impactor.

† Less than 30 micron A.E.D. in 8-stage Andersen cascade impactor and greater than 9.2 micron A.E.D. in 6-stage impactor.

TABLE V

ACTIVITY ANTICIPATED IN $^{239}PuO_2$ PARTICLES OF VARIOUS DIAMETERS

(Specific activity 1.34×10^{11} dpm/g Pu)

Diameter (micron)	Activity (dpm)
0.1	7.5×10^{-4}
0.3	0.02
1	0.76
2	6.1
3.3	27.2
5.5	126
9.2	590
30	2.4×10^{4}

by Radiation Monitoring (see Table III). Based upon the performance of the type of sampler used at this location, some loss of flow during the later stages of sampling is possible although not to the extent indicated by the difference in activity determined. A higher activity concentration for a different period is obtained by summation of all activity found in the eight-stage cascade impactor -- 6.4×10^{-3} dpm per cu. ft. The total emission indicated is approximately 20 times that shown by Daily Stack Samples for the same period. The Median Activity Diameter (M.A.D.) of the airborne particles sampled was approximately 3 micron A.E.D., indicating a relatively coarse aerosol. A PuO_2 sphere of 3 microns A.E.D. would show about 25 dpm and thus, if the activity present were all in the form of 3 micron PuO_2 particles only a single particle would be found in each 4 to 20 thousand cu. ft. of exhaust. The number of PuO_2 particles represented by the activity found on each stage of the impactor samples (based on the stage cut-off diameter) is shown on Table VI. Only in the 3 stages representing the finest size fractions was enough activity measured to indicate the present of more than 1 particle. In most cases, the activity measured was considerably less than should be found in a single particle (less than 1/100th the activity in a single particle in stage 2). Thus, the plutonium appears to be randomly attached to inert particles.

236Z (PLUTONIUM RECLAMATIONS) BLDG. EXHAUST

Single filter and cascade impactor samples were taken during the period 10/16/70 to 11/23/70. Flow rate for the filter sample was 50 cu. ft. per min. and 1 cu. ft. per min. for the impactor sample during the 38 days of sampling. Total volumes of samples were 2.65×10^6 and 5.42×10^4 cu. ft., respectively. A large quantity of moist, reddish-brown, granular material was found on the filter sample (see EXPERIMENTAL, Operating Experiences) and the rubber cement surface on impactor plates and impactor walls appeared oxidized. The activity concentration calculated from the filter sample was 4.0×10^{-7} dpm per cu. ft. and is probably low due to losses during dissolution, extraction and counting, resulting from the high salt content. Summation of the activity measured on all stages of the cascade impactor indicates an activity concentration for this stream of 10^{-4} dpm per cu. ft. (2×10^{-15} μCi per cu. cm or 3×10^{-3} MPC_{air} for continuous exposure, 168 hr/week). This quantity of activity represents the activity in a single PuO_2 particle of less than 0.1 micron A.E.D. per cu. ft. The M.A.D. for particles sampled from this stream was 3 micron A.E.D., and the

TABLE VI

CALCULATED NUMBER OF PuO_2 PARTICLES REPRESENTED BY ACTIVITY PRESENT ON VARIOUS IMPACTOR STAGES

Impactor Stage	Aerodynamic Fraction (stage cut-off)	Stack	Exhaust From 236Z Bldg.	Exhaust From 232Z Bldg.	26-Inch Vacuum System -- 1	26-Inch Vacuum System -- 2	309 Filter Room -- 1	309 Filter Room -- 2	HF System
Filter	*	4800	523	27000	1100	26000	150	15	2900
7	.3	55	27	650					
6	1	9	<1	53	24	47	1	<1	24
5	2	<1	<1	8	29	44	1	<1	6
4	3.3	<1	<1	4	17	2	1	<1	6
3	5.5	<1	<1	1	8	<1	40	<1	4
2	9.2	<1	<1	<1	8	<1	6	<1	2
1	†	<1	<1	<1	5	<1	43	<1	2
0	>30	<1	<1	<1					

* Less than 0.1 micron A.E.D. in 8 stage Andersen cascade impactor and less than 0.3 micron A.E.D. in 6 stage impactor (A.E.D. -- Aerodynamically Equivalent Diameter, particle exhibits aerodynamic characteristics equivalent to a sphere of stated diameter of unit density).

† Less than 30 micron A.E.D. in 8 stage Andersen cascade impactor and greater than 9.2 micron A.E.D. in 6 stage impactor.

activity distribution similar to that measured in stack gases -- both are roughly log-normally distributed with the 236Z sample having a slightly steeper slope. Using the higher activity concentration measured, 10^{-4} dpm per cu. ft., a single average PuO_2 particle would be present in each 2.5×10^5 cu. ft. of exhaust. Again, the activity found on the various impactor stages is insufficient to indicate the presence of PuO_2 particles beyond the 3 smallest size fractions (Table VI), and suggests the attachment of small active particles to larger inert particles. The activity measured in the 236Z Building exhaust represented only 0.2 percent of that emitted from the stack as measured by Daily Stack Samples (see Table III). As measured, this stream has only 1/10th the activity concentration of the stack gas and is aiding in diluting the activity contributed by other exhaust streams.

232Z (INCINERATOR) BLDG. EXHAUST

Single filter and impactor samples were taken during the period 12/2/70 to 1/11/71. The sampling equipment and flow rates were the same as used in the 236Z Building sample. Total volumes sampled during the 40 day sampling period were 2.85×10^6 cu. ft. for the filter and 5.76×10^4 cu. ft. for the impactor sample. A small quantity of highly active, large particles were found on the filter although the filter itself was not noticeably discolored. The activity concentration was at 5 times that measured in the stack gases and, since this stream contributes about 3 percent of the stack gases, the stream appears to be the major contributor of activity in the system. In this sampling period, the activity in the 232Z Building exhaust was approximately 150 to 300 percent of that measured by Daily Stack Samples taken by Radiation Monitoring. If these results were truly representative, activity may be accumulating in the area of the main exhaust plenum-base of the stack. The impactor was unaffected by sampling and the rubber cement remained unchanged. The M.A.D. determined was almost 8 microns A.E.D., considerably coarser than the two previous samples. The particles appeared to be distributed roughly log-normally. Although a greater amount of activity was present in the impactor samples, the activity measured was insufficient to indicate the presence of PuO_2 particles in the 3 coarse size fractions.

26-INCH VACUUM SYSTEM

Two filter samples and two impactor samples were taken ranging from 7 to 39 days in duration. Flow rates for both types of samples were one cu. ft. per minute. Sample volumes ranged from 1.01×10^4 to 5.62×10^4 cu. ft. The second filter was found wrinkled after the sampling period and may have allowed material to by-pass the filter. Interior surfaces and sampling plates of both cascade impactor samples were coated as previously described. The highest activity concentration measured (2.8 dpm per cu. ft.) in this study was found on the initial filter samples (taken 12/2/70 to 12/9/70). Values a tenth as high are calculated using the summation of activity on the impactor samples. Thus, the stream appears to have the greatest activity concentration of any measured. The flow from this system is small, only 0.05 percent of the total exhaust system, and thus the 26-inch vacuum system contributed a calculated 0.1 to 36.8 percent of the daily activity emitted as measured by stack monitors. Data from impactor samples on activity distribution into aerodynamically equivalent diameter size fractions vary by indicate a relatively fine aerosol. The M.A.D. values of 1.6 and less than 0.3 micron A.E.D. were found. In the latter samples, approximately 90 percent of the acitivity was found on the filter. The variation in size distributions may reflect the amount of moisture present as water droplets. Both distributions approximated a log-normal distribution of particles. Sufficient activity was present on all stages in the former sample to represent more than one PuO_2 particle of the stated size (see Table VI). The second impactor sample from this stream is similar to those previously discussed and did not indicate PuO_2 particles in the 3 coarsest size fraction.

ZONE 4 EXHAUST, 234-5Z BLDG. (309 FILTER ROOM)

Three filter and two impactor samples were taken from this location. Only one filter sample was usable -- the 4-inch glass fiber filter was torn soon after sampling was begun and the initial 8-inch by 10-inch glass fiber filter was lost in analysis. Sampling rate for the usable filter sample was 49 cu. ft. per minute during the 63 day sampling period for a total volume of 4.44×10^6 cu. ft. The impactor sampling rate was one cu. ft. per minute for 19.5 and 36 days respectively with total volumes of 2.81×10^4 and 2.88×10^4 cu. ft. Both 8-inch by 10-inch filters were completely black with shiny patches when retrieved. Calculated activity concentrations from filter and impactor samples were low, ranging from 4.5×10^{-6} to 10^{-3} dpm per cu. ft. The low value is calculated from the filter sample. The values calculated from the impactor samples may indicate re-entrainment of material from the floor[12,13] during entry for pump maintenance during sampling. The M.A.D. of both samples indicate a relatively coarse aerosol -- 9.5 and approximately 20 micron A.E.D. Both indicate log-normal distribution and have sufficient activity in all stages to represent a distribution of PuO_2 particles.

The activity concentration measured in the Zone 4 exhaust during these periods constituted up to 10 percent of the activity reported by daily stack samples. Since the zone 4 exhaust volume is some 18 percent of that emitted by the stack, this exhaust stream appears to aid in diluting other exhaust streams.

HF SYSTEM

A single set of filter and impactor samples was obtained during the period 12/7/70 to 1/17/71. Sample flow rate was one cu. ft. per minutes on both samples for the 36 day sampling period. Total volume of both samples was 5.18×10^4 cu. ft. Since the processing line served by the exhaust system was not in operation during the sampling period, the activity measured must be considered a system background level. The filter was found askew after sampling and the activity concentration indicated by this sample, 1.2×10^{-3} dpm per cu. ft., may be low. A higher value, 0.06 dpm per cu. ft., can be calculated using the activity measured in the impactor sample. This higher value appears more consistent with operating experiences (the HF System filters are among the first items checked in the event of high stack emissions). Even the higher activity level is only 2.3 percent of the activity emitted over the same period but, since the flow in the system is 0.2 percent of the total flow, the contribution of the exhaust stream may be significant when processing is resumed.

The activity distribution found for the material in this stream appears to be bi-modal indicating more than one particle formation (particles from process, corrosion in system, etc.). The M.A.D. was 7.4 micron.

AIR INTAKE

Three filter samples were taken of particles drawn into the building ventilation system. Flow through the 4-inch glass fiber filter was 72 cu. ft. per minute during the 13 day sampling period for a total volume of 1.32×10^6 cu. ft.

Two samples were obtained on 8-inch by 10-inch glass fiber filters at a flow of 53 cu. ft. per minute for periods of 15 and 34 days, with total volumes of air samples of 1.1×10^6 and 2.5×10^6 cu. ft., respectively. All filters collected were badly discolored by the particles deposited with the 4-inch filter especially heavily loaded with dust and insects. Activity measured on all samples was very low (see Table II) and indicated that 0.003 to 0.07 percent of the activity emitted from the stack was present in the air drawn into the ventilation system. Undoubtedly some of the activity present is normal background.

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