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EXPERIMENTAL PERFORMANCE EVALUATION
OF TWO STACK SAMPLING SYSTEMS IN A
PLUTONIUM FACILITY

J. A. Glissmeyer

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Pacific Northwest Laboratory
Richland, Washington 99352

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PREFACE

This report was written in 1977 to document work conducted during 1976 and 1977. The report was written for, and distributed to, the Atlantic Richfield Hanford Company, which, at that time, was the operating contractor responsible for the facilities studied. Because of this limited distribution, the report was never formally published or made publicly available. The current Hanford operating contractor, Westinghouse Hanford Company, has requested that the report be issued as a public document.

All efforts have been made to reproduce the document in its original format while remaining within the publishing guidelines established by the Pacific Northwest Laboratory (PNL). However, the report does contain references to internal operating procedures relevant to the operating contractor at the time the work was performed, which are no longer available, having been superseded over the intervening years. The report also contains numerous references to drawings in the Rockwell Hanford Operations Engineering Files. Copies of those drawings can be obtained by calling PNL Engineering Files (509) 376-0067.

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EXPERIMENTAL PERFORMANCE EVALUATION OF TWO STACK SAMPLING SYSTEMS IN A PLUTONIUM FACILITY

1.0 INTRODUCTION

The evaluation of two routine stack sampling systems at the Z-Plant plutonium facility operated by Rockwell International for USERDA is part of a larger study, sponsored by Rockwell and conducted by Battelle, Pacific Northwest Laboratories, of gaseous effluent sampling systems. The gaseous effluent sampling systems evaluated are located at the main plant ventilation stack (291-Z-1) and at a vessel vent stack (296-Z-3). A preliminary report, which was a paper study issued in April 1976, identified many deficiencies in the existing sampling systems and made recommendations for corrective action.⁽¹⁾ The objectives of this experimental evaluation of those sampling systems were as follows.

- Characterize the radioactive aerosols in the stack effluents.
- Develop a tracer aerosol technique for validating particulate effluent sampling system performance.
- Evaluate the performance of the existing routine sampling systems and their compliance with the sponsor's criteria.
- Recommend corrective action where required.

The tracer aerosol approach to sampler evaluation was chosen because the low concentrations of radioactive particulates in the effluents would otherwise require much longer sampling times and thus more time to complete this evaluation. The following report describes the sampling systems that are the subject of this study and then details the experiments performed. The results are then presented and discussed. Much of the raw and finished data are included in the appendices.

This study was initiated under the sponsorship of the Atlantic Richfield Hanford Company, which, at the time, was the operator of the above mentioned

facilities. The experimental program was completed and much of this report was written while ARHCO was the operating contractor. In June, 1977 the Rockwell International Company took over the operation of the subject facilities. Because this study evaluated the performance of equipment while ARHCO was the contractor, the term ARHCO is used throughout this report in reference to the facilities being tested. Since the conclusion of the experimental program, improvements to the subject effluent sampling systems were initiated by ARHCO and are being completed by Rockwell International.

2.0 SUMMARY

The evaluation of the radioactive particulate sampling systems at the 291-Z-1 and 296-Z-3 stacks, Z-Plant, consisted of two parts--a preliminary evaluation based on sampling theory⁽¹⁾ and this report on experiments conducted at the two stacks. The overall objectives of the study were to evaluate the compliance of the two sampling systems with Rockwell's Interim Criteria for Gaseous Sampling and Monitoring Systems (see Appendix I) and recommend corrective action where merited.

The experimental evaluation of the two sampling systems included the following tasks.

- Obtain locations and work platforms for representative sampling of stacks.
- Obtain velocity profiles at new sampling locations.
- Analyze stack effluents for trace levels of selected elements so a suitable tracer aerosol material could be chosen.
- Develop tracer aerosol technique.
- Check for aerosol channelling in stacks by measuring the tracer aerosol concentration at the locations to be used for comparison sampling.
- Evaluate the performance of the sampling systems for tracer aerosols by comparison against more representative samples taken from the new locations.
- Measure particle size distributions of plutonium bearing aerosols in the two stacks.

Tracer aerosols were used for the performance evaluation experiments because sampling for radionuclides would have required lengthier experiments because of their low concentration in the effluents. Tracer aerosols with mass median aerodynamic diameters of 1.3 and 8.5 μm were developed. The new locations used for collecting the samples for standards of comparison were at the 50-ft level of the 291-Z-1 stack and at the discharge of the 296-Z-3 stack.

The tracer concentration profile experiments showed only slight channelling of particulates in a limited cross section of the 291-Z-1 stack for the larger of the two tracer aerosols. At the 296-Z-3 stack slight channelling of both tracers along the area studied was found.

The plutonium bearing particles had activity mean aerodynamic diameters of 3.5 to 8.6 μm and 2.7 to 3.1 μm at the 291-Z-1 and 296-Z-3 stacks respectively. Because the performance of a sampling system generally decreases as particles become larger, the 291-Z-1 effluent presented the greater sampling challenge.

According to the comparison sampling experiment, the existing sampling system at the 291-Z-1 stack outperformed that at the 296-Z-3 stack. The samples extracted at the new locations made available for this study were used as standards of comparison. At both stacks the performance of the existing samplers was better for the finer than for the coarser tracer aerosol.

On the basis of these experiments, it was concluded that the two existing sampling systems do not comply with Rockwell's Interim Criteria for such systems. One requirement of the Criteria is that a sampling system collect 90% of a representative aliquot of 10 μm aerodynamic equivalent diameter* particles. There is, however, no clear definition as to what constitutes a satisfactory test of compliance.

The preliminary evaluation⁽¹⁾ pointed out other facets of the two sampling systems that do not satisfy other requirements of the Interim Criteria. The recommendations for corrective action given in that report are still valid and should continue to be carried out. Rockwell has taken some corrective measures already. The continuation of the development of the tracer aerosol validation technique is needed to be able to employ the technique routinely.

*AED, aerodynamic equivalent diameter, is the diameter of a sphere of density 1 g/cm^3 that behaves aerodynamically similar (has the same settling velocity) as the actual particle. For particles of diameter D and specific gravity relative to water sp.gr., the AED is roughly $D\sqrt{\text{sp.gr.}}$.

3.0 DESCRIPTION OF SAMPLING PROGRAMS

The 291-Z-1 stack exhausts filtered process enclosure and room ventilation air from the 234-5Z, 236-Z Plutonium Reclamation, 232-Z Incinerator, and 10 and 26-inch vacuum systems. The 296-Z-3 stack exhausts filtered cell and vessel ventilation air from the 241-Z Waste Sump and Storage Pit south of the 234-5Z Building. The exhaust flowrates through the 291-Z-1 and 296-Z-3 stacks are about 300,000 and 2,500 cfm respectively. Airborne particulates are suspected to be carriers of plutonium and americium and samples are extracted continuously from both stacks. The sample collection systems and sample analytical procedures will be described further.

3.1 291-Z-1 STACK SAMPLING SYSTEM

The system for collecting particulate samples from the 291-Z-1 stack is diagrammed in Figure 1. The sample is extracted downstream from the convergence of two ducts and just upstream from the entrance to the stack. The sampled air is extracted through three 2-in. pipe nipples and passes through a vertical length of 4-in. pipe, a horizontal length of 1 1/2-in. pipe, two valves and a piece of 3/8-in. O.D. Tygon tubing. The sampled particulates are collected on a 47 mm diameter Gelman^{*} Acropor 3 μ m pore filter. Sample flowrate is measured with a rotameter. The sample filter holder and the flowmeter are contained in the alpha monitor. During the course of our experiments the monitor was a Radeco Model 440 Alpha Monitor^{**} which was used to trigger high level release alarms. This monitoring system was described in greater detail in an earlier report.⁽¹⁾ The sample flowrate through the Radeco Monitor was 1.80 scfm. The Radeco Monitor has recently been replaced with an Eberline Alpha 3[†] and the new flowrate is about 1.4 scfm.

^{*}Gelman Instrument Company, 600 South Wagner Road, Ann Arbor, MI 48106.

^{**}Radeco/SAI, 4060 Sorrento Valley Blvd., San Diego, CA 92121.

[†]Eberline Instrument Corporation, P.O. Box 2108, Santa Fe, NM 87501.

FIGURE 1. Diagram of 291-Z-1 Stack Sampling System (Ref. 2-10)

The predicted performance in terms of sample bias caused by nonisokinetic* sample withdrawal and sample transport through the piping was given in the earlier report.^(1, pg 10-12) The nonisokinetic sample bias was calculated assuming all the sample was extracted through one nozzle and the bias caused by particle deposition was calculated assuming a flowrate of 2 cfm at 70°F, 29.92 in Hg. The effects of pipe fittings, valves and the side withdrawal of the sample from the 4-in. pipe (which may behave like a virtual impactor) were not considered. The predicted sample bias was intended to serve as an indicator of sampling problems and showed little bias for aerosols in the 1-2 μ m aerodynamic equivalent diameter (AED)** range and significant bias for particles larger than 5 μ AED.

3.2 296-Z-3 STACK SAMPLING SYSTEM

The 296-Z-3 stack is sketched in Figure 2 and the sampling system is diagrammed in Figure 3. The exhaust air temperature is about 153°F and the flowrate is about 2520 cfm (or 2190 scfm). The effluent sample is extracted from the centerline of the stack through a single nozzle probe. The air sample passes through six-90° bends, a flowmeter and a valve to a Hanford designed filter holder.⁽¹¹⁾ Particulates are collected on a 4-in. diameter asbestos and cellulose fiber filter paper[†] which is supported in the filter holder. The sample flowrate is about 1.5 scfm. The sampler piping is electrically heat traced to the sampler enclosure;⁽¹²⁾ however, there is no thermostatic control. The performance of the sample collection system was estimated in the earlier report^(1, pg 24-25) and was only slightly better than the 291-Z-1 stack sampler.

3.3 SAMPLE ANALYSIS PROCEDURE

The airborne particulate samples from both stacks are changed at least once each working day. If the alpha monitor at the 291-Z-1 stack indicates an

*Extracting a sample at a velocity different than the gas velocity approaching the nozzle orifice.

**The diameter of a sphere with density 1 g/cm³.

[†]Type HV-70, Hollingworth and Vose Co., 112 Washington Street, East Walpole, MA 02032.

FIGURE 2. 296-Z-3 (241-Z) Continuous Stack Sampler(13,14,15)

FIGURE 3. 296-Z-3 (241-Z) Continuous Stack Sampler(13,14,15)

unusual release the sample is changed more often. The sample filters are counted first in the 234-5Z lab for process control purposes.⁽¹⁶⁾ The "official" sample analysis is performed in the 222-S laboratory. Each sample is counted twice for one minute, the first count at least 6 hr after sample changing and the second count at least 4 hr later. The net long-lived alpha emitters are then calculated from the two counts. The counting instrument is a Beckman Wide Beta II Analyzer* which analyzes for gross beta and gross alpha emitters. The total alpha curies released during the period covered by a sample are calculated by Equation 1.

$$\text{Curies released} = \text{net } \frac{c}{\text{min}} \times \frac{C_i - \text{min}}{2.22(10^{12})_d} \times \frac{1}{0.7} \times \frac{\text{Stack Flowrate}}{\text{Sample Flowrate}} \quad (1)$$

$$\times \frac{d}{0.296 \text{ c}}$$

A sample filter collection efficiency of 70% and a gross alpha counting efficiency of 0.296 is assumed in the calculation. For total beta curies a counting efficiency of 0.383 is used.^(17,18) These calculations are performed by the AIRSAM computer program.

*Beckman Instruments Inc., P.O. Box C-19600, Irvine, CA 92713.

4.0 EXPERIMENTAL

The four experiments conducted at both the 291-Z-1 and 296-Z-3 stacks were:

- Background sampling for alpha emitters and candidate tracer materials,
- Tracer aerosol concentration profile at sampling locations,
- Comparison sampling - sampling for tracer simultaneously with both existing and control sampling systems, and
- Alpha emitter particle sizing.

Before performing these experiments some preliminary measurements were required. These will be described first and then the experiments will be described in turn.

4.1 PRELIMINARY MEASUREMENTS

Certain measurements were required at the locations available for sampling at each stack before proceeding with the other experiments. At the 291-Z-1 stack an opening at the 50-ft level on the South side was used for sampling access. Access was available at the stack base but it was felt that it was more likely to obtain representative samples farther up the stack and without too much difficulty. A scaffold was constructed with a covered work area at the top. At the 296-Z-3 stack, scaffolding was placed so samples could be taken at the stack discharge. Velocity profile measurements were made initially by ARHCO's Ventilation Balance Subsection. An Anemotherm Model 60 Thermal Anemometer^{*} was used at the 291-Z-1 stack and a pitot tube was employed for velocity measurements at the 296-Z-3 stack. Velocity readings fluctuated (especially at 291-Z-1) so average readings were recorded. The air velocities at sampling points were periodically checked with a Datametrix Model 800 VTP Airflow Multimeter.^{**} Both the Anemotherm and Datametrix anemometers were checked against a pitot tube in a wind tunnel.

^{*}Anemostat Corporation of America, 10 East 39th Street, New York 16, NY.

^{**}Datametrix is now a subsidiary of Gould Inc., Control and Systems Division, 340 Fordham Road, Wilmington, MA 91887.

Psychrometric data on each stack airflow was obtained with a sling psychrometer; however, the dry bulb temperature range was not sufficient for the 296-Z-3 effluent. At the 296-Z-3 stack the dry bulb temperature was obtained with a mercury thermometer and with a Minigraph Temperature Recorder.*

4.2 BACKGROUND SAMPLING

To estimate the concentration of alpha emitters and the background of candidate tracers in the effluents a series of samples were collected from each stack. The tracer aerosol was chosen on the basis of the data from this experiment.

Samples were collected using in-stack filter holders with the flowrate controlled to maintain isokinetic sample extraction. Figure 4 is a schematic diagram of the sampling equipment. Sampled particulates were collected on 47 mm diameter MF-Millipore cellulose ester membrane filters with a mean pore size of 5 μ m.** The membrane filter was backed-up with a 47-mm diameter glass fiber filter.*** Sample flowrate was measured with a Fisher-Porter Series 10A-3000 Rotameter.† Samples were collected for varying lengths of time. The sample flowrates were 1.2 and 1.5 scfm at the 291-Z-1 and 296-Z-3 stacks respectively. At the 291-Z-1 stack the filter holder was positioned 16 in. from the south wall by the probe shown in Figure 5. An exploded view of the filter holder-nozzle assembly is shown in Figure 6. At the 296-Z-3 stack the filter holder was positioned on the centerline and about 1 ft below the discharge. The filter holder was supported on a vertical tube which was suspended from a crossbar. The sampling system flowrates were calibrated in the lab using a calibrated venturi tube†† placed at the inlet nozzle of the assembled sampling equipment. Laboratory conditions were a satisfactory approximation to conditions inside the 291-Z-1 stack. At the 296-Z-3 stack there was a systematic error in

*Model M34A, Esterline Angus Instrument Corporation, P.O. Box 24000, Indianapolis, IN 46224.

**MF-Millipore Type SMWP 04700, Millipore Corp., Bedford, MA 01730.

***Gelman Type A, Gelman Instrument Company, 600 South Wagner Road, Ann Arbor, MI 48106.

†Fisher & Porter, Warminster, PA 18974.

††Radeco Model C812, Radeco/SAI, 4060 Sorrento Valley Blvd., San Diego, CA 92121.

FIGURE 4. Schematic Diagram of Background Sampling Equipment

the flowrate measurement because the air temperature was about 150°F; however, for the qualitative purpose of this experiment this flowrate control was sufficient.

Both the membrane and back-up filters from each sample were analyzed by X-ray fluorescence for traces of titanium, manganese, iron, nickel, copper, zinc and tin by BNW's Chemical Methods and Kinetics Section. Each sample was then dissolved and analyzed for total plutonium content (^{239}Pu + ^{240}Pu) by BNW's Technical Analysis Section as outlined in Appendix B. In addition, blank samples of the following filter media were analyzed by the X-ray fluorescence procedure for content of the same elements mentioned above: Gelman Type A Glass Fiber, Gelman Acropor AN3000, Hollingsworth & Vose HV-70, MF-Millipore SMWP, IPC #1478 oil treated.* The following blank filter media were analyzed for titanium and zinc only: Gelman Type E Glass Fiber, Nuclepore 6 μm pore size,** Millipore Mitex 5 μm pore size and Millipore Fluopore with 1 μm pore size. The Gelman Metrice1 VM-1 filter with 5 μm pores was analyzed for titanium and iron.

*The Institute of Paper Chemistry, Appleton, WI.

**Nuclepore Corporation, Pleasanton, CA 94577.

FIGURE 5. Probe for Background Sampling at 291-Z-1
(PNL Neg. No. 757249-1)

FIGURE 6. Exploded View of Filter Holder Assembly. Left to right: tightening ring, nozzle, membrane filter, support screens, filter holder.
(PNL Neg. No. 757249-3)

4.3 TRACER CONCENTRATION PROFILE

The purpose of this experiment was to characterize the concentration profile at the chosen sampling locations at each stack using a tracer aerosol with a small particle size. We used the titanium dioxide-zinc stearate blended aerosol described in Appendix A. (The zinc sulfide aerosol was not ready at this time.) The aerosol was generated using a Wright Dust Feed Mechanism^{*} and injected into the effluent airstream upstream of the exhaust fans to obtain the best possible mixing. The tracer aerosol was then sampled simultaneously at three positions along the available stack diameter. The titanium concentrations determined from the different locations were then compared to indicate how well mixed the aerosol was with the effluent. Based on the results of this experiment the sampling position to be used for the comparison sampling experiment was to be chosen.

4.3.1 291-Z-1

At the 291-Z-1 stack we originally intended to sample simultaneously at six positions on the North-South diameter of the stack at the 50-ft level; however, the number of positions was reduced to three because there was inadequate power for the six vacuum pumps. Figure 7 shows the layout of the sampling positions and Table 1 lists the data pertinent to each position. The sampling positions were located in the regions of high, average, and low velocities (numbers 6, 4 and 1 respectively) along the stack diameter. The chosen diameter roughly bisects the velocity profile. Figure 8 shows the tracer injection and sampling locations and Figure 9 shows the Wright Dust Feeder positioned to inject tracer aerosol into the ventilation duct near the 234-5Z Building.

Each of the three samplers was assembled as shown in Figures 4 and 6 and used the same filters as in the background sampling experiment. The flowmeter set points were also calibrated as in the background sampling experiments except a dry test meter^{**} was used instead of the venturi meter. The accuracy

^{*}L. Adams Ltd., London, England, Importer BGI Inc., Waltham, MA 02154. (19)

^{**}American Dry Test Meter Model DTM-115-2, Singer, American Meter Division, Burlingame, CA 94010.

FIGURE 7. Sampling Positions at 291-Z-1 Stack
50-ft Level

TABLE 1. 291-Z-1 Concentration Profile Samplers

	Position		
	1	4	6
Avg. Air Velocity, ft/min	~1300	~1500	~1650
Nozzle Inlet Diam., in.	.382	.355	.350
Isokinetic Flowrate, scfm	1.03	1.07	1.10
Flowmeter	Dwyer RMC-102	Fischer-Porter 10A-3000	Fischer-Porter 10A-3000
Flowmeter Setting	86 scfh (1.43 scfm)	1.37 scfm	1.34 scfm
Exposed Filter Area (as determined from nozzle outlet diameter), cm ²	10.07	9.80	10.24

of flowrate control was probably $\pm 10\%$. The filter holders were suspended in the airflow by a length of unistrut as shown in Figure 10 (the flowmeter instrumentation in the figure is that used for the comparison sampling experiment, however).

FIGURE 8. Photograph Showing 234-5 Z Building, Tracer Injection Point and Sampling Location on 291-Z-1 Stack (PNL Neg. No. 769230-1)

FIGURE 9. Wright Dust Feeder Positioned to Inject Tracer Into Ventilation Dust from 234-5 Z Building
(PNL Neg. No. 771455-7)

FIGURE 10. Sampling Probe Used for Concentration Profile and Comparison Sampling Experiments at 291-Z-1 Stack; Flowmeter Instrumentation Shown Was for the Comparison Sampling Experiment (PNL Neg. No. 770376-2)

Runs of the experiment lasted 23-72 hr; however, tracer injection took 3-4 hr and 31-33 g of tracer were injected. Both the membrane and glass fiber filters were analyzed for titanium using the X-ray fluorescence technique outlined in Appendix B. Blank membrane and glass fiber filters were analyzed with each run. Eleven runs were completed; however, the data from the first six runs were discarded because the filter deposit area (an important parameter for converting the analysis result from $\mu\text{g}/\text{cm}^2$ to total μg) was inaccurately controlled.

4.3.2 296-Z-3

The tracer concentration profile experiment at the 296-Z-3 stack proceeded generally the same way as at the other stack but with some refinements in the areas of flowrate control and sample analysis.

The titanium dioxide-zinc stearate tracer described in Appendix A was injected using the Wright Dust Feeder as shown in Figure 11 and sampled at the point shown in Figure 12. A schematic diagram of one of the three sampling systems is shown in Figure 13 and Table 2 lists the pertinent sampler data such as flowrate, nozzle diameter, etc. The sample filter holders were assembled as shown in Figure 6 and supported as shown in Figure 14. The samplers were arrayed as sketched in Figure 15 along a stack diameter which bisected the flow profile. The sampling equipment in place is shown in Figure 16.

Six runs of this experiment were completed successfully. During each run flowrates were adjusted by observing the vacuum pressure and temperature at each flowmeter and calculating the flowmeter setpoint using Equation 2 where atmospheric pressure was approximated as 30-in. Hg, P was the vacuum pressure (in. Hg) and T was the Temperature ($^{\circ}\text{F}$).

$$\text{Flowmeter Set Point} = \text{Desired scfm} \sqrt{\frac{30}{30-P}} \times \frac{460 + T}{530} \quad (2)$$

After the stable flowmeter setpoint was achieved tracer injection was begun. During the injection of the tracer the flowmeters were adjusted as needed. Tracer injection lasted 1 hr, total sampling run time lasted from 74 to 97 min and 2.9 - 3.2 g of tracer were injected.

FIGURE 11. Wright Dust Feeder In Position to Inject Tracer Aerosol
(PNL Neg. No. 771455-4)

FIGURE 12. 296-Z-3 Stack Showing Tracer Injection and Sampling Points
(PNL Neg. No. 770376-6)

FIGURE 13. Schematic Diagram of Sampling Equipment Used for Concentration Profile and Comparison Sampling Experiments

TABLE 2. 296-Z-3 Concentration Profile Samplers

	Position		
	1	2	3
Ave. Air Velocity, ft/min	2190	2330	2600
Nozzle Inlet Diam, in.	0.367	0.352	0.344
Isokinetic Flowrate, cfm	1.61	1.57	1.67
scfm	1.39	1.36	1.44
Exposed Filter Area (as determined from nozzle outlet diameter) cm ²	9.80	10.36	10.24

TABLE 2. 296-Z-3 Concentration Profile Samplers

FIGURE 14. Support For Sampler Filter Holders
(PNL Neg. No. 770376-5)

FIGURE 15. Layout of Samplers at 296-Z-3 Stack for Concentration Profile and Comparison Sampling Experiments

FIGURE 16. 296-Z-3 Stack Sampling Equipment for Concentration Profile and Comparison Sampling Experiments
(PNL Neg. No. 770376-3)

At the conclusion of each run the filters were removed from the filter holders and each nozzle interior was washed with concentrated sulfuric acid. Analyses were performed by BNW's Chemical Methods and Kinetics Section. Each nozzle wash was neutralized and the precipitate was collected on a MF-Millipore filter. These wash filters and the other sample filters were analyzed as outlined in Appendix B. Blank membrane and glass fiber filters were submitted for analysis with each run as controls.

4.4 COMPARISON SAMPLING EXPERIMENT

The purpose of this experiment was to assess the performance of ARHCO's existing sampling systems by comparing samples collected by the existing systems with samples collected by control sampling systems. Background samples showed that the concentrations of radioactive particulates in both stacks were quite low so it was decided to use tracer aerosols for this experiment to reduce the sampling time. Two tracer aerosols were developed for this experiment. One tracer was the titanium-dioxide (TiO_2) aerosol with a mass mean aerodynamic diameter (MMAD) of $1.3 \mu\text{m}$ used in the concentration profile experiment. The other was a zinc sulfide (ZnS) based aerosol with a MMAD of 8-9.5 μm . (Both aerosols are described in Appendix A and were generated using the Wright Dust Feed Mechanism as explained in Appendix A.) The use of the two tracers enabled the evaluation of the existing sampling systems' performance with particulates of two particle size distributions and it was anticipated that the performance test would be more rigorous with the ZnS than with the TiO_2 aerosol. Both particle size distributions were within the range of typical radioactive particulates encountered in such facilities. (20,21)

The samples to serve as standards for comparison were obtained from the locations at the 50-ft level of the 291-Z-1 stack and the discharge of the 296-Z-3 stack as already described. The initial results of the concentration profile experiment indicated that the concentration of the TiO_2 aerosol was not uniform along the sampled diameters. For this reason we decided to collect the control samples from the same locations used for the concentration profile experiment. The sampling method was also the same as with the concentration profile experiment but with some refinements.

The components of each sampler were arranged as shown in Figure 13 and the filter holders were assembled as shown in Figure 6. Each sample was collected in the stack with the filter holders and nozzles positioned as shown in Figures 7 and 15. The assembled sampling equipment is shown in Figures 10, 14 and 16. Stack air velocities were rechecked at the sampling points with the Datametrics 800 VTP Airflow Multimeter and some new sampler nozzles were made. Sampler measurements and flowrates for the comparison sampling experiment are listed in Table 3. Tracers were injected as in the concentration profile experiment (see Figures 8, 9, 11 and 12) and as described in Appendix A.

TABLE 3. Sampler Data For Comparison Sampling Experiment

a. 291-Z-1 Stack:

	Position			
	<u>1</u>	<u>4</u>	<u>6</u>	<u>ARHCO</u>
Ave. Air Velocity, ft/min	1211	1512	1514	
Nozzle Inlet Diam. in.	0.358	0.356	0.357	
Isokinetic Flowrate, scfm	0.83	1.03	1.03	
Exposed Filter Area, cm ²	10.02	10.29	10.20	14.37
Ave. Flowrate, scfm*	0.92	1.04	1.06	1.80

b. 296-Z-3 Stack:

	<u>1</u>	<u>2</u>	<u>3</u>	<u>ARHCO</u>
Ave. Air Velocity, ft/min	2190	2330	2600	
Nozzle Inlet Diam., in.	0.367	0.352	0.344	
Isokinetic Flowrate, cfm	1.61	1.57	1.67	
scfm	1.40	1.36	1.45	
Exposed Filter Area, cm ²	9.80	10.36	10.25	57.17
Ave. Flowrate, scfm*	1.38	1.34	1.41	1.53

*These flowrates were measured with a calibrated mass flowmeter.

Flowrate control for Battelle's samplers was accomplished in the same way as described in the concentration profile experiment at the 296-Z-3 stack. It was originally intended to precalibrate each sampler with a mass flowmeter; however, its receipt was delayed several months. Instead, each sampler flowrate was measured with the calibrated mass flow meter^{*} after the experiments were completed and under actual or simulated operating conditions. The average flowrates measured are recorded in Table 3 and the maximum variation was $\pm 5\%$. These values were used for data reduction.

The experimental runs and analyses were performed in about the same manner as the concentration profile experiments already described. The samples from ARHCO's existing systems were collected on the same filter media as used in the control samplers.** ARHCO's samples were collected simultaneously with the control samples. ARHCO's samples were then analyzed in the routine way and set aside for the analysis of tracer content. The control sampler nozzles were washed with 6N H_2SO_4 after each run. In some instances the nozzle washes from the three samplers were combined for analysis and in other cases they were analyzed separately. The X-ray fluorescence analytical procedure is detailed in Appendix B. Three replicate analyses were made of each sample at randomly chosen spots on the filter. The back-up glass fiber filters were not analyzed because the experience from the concentration profile experiment showed no detectable tracer collection on that filter. The technique for precipitating tracer from the nozzle washes is also detailed in Appendix B.

4.5 PARTICLE SIZING

Particle size distribution data for both effluents was needed to better interpret the results of the comparison sampling and concentration profile experiments. The particulates of main interest in this study were those emitting alpha radiation which we expected to be caused by traces of plutonium on particles. No effort was made to characterize the source or chemistry of

*The mass flowmeter consisted of the following components: Sierra Model 500-8 Mass Flow Transducer, Model 500-PSL Power Supply/Linearizer, and Model 439 Digital Readout, Sierra Instruments Inc., Carmel Valley, CA 93924.

**MF-Millipore, SMWP, 5 μm pore size.

the particulates. As will be described, particulate sizing was performed using high flowrate cascade impactors at both stacks and also a low flowrate cascade impactor at the 296-Z-3 stack.

4.5.1 High Flowrate Impactor--291-Z-1

At the 291-Z-1 stack an Anderson Model 65-800^{*} was used to obtain size fractionated particulate samples of the effluent. This impactor operates at a flowrate of 20 scfm and is shown in exploded view in Figure 17. The cascade impactor is a series of stages, each composed of a jet plate and a glass fiber impaction surface. The stages are followed by a final back-up Gelman Type A glass fiber filter. The sampled air passes through the series of jet plates where particulates collect on the impaction surfaces and on the back-up filter. Because the jet dimensions are different for each stage the particulates are fractionated between the impaction surfaces according to their size and aerodynamic behavior. Each stage has a characteristic aerodynamic equivalent cutoff diameter and the material collected on the impaction surface will have a mean aerodynamic equivalent diameter (AED) equal to that cutoff diameter. The back-up filter collects the particles that are smaller than the cutoff diameter of the last stage. With analyses of the material collected on each impactor surface and the back-up filter the aerosol particle size distribution can be reconstructed in terms of AED.

The assembled cascade impactor and the sampling probe are shown in Figures 18 and 19. The sampling probe was fabricated to have a smooth bore to the impactor. The probe had a 1.610 in ID nozzle and was about 40 in. long. The nozzle was positioned at sampling location #6 (as described earlier) because the initial data from the concentration profile experiment showed that the tracer concentration there was the average of the three locations. Flowrate control was maintained at 20 scfm by a Sierra Model 310 High Volume Flowrate Controller and the flowrate for some of the runs was recorded on a strip chart recorder. The air mover was a standard high volume blower.

^{*}Anderson 2000 Inc., P.O. Box 20769, Atlanta, GA 30320.

FIGURE 17. Exploded View of Anderson Model 65-800 Four Stage High Volume Cascade Impactor
(PNL Neg. No. 771455-15)

FIGURE 18. Assembled Cascade Impactor Sampler for 291-Z-1 Stack
(PNL Neg. No. 771455-12)

FIGURE 19. High Flowrate Impactor In-Place at 201-Z-1 Stack
(PNL Neg. No. 769230-7)

Three runs of this experiment were completed, each of them lasting about two weeks. After each run the impactor was disassembled and each impaction surface and filter was folded inward, labeled, and placed in a container. Impactor jet plate surfaces adjacent to each impaction surface were wiped with tissues soaked in 6N-nitric acid and the tissues were added to their respective collection surfaces. Probe and other impactor inlet component interiors were washed with 6N-nitric acid and wiped with HNO_3 soaked tissues. These washes and wipes were combined and the tissues were dissolved as the total wash volume was reduced by evaporation. The wash solution and the combined tissues and glass fiber impaction surfaces were analyzed for total plutonium content as detailed in Appendix B. ARHCO's routine samples taken concurrently with runs #2, #3 (except for eight missing hours) were also analyzed for total plutonium.

4.5.2 High Flowrate Impactor--296-Z-3

The Anderson Model 65-800 high flowrate cascade impactor was used to obtain particle size information at the 296-Z-3 stack also. The impactor assembly used is shown in Figure 20. Three runs were performed using the impactor and the procedure was similar to that described for the 291-Z-1 stack. The high volume flowrate controller was available in time to be used only on the third run. For the first two runs a U-tube manometer was used to measure the pressure differential between the ambient pressure and vacuum pressure behind the impactor. The pressure reading set point was obtained in the lab at a flow of 20 scfm using a calibrated venturi* placed at the nozzle inlet. The setpoint was corrected for the elevated stack temperature. The inlet probe was a short, smooth bore nozzle with an opening of 1.33 in. ID placed on the centerline of the stack (position #2). Sample handling was the same as for the other stack, except that there were no tissue wipes for the first run. ARHCO's routine samples corresponding to the impactor run #1 were the only ones available for the total plutonium analysis by our lab.

*Radeco Model C8528 Calibrated Air Flow Indicator.

FIGURE 20. High Flowrate Cascade Impactor In-Use at 296-Z-3 Stack
(PNL Neg. No. 771455-2)

4.5.3 Low Flowrate Impactor--296-Z-3

One run using a Sierra Model 226 Source Cascade Impactor was made at the 296-Z-3 stack to gain additional particle size information for plutonium bearing particles. This cascade impactor operates on the same principle as the high flowrate impactor but it has six fractionating stages, operates at a low flowrate and is placed entirely in the stack airflow. Figure 21 is an exploded view of this impactor and Figure 22 shows the assembled impactor. Figure 23 shows the impactor positioned on the stack centerline. The flowrate through the impactor was 0.87 scfm. Flowrate was controlled by a Sierra Model 250-10 Constant Flow Air Sampler (shown in Figure 24) which was calibrated with the Sierra Model 508 Mass Flowmeter. The equivalent cutoff diameters for each stage were calculated for the flowrate as directed by the impactor manufacturer. The one run lasted nearly 19 days. When the impactor was retrieved we found that it had been pushed over to one edge of the stack since it was last checked. This may lessen the reliability of the sample somewhat since the aerosol concentration profile was not found to be uniform. The impactor samples were prepared and analyzed in the same manner as was explained for the 291-Z-1 stack particle sizing experiment. The sample analysis was for total plutonium. ARHCO's routine samples collected concurrently were also analyzed by the same procedure for total plutonium. The analytical procedure was as detailed in Appendix B.

4.6 SCHEDULE OF EXPERIMENTS

The experiments described were conducted from January 21, 1976 to February 19, 1977. Figure 25 shows the time periods during which samples were collected.

FIGURE 21. Exploded View of Low Flowrate Cascade Impactor
(PNL Neg. No. 771455-14)

FIGURE 22. Assembled Low Flowrate Cascade Impactor
(PNL Neg. No. 771455-3)

FIGURE 23. Low Flowrate Cascade Impactor In Position on 296-Z-3 Stack
(PNL Neg. No. 771455-10)

FIGURE 24. Sierra Model 250-10 Constant Air Flow Controller Panel

FIGURE 25. Time Periods During which Samples were Collected

5.0 RESULTS

The results of the experiments will be presented in the same order as they were described.

5.1 PRELIMINARY MEASUREMENTS

The initial velocity profiles of the 296-Z-3 and 291-Z-1 stacks are shown in Appendices C.1 and C.2. In both stacks the highest velocities were located off-center and the profiles were roughly symmetrical. Measurements taken later at selected points in the 291-Z-1 stack are recorded in Appendices C.3 and C.4.

The velocities in the 296-Z-3 stack were relatively constant but the velocity at any given point in the 291-Z-1 stack rapidly varied over a wide range so that the readings recorded were only estimates of the average. An example of this is shown in Appendix C.4 which lists the final measurements taken in January 1977. The readings recorded in March 1976 were used in designing the concentration profile experiment and the readings of October 1976 were used in the design of comparison sampling and particle sizing experiments (Appendix C.3). The flowrates of the 291-Z-1 and the 296-Z-3 stacks were respectively about 302,000 and 2,520 cfm (300,000 and 2,190 scfm). The relative humidities of the 291-Z-1 and 296-Z-3 stacks were 54% and 8% yielding dewpoints of 53°F and 44°F respectively.

5.2 BACKGROUND SAMPLING EXPERIMENT

To obtain data on the background concentration of selected elements four samples were collected from each stack and the raw data are listed in Appendix D.1. The measurable concentrations of elements are listed in Appendix D.2. There were measurable concentrations of iron, zinc, tin and plutonium in the 296-Z-3 stack and iron, copper, zinc, tin and plutonium were found in the 291-Z-1 stack. The concentration of iron in the 291-Z-1 stack was an order of magnitude greater than at the 296-Z-3 stack. These four and subsequent samples from the 291-Z-1 stack contained visible particles which appeared like rust.

The level of plutonium (^{239}Pu and ^{240}Pu) at the 296-Z-3 stack was fairly constant over this period (February-March 1976) and ranged from 2.0 - 2.7 (10^{-14}) $\mu\text{Ci/std. ml.}$ In contrast, the plutonium concentration varied widely over the same period at the 291-Z-1 stack; from 3.4(10^{-14}) to 2.5(10^{-12}) $\mu\text{Ci/std. ml.}$ For comparison, the Table I limit of the ERDA Operations Manual Chapter 0524, Appendix, Annex A is 2×10^{-12} $\mu\text{Ci/ml.}$

Titanium and manganese were not detectable in either stack. They were considered prime candidates for tracer aerosols but a suitable compound of manganese was not found. Although there was some zinc present in both stacks, a zinc sulfide tracer was developed and in use its concentration was 3-4 orders of magnitude above that of the background zinc.

5.3 TRACER CONCENTRATION PROFILE EXPERIMENT

The raw data for the concentration profile experiments described in Section 4.3 are listed in Appendices E.1a and E.1b. The concentration of titanium at each location and for each run was calculated as detailed in Appendix E.1c.

A two-way analysis of variance was performed using the concentration data from each stack. For both stacks this analysis showed that on the average there were significant differences between the tracer concentrations at each location, although the differences were small as shown in Table 4.

TABLE 4. Results of Original Concentration Profile Experiment

	<u>Location</u>	<u>Mean Titanium Concentration $\mu\text{g/std. ft}^3 \times 10^{-2}$</u>
291-Z-1 Stack 5-runs	1	2.69
	4	3.63
	6	3.49
296-Z-3 Stack 6-runs	1	7.28
	2	7.65
	3	6.54

On the basis of these data we decided to use the same three sampling locations at each stack rather than just one location per stack to collect our samples for the comparison sampling experiment.

Because the comparison sampling experiment was so similar to the concentration profile experiment the results provided more data on the concentration profiles in the stacks. Data was also available for a tracer with a larger average particle size; i.e., zinc sulfide. Only those samples for which the nozzle washes were individually analyzed (Wash Code = 3) rather than combined provided data useful for this experiment. This additional data for the TiO_2 and ZnS tracers are listed in Appendices E.2a-b. The tracer concentrations were calculated as shown in Appendix E.2c from the raw data listed in Appendix F. Combining this data with the original data yields the mean concentrations tabulated in Table 5.

TABLE 5. Mean Tracer Concentrations at Various Locations in 291-Z-1 and 296-Z-3 Stacks

Stack	Sampling Location	Tracer			
		TiO_2 MMAD = 1.3 μm		ZnS MMAD = 8 - 9.5 μm	
		No. of Runs	$\mu\text{g}/\text{std. ft}^3$	No. of Runs	$\mu\text{g}/\text{std. ft}^3$
291-Z-1		10		4	
	1		0.185		0.157
	4		0.208		0.185
	6		0.209		0.168
	Grand Mean		0.201		0.170
296-Z-3		15		6	
	1		8.50		7.53
	2		8.47		9.00
	3		7.87		7.20
	Grand Mean		8.28		7.91

A two-way analysis of variance for the TiO_2 data at the 291-Z-1 stack showed no significant difference between the mean concentrations at the three locations; however, similar analyses showed that the differences in concentrations were significant in the other three cases. Although the concentrations were not very different from the grand mean in any instance (14% at most) the differences were consistent enough in three of the four cases to be statistically significant. As expected, the differences between locations were more pronounced for the ZnS tracer than for the TiO_2 because of the larger mean particle size of the ZnS.

5.4 COMPARISON SAMPLING EXPERIMENTS

The raw data for the comparison sampling experiments are listed in Appendices F.1-F.4. These appendices list the duration, amount of tracer used and the sample analyses for each run. Appendix F.5 lists the analyses of blank nozzle wash solutions which were used to obtain background levels of the tracer elements in the wash solutions. The averaged background levels were subtracted from the wash analyses to compensate for trace contaminants in the solutions. The calculated tracer concentrations measured with Battelle's and ARHCO's samplers for each run are listed in Appendices F.6-F.9. For each run the amount of tracer that should have been collected by each sampler was estimated and the percent recovery was then calculated. The ratios of the recovery as measured with Battelle's and ARHCO's samplers were calculated as well as the average ratio, the standard deviation of the ratios, the standard deviation of the mean, average recoveries and the ratios of averaged recoveries. These calculations were made for each stack and each tracer aerosol and the results are tabulated in Appendices F.6-F.9. A computer program, COMEXP, was written to perform most of the calculations using the data from the more than 900 analyses and measurements. The calculational procedures are detailed in Appendix F.10 and the outputs formed the basis of Appendices F.1 to F.9.

The results of the comparison sampling experiments are summarized below in Table 6. The "recovery performance ratio", which is equivalent to the

ratio of concentrations, was calculated for each run and the average for each experiment is shown in the table. These ratios do not tell the whole story and it is not recommended that they be used to correct past calculated emission values. As shown in Appendices F.6-F.9, the spread in the ratios for the runs in an experiment was considerable in three of the four experiments. The standard deviations of the means were also large for the same three experiments.

TABLE 6. Summarized Results of Comparison Sampling Experiments (See Appendices F.6-F.9)

<u>Stack</u>	<u>Tracer</u>	<u>Runs</u>	<u>Average % Recovery For Sampler</u>		<u>Recovery Performance Ratio Battelle/ARHCO</u>	
			<u>Battelle</u>	<u>ARHCO</u>	<u>Mean, S_x All Runs</u>	<u>Std. Deviation of Mean, S_x</u>
291-Z-1	TiO ₂	9	87	82	1.07	0.041
	ZnS	10	33	3	20.3	6.6
296-Z-3	TiO ₂	11	76	14	5.86*	3.4*
	ZnS	11	38	1	46.7	5.0

*Excludes Run 4 which appears to be an outlier for which we have no apparent explanation.

5.5 PARTICLE SIZING RESULTS

The raw data for the particle sizing experiments at both stacks are listed in Appendices G.1 and G.2. Included in those appendices are the data for the corresponding samples collected by ARHCO's existing sampling systems and later analyzed by the same method as the impactor samples. The results were in terms of d/min. of total plutonium and the concentrations were calculated as $\mu\text{Ci}/\text{std. ml.}$

The method for reducing the cascade impactor data to particle size distributions is detailed in Appendix G.3. The cumulative percent of plutonium versus stage cutoff particle size is plotted on log-normal

probability paper. The size distributions for the runs at the 291-Z-1 and 296-Z-3 stacks are shown in Figures 26 and 27. The intersection of the size distribution curve with the 50% line indicates the activity mean aerodynamic diameter, AMAD, for the distribution. Fifty percent of the plutonium was associated with particles larger than the AMAD and 50% with particles smaller than the AMAD. The slope of the size distribution curve is related to the geometric (activity) standard deviation. The indicated AMAD's for each run are listed in Appendices G.1 and G.2 and in Figures 26 and 27.

As indicated in Appendices G.1 and G.2 a significant portion of plutonium entering the samplers was deposited in the probe and inlet parts of the impactor during many of the runs. Because the larger particles tend to deposit in the probe and impactor inlet the size distributions may be more coarse than indicated by the impactor data. An extreme estimate of the largest AMAD for each run can be found by adding the deposited activity to the first impactor stage and reducing the data in the same manner as before. The AMAD's calculated when the inlet losses are both ignored and included are listed in Appendices G.1 and G.2 and summarized in Table 7. Where the percent deposited in the inlet was largest the difference between the AMAD's calculated both ways was the greatest. Because the probe configuration used at the 296-Z-3 stack was simpler than that used at the 291-Z-1 stack the percent activity deposited in the inlet was lower for the runs made at the 296-Z-3 stack. The actual AMAD's are in between the two values; however, the lower values will be used in discussing the size distributions later in the report.

The data for Run 3 at the 291-Z-1 stack may not be reliable because about one-third of one sample was lost during lab work and the resulting analysis was only an estimate. The data from the other two runs were comparable to data obtained several years ago by Mishima⁽²⁰⁾ at the 291-Z-1 stack.

FIGURE 26. Plutonium Bearing Particle Size Distribution in 291-Z-1 Stack

FIGURE 27. Plutonium Bearing Particle Size Distribution in 296-Z-3 Stack
*Low Flowrate Impactor

TABLE 7. Summarized Activity Mean Aerodynamic
Diameters for Plutonium Bearing Particles

<u>Stack</u>	<u>Run</u>	<u>Ignoring Inlet Deposit</u>	<u>Including Inlet Deposit</u>	<u>Activity Percent Deposited in Inlet</u>
291-Z-1	1	6.8	15	46%
	2	8.6	26	59%
	3	3.5	6.2	23%
296-Z-3	1	3.1	3.5	7%
	2	2.8	3.6	14%
	3	3.0	3.7	16%
	4*	2.7	9.6	29%

*Low flow rate impactor

5.6 ANALYSES OF ARHCO SAMPLES

During the course of the background and cascade impactor sampling experiments several of ARHCO's routine air samples were analyzed by BNW's Technical Analysis Section for total plutonium content. The results of these analyses and the total curies of plutonium released during the sampling periods are recorded in Appendices H.1 and H.2 for the 291-Z-1 and 296-Z-3 stacks. For purposes of comparison the stack releases as measured by Battelle's samples and as reported in ARHCO's AIRSAM reports are also given in the same appendices. ARHCO's releases were reported in terms of gross alpha whereas our analyses were for plutonium only so the results are not exactly comparable. Furthermore, an impactor sample tends to underestimate the quantity of total particulates versus a single filter sample because of interstage losses unaccounted for as illustrated by Mishima's data. (20, p. 23)

6.0 DISCUSSION

In this section the relationship of the experiments to the subject of this study will be discussed. The particle sizing, concentration profile and comparison sampling experiments plus the analyses of some concurrent ARHCO samples contribute to evaluating the performance of the existing routine sampling systems at the two stacks.

The particle sizing experiments showed that the plutonium bearing aerosols have different activity mean aerodynamic diameters at the two stacks--3.5 to 8.6 μm at the 291-Z-1 stack versus 2.7 to 3.1 μm at the 296-Z-3 stack. The aerosol at the 291-Z-1 stack presents more of a sampling challenge than the aerosol at the 296-Z-3 stack, other things being equal.

The concentration profile experiments showed that along the stack diameters studied there was small stratification of the tracer aerosols except for the TiO_2 aerosol at the 291-Z-1 stack. The experiment also showed that the stratification was more pronounced for larger particles such as the ZnS than for smaller TiO_2 particles. In all cases the stratification was not severe. On the average the tracer concentration at any location did not vary more than 14% from the mean for that tracer and stack. The results of this experiment indicate that if the performance of the existing sampling systems is very poor, it would not be caused by stratification; however, only limited areas of the stacks were studied so there is the possibility that stratification may be more pronounced elsewhere on the stack cross sections.

The comparison sampling experiments showed that neither of the two sampling systems collected a sample of particles as large as 10 μm AED that was 90% of a representative sample as required in ARHCO's Sampling Criteria. Of the zinc sulfide tracer about 52% of it by weight was smaller than 10 μm AED. To satisfy the Interim Criteria then, ARHCO's samples should have collected 47% as much of the tracer as Battelle's samples normalized for flowrate. By the same reasoning, ARHCO's sample should have picked up 78% as much of the TiO_2 tracer as Battelle's when normalized for flowrate. At the 291-Z-1 stack, the existing routine sampler failed to meet these criteria for the zinc sulfide

tracer but met it for the titanium dioxide tracer. The routine sampler did not meet these criteria for either tracer at the 296-Z-3 stack.

We assumed in this discussion that Battelle's samples were representative; however, a look at the average recoveries listed in Table 6 raises some questions. Battelle's samples were clearly more representative than ARHCO's but a great deal of the injected tracer was unaccounted for--especially the zinc sulfide. It is possible that either the tracer deposited in the ventilation systems upstream of the samplers or there was severe stratification in the uninvestigated regions of the airflow and Battelle's sampler was not located to sample representatively. There may still be other feasible explanations; however, further investigation of the technique to explain this performance is indicated.

The preliminary report on the two existing sampling systems⁽¹⁾ estimated better routine sampler performance at the 296-Z-3 stack than the 291-Z-1 stack. The data from the comparison sampling experiment do not support this conclusion. Possibly the tracer size distributions were altered in one or both stacks by deposition of the tracer in the exhaust systems.

For the Battelle samples of the zinc sulfide tracer from the 296-Z-3 stack where the Wash Code = 3*, the amount found in the nozzles ranged from 4-21% of the total sample. The nozzle sample comprised less than 4% of the total sample in all the other comparison sampling experiments. This illustrates one case where it is difficult to collect on the filter 90% of a representative sample of large particles even with an in-stack sampler. This also illustrates the value of accounting for material deposited in the probes.

The two instances where Battelle's samples could be compared against the routine samples on the basis of plutonium emitted (see Appendix H.1) from the 291-Z-1 stack tend to support the conclusion that the routine sampler doesn't collect samples that satisfy the Interim Sampling Criteria for particles as large as 10 μm . This is as expected because the routine sampler performed

*Where nozzle washes were analyzed separately rather than combined.

poorly for the zinc sulfide tracer (see Appendix A) which had a size distribution similar to the plutonium aerosol.

The two cases where samples from the 296-Z-3 stack could be compared on the basis of plutonium emitted neither support nor refute the conclusion made concerning the routine sampler's compliance with the Interim Criteria (see Appendix H.2). The plutonium bearing aerosol in the exhaust did not present a severe challenge to the sampling system. It was surprising that the 296-Z-3 sampler performed better for the plutonium than for the TiO_2 tracer, which had a finer size distribution. Although a comparison of only two samples is a narrow base for judgement, this observation is consistent with the speculation that the tracer particle size distributions were altered in the exhaust system. It was also interesting that at the 296-Z-3 stack the reported total alpha curies emitted were more conservative than the amounts of plutonium emitted as derived from our analyses.

7.0 CONCLUSIONS

The following are the major conclusions reached as a result of this study.

- Neither of the existing routine sampling systems at the 296-Z-3 and 291-Z-1 stacks satisfy the Interim Criteria for collection of particulate samples as demonstrated with the tracer aerosols.
- The two routine sampling systems tested performed better for smaller particulates than for larger ones.
- The exploration of a limited area in each stack did not show any serious channeling of particulates.
- The plutonium bearing particulates have activity median aerodynamic diameters of 2.7 to 3.1 μm at the 296-Z-3 stack and 3.5 to 8.6 μm at the 291-Z-1 stack.
- The tracer aerosol sampler validation concept is a workable tool for testing existing particulate sampling systems.

Improvements in the way particulates are sampled in the two stacks appear necessary. As of this writing Rockwell has undertaken to design and install significantly improved particulate sampling systems following the recommendations of the preliminary report.⁽¹⁾ Those recommendations are consistent with the findings of this study and their continued implementation is adviseable. Continued development of the tracer aerosol sampler validation technique to the stage where it can be routinely used with confidence is recommended.

NOMENCLATURE

AED - aerodynamic equivalent diameter
atm - atmospheres pressure
c - counts
cfm - cubic feet per minute
Ci - curies
cm - centimeters
d - disintegrations
D - particle diameter
dpm - disintegrations per minute
ft - feet
g - grams
hr - hours
ID - inside diameter
in. - inches
in. Hg - inches of mercury, pressure
in. W.G. - inches of water, pressure
min - minutes
ml - milliliters
mm - millimeters
O.D. - outside diameter
scfh - cubic feet per hour at standard conditions, i.e., 1 atm
absolute, 70°F
scfm - cubic feet per minute at standard conditions
sp. gr. - particle specific gravity relative to water

std. ft³ - cubic feet at standard conditions

std. ml. - milliliter at standard conditions

S_x - standard deviation of samples

$S_{\bar{x}}$ - standard deviation of mean of samples

°F - degrees Fahrenheit

μCi - micro (10⁻⁶) curies

μg - micro (10⁻⁶) grams

μm - micro (10⁻⁶) meters

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2. Drawing H-2-16017, "234-5 and 291-Z Buildings, Breeching to Exhaust Fan Tunnel and Stack Foundation," Rockwell Hanford Operations Engineering Files.^(a)
3. Drawing H-2-16183, "234-5 Building Z, 252-Z-2, 291-Z Building, Roof Plan and Sections," Rockwell Hanford Operations Engineering Files.^(a)
4. Drawing H-2-16015, "291-Z and 252-Z-2 Buildings, Exhaust Fan Tunnels and Stack Foundation," Rockwell Hanford Operations Engineering Files.^(a)
5. Drawing H-2-16016, "291-Z and 252-Z-2 Buildings, Sections and Details for Exhaust Fan Tunnels," Rockwell Hanford Operations Engineering Files.^(a)
6. Drawing H-2-16412, "Exhaust Fans, Ventilation Piping and Drainage," Rockwell Hanford Operations Engineering Files.^(a)
7. Drawing H-2-16485, "Building 291-Z, Breathing Air System and Air Monitoring System in Building 291-Z," Rockwell Hanford Operations Engineering Files.^(a)
8. Drawing H-2-23114, "Exhaust Ventilation and Service Piping Arrangement, Services to 232-Z," Rockwell Hanford Operations Engineering Files.^(a)
9. Drawing H-2-29648, "Ventilation and Service Piping Modification, Plutonium Reclamation Facility," Rockwell Hanford Operations Engineering Files.^(a)
10. Certified Vendor Information File 1312, Drawing C-5796-2, "200 ft x 13 ft-6 in. ID Reinforced Concrete Chimney," Rockwell Hanford Operations Engineering Files.^(a)
11. Drawing H-2-24543, "In-Line Air Sampler Assembly and Details," Rockwell Hanford Operations Engineering Files.^(a)
12. Drawing H-2-26146, "Electrical, 241-Z Sump Continuous Stack Sampler," Rockwell Hanford Operations Engineering Files.^(a)

^(a) Copies can be obtained through Pacific Northwest Laboratory Engineering Files (509) 376-0067.

13. Drawing H-2-26135, "241-Z Sump Continuous Stack Sampler," Rockwell Hanford Operations Engineering Files.^(a)
14. Drawing H-2-43549, "Waste Sumps Facility Equipment and Piping Arrangement Plans," Rockwell Hanford Operations Engineering Files.^(a)
15. Drawing H-2-43552, "Exhaust System 421-Z Sump Arrangement and Sections," Rockwell Hanford Operations Engineering Files.^(a)
16. Counting Air Samples at 234-5 Process Control Laboratory, AIS-0IA, ARHCO.^(b)
17. An Outline of Environmental Air Sample Analysis, ARH-869, ARHCO, 1968.^(b)
18. Emergency and Holiday Coverage of 222-S Counting Room, Technical Services Section, Laboratory Operations Procedure No. ECOVCR-0IA, ARHCO, 1973.^(b)
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21. Mishima, J., and L. C. Schwendiman. Characterization of Radioactive particles in the 234-5Z Building Gaseous Effluent. December 1973. BNWL-B-309, Pacific Northwest Laboratory, Richland, Washington.

(a) Copies can be obtained through Pacific Northwest Laboratory Engineering Files (509) 376-0067.

(b) ARHCO internal procedure in place at time of work (1977).

APPENDIX A

TRACER AEROSOLS

APPENDIX A

TRACER AEROSOLS

Because the tracer aerosols were first developed and tested as part of this study, a description of their composition and generation is warranted.

The TiO_2 tracer aerosol was a blend of 87.7% titanium dioxide and 12.3% zinc stearate by weight. Titanium dioxides furnished by both Baker^{*} and MCB^{**} were used and both yielded blends of similar size characteristics. The zinc stearate was a product of Synpro^{***} and was used to improve the packing and surface characteristics of the tracer cake in the Wright Dust Feeder.[†]

The TiO_2 was photographed with a Scanning Electron Microscope and the micrographs were analyzed using a Quantimet Image Analyzer which produced a size distribution. From these data a mass mean diameter of 1.5 μm and a mass mean aerodynamic diameter (MMAD) of 3.1 μm were estimated. The size distribution of the blended tracer as generated by the Wright Dust Feeder was determined by a high volume cascade impactor^{††}. Runs were made using both the Baker and MCB titanium dioxides. The data are shown in Figure A.1. The mass median aerodynamic diameter of the titanium dioxide tracer as produced by the Wright Dust Feeder was 1.3 μm .

A second tracer aerosol with a larger size distribution was also desired. Several compounds were screened by SEM microphotography and Quantimet Image Analysis for particle size distribution. These were zinc sulfide, zinc oxide, chromium oxide, and zinc dust. The zinc sulfide was selected for further use,

*J. T. Baker Chemical Co., Phillipsburg, NJ.

**MCB Manufacturing Chemists, Norwood, OH.

***Synthetic Products Co., Cleveland, OH.

†Manufactured by L. Adams, Ltd., London, England, and distributed by BGI Inc., Waltham, MA.

††Model 65-800, Anderson 2000 Inc., Atlanta, GA.

FIGURE A.1. Size Distribution Data for Titanium Dioxide-Zinc Stearate Tracer Generated with Wright Dust Feeder

but some of the other compounds may also have been suitable with further development. The zinc sulfide was a commercial fluorescent pigment^{*} but was coarser than desired. Ball milling to reduce the size of the zinc sulfide proved cumbersome and slow so a quantity was sent to Majac^{**} to have a suitable size fraction separated from the bulk material by a type of air classification. The product zinc sulfide was then blended with the same zinc stearate as mentioned before to improve the handling characteristics for the Wright Dust Feeder. The blend proportions were 87.7% zinc sulfide and 12.3% zinc stearate by weight. The size distribution of the zinc sulfide blended tracer as generated by the Wright Dust Feeder was determined with the high volume cascade impactor. The size distribution data are plotted in Figure A.2. Ignoring impactor inlet losses, the MMAD was 8.5 μm . Accounting for the inlet losses by adding them to the first impactor stage yielded a MMAD of 10.5 μm .

During the experimental program the tracer aerosols were generated by the Wright Dust Feeder. Bottled breathing air was fed to the Wright Dust Feeder and the outlet from the feeder was situated inside the opening to the ventilation duct (see Figures 9 and 11). The tracer aerosol feed rate was regulated by the gear train on the Dust Feeder. Because the dilution of the tracer in the 291-Z-1 stack was many times greater than in the 296-Z-3 stack, a much higher feed rate was required at the former in order to complete a run in a reasonable length of time. A feed rate of 0.33 g tracer/min was achieved for the 291-Z-1 stack which meant operating the Dust Feeder near its maximum rate. A feed rate of only about 0.08 g tracer/min was required at the 296-Z-3 stack.

^{*}Fluorescent Pigment 2210, United States Radium Co., Morristown, NJ.

^{**}Majac Division, Donaldson Company, Inc., Tulsa, OK.

FIGURE A.2. Size Distribution Data for Zinc Sulfide Tracer
Generated with Wright Dust Feeder

APPENDIX B

X-RAY FLUORESCENCE AND PLUTONIUM ANALYSIS PROCEDURES

APPENDIX B

X-RAY FLUORESCENCE AND PLUTONIUM ANALYSIS PROCEDURES

The following analytical procedures were employed by BNW's Technical Analysis and Chemical Kinetics and Methods sections. The invaluable assistance of Kirk Nielson, Jim Langford, L. J. Kirby and their colleagues is acknowledged. The description of the X-ray fluorescence analysis is quoted from materials supplied by Langford. The description of the plutonium analysis is paraphrased from information supplied by Kirby.

X-Ray Fluorescence

X-ray fluorescence was employed in the analysis of background and tracer aerosol samples. Filter samples were simply mounted and analyzed directly in triplicate (as were all other samples). Nozzle washes were all 6 N H_2SO_4 solutions and were analyzed as described below by Langford. The QA procedures are also as described by Langford.

Procedure for Determining Zinc

All samples being analyzed were in 6 N H_2SO_4 and contained only trace contaminants. Analysis was by precipitation of the desired constituent, then measurement on the X-ray fluorescence apparatus.

1. To each sample wash in its original vial, add 1 drop of 0.1% Phenol Red.
2. Neutralize the sample with NH_4OH and swirl to mix well.
3. Add about 1 ml of Thioacetamide powder to each vial and mix well. Cap the vial loosely to allow pressure release and let set 10 minutes. The solution will remain hot from the heat of neutralization.
4. Filter the solution through a 0.45 μm Millipore, then mount the filter onto a 35 mm slide holder and count on the XRF.
5. Count the sample for 15 minutes (900 seconds), Regulator at 500, Zr secondary, Zr filter, in channels 333 to 355.

6. Include the $131 \mu\text{g Zn/cm}^2$ standard for energy calibration and for calculation standard.
7. Standard Zinc solution run with each set.

Procedure for Determining Titanium

The samples being analyzed were all in 6 N H_2SO_4 and contained trace contaminants. Analysis was by precipitation of the desired constituent, then measured on the X-ray fluorescence apparatus.

1. Combine the sample washes for one run into a clean bottle.
2. Add 1 drop of 0.1% Phenol Red Indicator plus 0.5 mg of Al^{+++} .
3. Neutralize with NH_4OH and let set 10 minutes.
4. Filter the solution through a $0.45 \mu\text{m}$ Millipore filter.
5. Mount the filter in a 35 mm slide holder and analyze the sample for Ti by X-ray fluorescence.
6. Count the sample by XRF for 900 seconds, Regulator at 500, Cu secondary, no filter, in channels 166 to 188.
7. Include the $81 \mu\text{g Ti/cm}^2$ standard for calibration of energy and for calculation.
8. Standard Ti solution run with each set.

Quality Assurance Procedures for X-R-F Analysis of TiO_2 And ZnS Loaded Millipore Filters

Since the loading of TiO_2 and ZnS on the millipore filters was consistently below 100 mg/cm^2 , samples could be regarded as thin films and required no absorption corrections. Quantities of Ti and Zn were thus directly proportional to the respective K_α X-ray peak areas. A thin film standard* of either Ti or Zn was analyzed with each set of 16 samples, and all resulting peak areas were compared directly to the standard for quantitative analysis. Net peak areas were determined by the Covell method** using standard software

*Micromatter Corp., Seattle, WA.

**D. F. Covell, Anal. Chem. 31, 1785 (1959).

in the multichannel analyzer system (Model 4410, Nuclear Data, Palatine, IL, N.D. Programs 41-1060 and 41-1104). Accuracy of the thin film standards was verified by use of a multi-element calibration curve and analyses of NBS and USGS standard reference materials*. Appropriate blanks were analyzed with each filter set and three replicate analyses of each sample were done. Interferences by other elements in the samples were investigated and found to be insignificant. Analysis was accomplished using a Model 810 Kevex vacuum X-ray spectrometer and excitation system.

Plutonium

The background experiment and the particulate sizing experiment samples were analyzed for total plutonium. Glass fiber filters, membrane filters and HNO_3 solutions were the sample matrices analyzed. The glass fiber filters were heated overnight at 500°C in a muffle furnace to remove the organic binder. The siliceous material was destroyed and the residues solubilized with treatments of HF and HNO_3 . The membrane filters were dissolved in $\text{HNO}_3\text{-H}_2\text{O}_2$ and then treated with HF to dissolve the siliceous material. The liquid samples were also treated with HF and HNO_3 to destroy any siliceous materials.

All of the resulting solutions were evaporated to dryness and the residues were dissolved in 7.7 N HNO_3 . The plutonium was then separated out from the samples by standard ion exchange procedures. The Pu fractions were plated on dishes and counted with zinc sulfide scintillation detectors. Spiked filters and blank filters were analyzed simultaneously for quality control. Standard documented quality control procedures were followed to insure counter and background stability.

*K. K. Nielson, BNWL-SA-5785, Anal. Chem. 49 (1977) (in Press).

APPENDIX C

VELOCITY TRAVERSE DATA

AVERAGE VELOCITY = 2358 ft/min

STACK DIAMETER = 14 in.

EXHAUST FLOWRATE = 2521 ft³/min = 1983 SCFM

PRESSURE = 0.15 IN W.G.

TEMPERATURE = 150°F

APPENDIX C.1

296-Z-3 Stack Velocity Traverse

STACK INSIDE DIAMETER 188-1/2in.

AVERAGE VELOCITY = 1560ft/min

AVERAGE FLOWRATE = 302,000CFM ~ 300,000SCFM

TEMPERATURE ~ 74°F

PRESSURE = 0.20 in W. G.

APPENDIX C.2

Initial 291-Z-1 Stack Velocity Traverse
with Flange Unplugged

APPENDIX C.3

Additional 291-Z-1 Stack Velocity Data

The following data were taken at six locations spaced equally across the stack.

<u>Location</u>	<u>Distance From Flange Outer Edge, in.</u>	<u>Velocities ft/min</u>		
		<u>March 1976</u>	<u>October 25, 1976</u> <u>Hi - Lo - Estimate</u>	
1	179	1300	850 - 1600	1200
2	152	1350		
3	125	1500		
4	98	1550	1334 - 1658	1520
5	71	1725		
6	44	1650	1200 - 1700	1510

APPENDIX C.4

Final Velocity Traverse of 291-Z-1 Stack

■ Refers to points located by the formula:

$$R_n = \sqrt{\frac{(n - 1/2) a}{\pi}}$$

where R_n is the radius from the stack center to the point, a is 1/6th of the stack area and n is the number identifying each of the six equal areas. The average velocity of these points is 1436 std ft/min. This is the common way of laying out the velocity traverse.

◆ Refers to six points spaced equally across the stack (see Appendix C.3) plus the centerpoint.

APPENDIX D

DATA FROM BACKGROUND SAMPLING EXPERIMENT

APPENDIX D.1

Background Sampling Experiment Analytical Data

		ELEMENTAL X-RAY FLUORESCENCE ANALYSES - $\mu\text{g}/\text{cm}^2$ OF FILTER							RADIOCHEMICAL
SAMPLE	FILTER	Ti	Mn	Fe	Ni	Cu	Zn	Sn	Pu d/min
296-Z-3 STACK SAMPLE FLOWRATE = 1.5 SCFM									
1	MEMBRANE	0.165	<0.0059	0.378	0.0054	0.072	0.092		9.28
$\Delta t = 4257 \text{ min}^*$	BACK-UP	76.2	1.21	3.66	0.030	≤ 0.031	187		1.68
2	MEMBRANE	0.158	0.018	0.931	0.0088	0.079	0.135		16.9
$\Delta t = 10,055 \text{ min}$	BACK-UP	73.7	1.19	3.62	0.035	≤ 0.029	189		3.21
3	MEMBRANE	0.136	<0.0068	0.971	0.011	0.074	0.095	0.84	21.4
$\Delta t = 11,550 \text{ min}$	BACK-UP	75.5	1.17	3.63	0.033	≤ 0.030	178	0.99	1.52
4	MEMBRANE	0.105	0.011	0.661	0.013	0.077	0.224		5.33
$\Delta t = 4290 \text{ min}$	BACK-UP	74.2	1.20	3.79	0.042	≤ 0.030	180		2.93
291-Z-1 STACK SAMPLE FLOWRATE = 1.2 SCFM									
1	MEMBRANE	DAMAGED NOT ANALYZED							8.7
$\Delta t = 4240 \text{ min}$	BACK-UP	73.0	1.22	6.45	0.097	0.138	174		2.23
2	MEMBRANE	0.061	0.013	2.87	≤ 0.0036	0.083	0.243		1800
$\Delta t = 10,050 \text{ min}$	BACK-UP	73.7	1.19	10.1	0.028	≤ 0.031	182		66.1
3	MEMBRANE	<0.017	0.017	2.93	≤ 0.0041	0.084	0.216	0.41	133
$\Delta t = 11,530 \text{ min}$	BACKUP	78.3	1.22	4.81	0.034	≤ 0.028	183	0.41	294
4	MEMBRANE	0.032	<0.0059	1.15	≤ 0.0031	0.090	0.075		14.8
$\Delta t = 4265 \text{ min}$	BACK-UP	82.7	1.35	6.03	0.029	≤ 0.029	193		56.0
BLANK FILTERS									
IPC NO. 1478 TREATED		<0.019	0.014	0.42	<0.0035	0.048	0.30	≤ 0.29	
GELMAN TYPE A GLASS FIBER		72.8	1.33	3.24	0.045	<0.020	176	≤ 0.26	
GELMAN TYPE A LOT 8064 LIGHT MAT		0.55- 0.83							
GELMAN TYPE A LOT 8064 HEAVY MAT		17.07- 18.95							
GELMAN ACROPOR AN3000		10.4	<0.04	<0.03	<0.016	0.033	0.045	≤ 0.24	
HV-70		12.0	0.94	216	<0.028	0.068	33	≤ 0.30	
MF - MILLIPORE SMWP		<0.18	<0.041	<0.035	<0.018	0.073	0.021	≤ 0.25	
GELMAN TYPE E GLASS FIBER		37					173		
GELMAN METRICEL VM-1		0.17- 0.97		12					
MILLIPORE MITEX		0.014- 0.042					0.003- 0.017		
MILLIPORE FLUOPORE		0.026					0.003		
NUCLEPORE		<0.008					0.001- 0.005		

*ELAPSED TIME

APPENDIX D.2
Effluent Background Concentrations

	Run	mo-day-time		Measureable Elemental Concentrations μg/std. ft ³ *				Pu μCi/std. ml**
		On	Off	Fe	Cu	Zn	Sn	
296-Z-3 Stack	1	2-9-1030	2-12-0927	1.2E-3		1.1E-4		2.7E-14
	2	2-12-0935	2-19-0910	8.6E-4		8.0E-5		2.1E-14
	3	2-19-0920	2-27-0950	7.8E-4		4.4E-5	7.8E-4	2.1E-14
	4	2-27-0950	3-1-0920	1.9E-3		3.3E-4		2.0E-14
291-Z-1 Stack	1	2-9-1100	2-12-0940					3.4E-14
	2	2-12-0950	2-19-0920	8.2E-3	9.1E-6	1.9E-4		2.5E-12
	3	2-19-0930	2-27-0940	3.3E-3	8.7E-6	1.4E-4	2.2E-4	4.9E-13
	4	2-27-0945	3-1-0850	7.6E-3	3.5E-5	1.1E-4		2.2E-13

There were no measureable concentrations of Ti, Mn

$$* \text{Concentration } \mu\text{g}/\text{std. ft}^3 = \frac{\mu\text{g}}{\text{cm}^2} \times 10.2 \text{ cm}^2 \times \frac{\text{min.}}{\text{S.F. std. ft}^3} \times \frac{1}{\Delta t \text{ min.}} \quad (\text{D-1})$$

where the filter deposit area was 10.2 cm² and the sample flowrate, S.F., was 1.5 SCFM and 1.2 SCFM at the 296-Z-3 and 291-Z-1 stacks respectively. The μg/cm² was the total of the membrane and glass fiber filter analyses.

$$** \text{Pu concentration } \mu\text{Ci}/\text{std. ml} = \frac{d}{\text{min.}} \times \frac{\mu\text{Ci} - \text{min.}}{2.22\text{E-}6d} \times \frac{\text{min.}}{\text{S.F. std. ft}^3} \times \frac{1}{\Delta t \text{ min.}} \times \frac{\text{ft}^3}{28,320 \text{ ml}} \quad (\text{D-2})$$

where S.F. is the sample flowrate and d/min. is the sum of activity on both the membrane and glass fiber filters.

NOTE: To save space computer format used for scientific notation, thus E-6 equals 10⁻⁶.

APPENDIX E

CONCENTRATION PROFILE EXPERIMENT DATA

APPENDIX E.1a

Concentration Profile, Titanium Analyses, 291-Z-1 Stack

Run and Elapsed Time, min.	Location	Membrane Filter Analyses μg/cm ²			Back-up Glass Fiber Filter Analysis μg	Nozzle Wash Analysis μg	Total Ti on Sample μg	Ti Conc μg/std. ft ³ x10 ²
		1	2	3				
Run 7* 1450 min.	1	4.62	4.62	4.57	19.18	5.54	51.9	3.47
	4	6.22	6.31	6.28	20.03	3.96	65.4	4.22
	6	6.03	5.96	5.89	18.69	5.05	66.1	4.14
	Blank Filters	0.005			18.95			
Run 8 4320 min.	1	4.52	4.60	4.48	18.57	4.26	49.8	1.12
	4	6.79	6.87	6.81	19.02	4.95	71.7	1.55
	6	6.48	6.58	6.59	19.05	7.72	74.7	1.57
	Blank Filters	0.01			0.83			
Run 9 1425 min.	1	4.35	4.26	4.22	0.81	5.74	47.8	3.26
	4	6.11	5.90	6.00	0.70	7.33	65.2	4.27
	6	4.80	4.77	4.68	18.05	4.06	51.7	3.30
	Blank Filters	0.10			17.62			
Run 10 1425 min.	1	0.15 [†]	3.68	3.79	18.29	3.56	41.1	2.80
	4	6.41	6.08	6.00	17.64	4.06	64.4	4.22
	6	6.02	5.95	5.79	17.19	5.45	66.0	4.21
	Blank Filters	0.01			17.07			
Run 11 1400 min.	1	3.91	4.04	3.98	0.56	0.556	40.5	2.81
	4	5.81	5.98	5.93	0.66	0.426	58.3	3.89
	6	6.31	6.34	6.38	0.70	0.706	65.5	4.25
	Blank Filters	0.01			0.59			

* Runs 1-6 were discarded.

† Considered as an outlier and not included in calculations.

APPENDIX E.1b

Concentration Profile Titanium Analyses, 296-Z-3 Stack

Run and Elapsed Time, min.	Location	Membrane Filter Analyses μg/cm ²			Back-up Glass Fiber Filter Analysis μg	Nozzle Wash Analysis μg	Total Ti on Sample μg	Ti Conc μg/std. ft ³
		1	2	3				
Run 3* 95 min.	1	85.63	83.14	82.98	0.55	13.5	836	6.33
	2	85.33	81.51	84.04	0.64	15.5	882	6.83
	3	69.08	69.84	70.01	0.59	12.2	725	5.30
	Blank Filters	0.01			0.52			
Run 4 88 min.	1	79.40	78.01	77.21	0.58	14.6	781	6.38
	2	76.18	75.04	76.15	0.54	13.9	799	6.68
	3	68.01	68.50	68.08	0.68	13.6	712	5.62
	Blank Filters	0.01			0.63			
Run 5 83 min.	1	94.08	93.00	93.85	0.57	7.75	925	8.02
	2	90.12	93.22	89.35	0.61	10.19	952	8.43
	3	82.24	83.03	81.93	0.57	12.39	856	7.16
	Blank Filters	0.01			0.60			
Run 6 97 min.	1	86.00	88.66	87.67	0.63	6.47	863	6.40
	2	86.38	87.49	84.69	0.59	5.65	898	6.81
	3	79.59	80.11	79.32	0.63	7.45	823	5.89
	Blank Filters	0.01			0.57			
Run 7 74 min.	1	87.35	91.11	90.05	0.66	4.84	882	8.57
	2	84.56	88.59	86.83	0.60	5.23	903	8.97
	3	77.36	81.19	80.86	0.56	4.72	822	7.71
	Blank Filters	0.002			0.57			
Run 8 77 min.	1	85.46	86.63	88.33	0.62	4.87	856	7.99
	2	80.92	85.40	79.77	0.64	6.22	856	8.17
	3	80.77	83.69	80.21	0.64	6.24	841	7.59
	Blank Filters	0.004			0.60			

*Runs 1-2 were discarded.

APPENDIX E.1c

Concentration Profile Experiment Calculations

The triplicate analyses of the membrane filters were averaged and then the titanium background in the blank filter was subtracted. The total titanium per location sample was then calculated as:

$$\begin{aligned} \text{Total Ti, } \mu\text{g} &= (\text{Average filter analysis } \frac{\mu\text{g}}{\text{cm}^2} \times \text{Deposit Area, cm}^2) \\ &+ \text{Nozzle } \mu\text{g} \end{aligned} \quad (\text{E-1})$$

The titanium concentration was then calculated as follows:

$$\text{Ti Cone } \mu\text{g/std. ft}^3 = \frac{\text{Total Ti } \mu\text{g}}{\text{Sample Flowrate scfm} \times \text{Elapsed Time min.}} \quad (\text{E-2})$$

The analyses of the back-up glass fiber filters did not enter into the calculations. The titanium tracer content on the back-up filters were taken to be zero. Although all the glass fiber filters were from the same lot there were two visually distinguishable types with different amounts of titanium as shown in Runs 8 and 9, 291-Z-1 stack.

APPENDIX E.2a

Additional Concentration Profile Data from Comparison Sampling Experiment -- TiO_2 Tracer

	RUN	LOCATION	TOTAL TITANIUM IN SAMPLE μg	TITANIUM CONCENTRATION $\mu\text{g}/\text{std ft}^3$
<u>291-Z-1 STACK</u>	R3	1	51.3	0.372
		4	62.8	0.402
		6	59.3	0.373
	R4	1	35.2	0.258
		4	60.0	0.390
		6	60.1	0.383
	R5	1	48.2	0.354
		4	61.0	0.397
		6	59.1	0.377
	R6	1	50.4	0.386
		4	62.3	0.422
		6	62.3	0.414
	R7	1	49.1	0.349
		4	45.7	0.287
		6	60.1	0.370
<u>296-Z-3 STACK</u>	R1	1	1088	7.04
		2	1090	7.26
		3	1061	6.72
	R3	1	1120	9.02
		2	1031	8.55
		3	1041	8.20
	R4	1	1135	10.5
		2	994	9.51
		3	1082	9.84
	R5	1	1207	11.4
		2	1165	11.3
		3	1188	10.9
	R6	1	1067	9.32
		2	1055	9.49
		3	1054	9.00
	R7	1	674	6.78
		2	599	6.21
		3	618	6.09
	R8	1	1271	12.3
		2	1167	11.6
		3	1172	11.1
	R9	1	952	8.74
		2	944	8.92
		3	925	8.30
	R10	1	982	8.78
		2	907	8.36
		3	989	8.66

APPENDIX E.2b

Additional Concentration Profile Data from Comparison
Sampling Experiment -- ZnS Tracer

	<u>RUN</u>	<u>LOCATION</u>	<u>TOTAL ZINC IN SAMPLE μg</u>	<u>ZINC CONCENTRATION $\mu\text{g}/\text{std ft}^3$</u>
<u>291-Z-1 STACK</u>	R7	1	15.4	0.166
		4	20.1	0.191
		6	18.9	0.177
	R10	1	15.7	0.163
		4	18.9	0.173
		6	19.2	0.173
	R11	1	13.8	0.135
		4	19.0	0.164
		6	16.8	0.143
	R12	1	15.3	0.164
		4	22.1	0.211
		6	19.4	0.181
<u>296-Z-3 STACK</u>	R1	1	696	8.13
		2	755	9.09
		3	642	7.34
	R2	1	536	7.93
		2	632	9.62
		3	547	7.91
	R3	1	480	6.22
		2	595	7.93
		3	521	6.60
	R4	1	472	6.58
		2	607	8.71
		3	465	6.34
	R6	1	540	7.82
		2	628	9.37
		3	542	7.69
	R7	1	541	8.52
		2	574	9.31
		3	476	7.35

APPENDIX E.2c

Calculations for Additional Concentration Profile Data

The values tabulated in Appendices E.2a and E.2b were calculated essentially the same as the comparison sampling experiment calculations detailed in Appendix F using a slightly different version of the computer program (COMEXP2). Only those runs for which the nozzle washes were analyzed separately were used for concentration profile data. The total amount of tracer collected at each sample location was calculated from the background corrected data for membrane filter and nozzle wash analyses. The tracer concentration at each location was then calculated as follows from the total tracer collected and flowrate at each of the control samplers.

$$\text{Tracer Conc } \mu\text{g/std ft}^3 = \frac{\text{total } \mu\text{g at location}}{\text{sampler flowrate, scfm} \times \text{sampling time min.}} \quad (\text{E-3})$$

APPENDIX F

DATA FROM COMPARISON SAMPLING EXPERIMENTS

APPENDIX F.1

Comparison Sampling, 291-Z-1 Stack, TiO₂ Tracer Data

RUN	SAMPLER LOCATION	FILTER ANALYSES			WASH ANALYSES		
		TITANIUM, $\mu\text{g}/\text{cm}^2$			TITANIUM, μg		
		1	2	3	1	2	3
R3	SAMPLING TIME 150 MIN. -- GRAMS INJECTED 32.4				WASH CODE 3		
	B1	5.12	5.16	5.08	0.72	0.7	0.69
	B4	5.05	5.06	5.05	1.07	1.11	1.23
	B6	5.75	5.48	5.3	0.6	0.55	0.56
	ARHCO	6.69	6.63	6.69			
	BACKGROUND	0.011	0.0092	0.013			
R4	SAMPLING TIME 149 MIN. -- GRAMS INJECTED 31.4				WASH CODE 3		
	B1	3.54	3.54	3.47	0.33	0.43	0.48
	B4	5.33	5.27	5.79	0.65	0.63	0.63
	B6	5.93	5.91	5.86	0.51	0.54	0.39
	ARHCO	6.03	6.15	6.18			
	BACKGROUND	0.0045	0.0026	0.013			
R5	SAMPLING TIME 145 MIN. -- GRAMS INJECTED 31.4				WASH CODE 3		
	B1	4.33	4.23	4.31	0.39	0.57	0.46
	B4	5.94	5.92	5.93	0.62	0.7	0.65
	B6	5.93	5.4	5.77	0.56	0.57	0.4
	ARHCO	7.32	7.68	7.59			
	BACKGROUND	0.016	0.011	0.0096			
R6	SAMPLING TIME 142 MIN. -- GRAMS INJECTED 31.5				WASH CODE 3		
	B1	5.09	5.06	4.97	0.3	0.25	0.42
	B4	6.11	6.07	6.01	0.41	0.42	0.47
	B6	6.17	6.14	6.04	0.51	0.45	0.44
	ARHCO	7.37	7.31	7.77			
	BACKGROUND	0.011	0.00044	0.017			
R7	SAMPLING TIME 153 MIN. -- GRAMS INJECTED 31.4				WASH CODE 3		
	B1	4.32	4.37	4.32	1.25	1.26	1.29
	B4	4.47	4.44	4.44	0.46	0.54	0.47
	B6	5.93	5.9	5.86	0.4	0.54	0.51
	ARHCO	5.26	5.23	5.32			
	BACKGROUND	0.0038	0.0066	0.01			
R8	SAMPLING TIME 181 MIN. -- GRAMS INJECTED 22.1				WASH CODE 1		
	B1	2.96	2.34	2.34	0.97	1	1.03
	B4	3.51	3.52	3.53			
	B6	3.65	3.69	3.68			
	ARHCO	3.13	3.35	3.27			
	BACKGROUND	0.0079	0.0091	0.0028			
R9	SAMPLING TIME 146 MIN. -- GRAMS INJECTED 29				WASH CODE 1		
	B1	3.31	3.33	3.34	0.91	0.93	1.05
	B4	4.25	4.23	4.23			
	B6	4.23	4.26	4.29			
	ARHCO	4.9	4.37	4.33			
	BACKGROUND	0.0027	0.0019	0.0099			
R11	SAMPLING TIME 152 MIN. -- GRAMS INJECTED 33.1				WASH CODE 1		
	B1	3.82	3.34	3.86	1.12	1.12	1.32
	B4	4.68	4.7	4.69			
	B6	4.64	4.63	4.65			
	ARHCO	5.55	5.65	5.59			
	BACKGROUND	0.013	0.0051	0.011			
R12	SAMPLING TIME 155 MIN. -- GRAMS INJECTED 32.2				WASH CODE 1		
	B1	3.73	3.71	3.71	1.07	0.96	1.12
	B4	4.49	4.54	4.48			
	B6	4.42	4.45	4.43			
	ARHCO	4.79	5.05	5.02			
	BACKGROUND	0.0033	0.0054	0.015			

NUMBER OF RUNS - 9
 STACK FLOWRATE - 300,000 SCFM
 BACKGROUND FILTER DEPOSIT AREA - 17.35 cm²
 AVERAGE WASH BACKGROUND - 0.54 μg

SAMPLER	FLOW SCFM	DEPOSIT cm ²
B1	0.92	10.02
B4	1.04	10.29
B6	1.06	10.2
ARHCO	1.3	14.37

APPENDIX F.2

Comparison Sampling, 296-Z-3 Stack, TiO₂ Tracer Data

RUN	SAMPLER LOCATION	FILTER ANALYSES			WASH ANALYSES		
		TITANIUM, $\mu\text{g}/\text{cm}^2$			TITANIUM, μg		
		1	2	3	1	2	3
R1	SAMPLING TIME - 112 MIN. -- GRAMS INJECTED - 3.5				WASH CODE - 3		
	B1	111	113	108	3.61	3.79	3.71
	B2	106	107	102	2.42	2.45	2.39
	B3	105	104	101	2.81	2.76	2.99
	ARMCO	3.63	3.62	3.57			
	BACKGROUND	0.009	0.017	0.021			
R2	SAMPLING TIME - 85 MIN. -- GRAMS INJECTED - 3.6				WASH CODE - 2		
	B1	116	116	116	0	0	0
	B2	105	104	103	1.75	1.73	1.73
	B3	102	100	104	2.9	2.91	3.02
	ARMCO	1.39	1.4	1.43			
	BACKGROUND	0.013	0.008	0.009			
R3	SAMPLING TIME - 90 MIN. -- GRAMS INJECTED - 3.5				WASH CODE - 3		
	B1	114	112	116	3.2	3.36	3.27
	B2	100	99	99	2.77	2.8	2.6
	B3	103	102	99	2.69	2.62	2.63
	ARMCO	3.85	3.8	3.91			
	BACKGROUND	0.016	0.013	0.005			
R4	SAMPLING TIME - 78 MIN. -- GRAMS INJECTED - 3.5				WASH CODE - 3		
	B1	118	119	111	2.44	2.31	2.28
	B2	95	94	98	3.79	3.74	3.79
	B3	106	106	104	3.04	3.01	3
	ARMCO	0.11	0.11	0.096			
	BACKGROUND	0.005	0.009	0.01			
R5	SAMPLING TIME - 77 MIN. -- GRAMS INJECTED - 3.6				WASH CODE - 3		
	B1	121	121	127	2.26	2.13	2.16
	B2	112	112	113	2.4	2.48	2.25
	B3	116	115	116	2.73	2.3	2.93
	ARMCO	4.19	4.24	4.26			
	BACKGROUND	0.015	0.013	0.006			
R6	SAMPLING TIME - 83 MIN. -- GRAMS INJECTED - 3.5				WASH CODE - 3		
	B1	109	108	109	3.01	2.98	2.96
	B2	101	101	103	2.59	2.56	2.62
	B3	103	103	102	1.91	1.92	1.89
	ARMCO	3.55	3.59	3.61			
	BACKGROUND	0.006	0.008	0.01			
R7	SAMPLING TIME - 72 MIN. -- GRAMS INJECTED - 2.5				WASH CODE - 3		
	B1	68.2	68.2	69.1	2.71	3.09	2.93
	B2	57.3	57.3	58	3.24	3.15	3.18
	B3	59.9	59.8	60.5	3.38	3.3	3.23
	ARMCO	3.33	3.39	3.37			
	BACKGROUND	0.0013	0.0045	0.00055			
R8	SAMPLING TIME - 75 MIN. -- GRAMS INJECTED - 4.1				WASH CODE - 3		
	B1	129	129	130	4.2	4.35	4.25
	B2	113	112	112	4.4	4.33	4.28
	B3	114	114	114	3.91	4.05	4.01
	ARMCO	2.93	2.97	2.96			
	BACKGROUND	0.0013	0.0087	0.018			
R9	SAMPLING TIME - 79 MIN. -- GRAMS INJECTED 3.5				WASH CODE - 3		
	B1	96.9	96.4	96.8	8.55	8.78	8.63
	B2	99.8	90.1	91	4.92	4.95	4.95
	B3	88.7	89.2	90.4	8.49	8.59	8.56
	ARMCO	5.75	5.72	5.76			
	BACKGROUND	0.012	0.013	0.013			
R10	SAMPLING TIME - 81 MIN. -- GRAMS INJECTED - 3.9				WASH CODE - 3		
	B1	98.3	99.4	101	6.45	6.35	6.43
	B2	86	87	88.1	6.33	6.23	6.34
	B3	94.5	95.7	96.4	10.1	10.1	10.3
	ARMCO	6.38	6.52	6.51			
	BACKGROUND	0.019	0.001	0.017			
R11	SAMPLING TIME - 77 MIN. -- GRAMS INJECTED - 3.6				WASH CODE - 2		
	B1	94.2	93.5	94.1	7.5	7.55	7.71
	B2	85.9	85.8	87	0	0	0
	B3	87.3	87.2	86.2	7.53	7.55	7.63
	ARMCO	5.33	5.36	5.39			
	BACKGROUND	0.0071	0.012	0.01			
NUMBER OF RUNS - 11		SAMPLER			FLOW SCFM		
STACK FLOWRATE - 1983 SCFM		B1			1.38		
BACKGROUND FILTER DEPOSIT AREA - 17.35 cm ²		B2			1.34		
AVERAGE WASH BACKGROUND - 0.54 μg		B3			1.41		
		ARMCO			1.53		
					DEPOSIT cm ²		
					9.8		
					10.36		
					10.25		
					57.17		

APPENDIX F.3

Comparison Sampling, 291-Z-1 Stack, ZnS Tracer Data

RUN	SAMPLER LOCATION	FILTER ANALYSES			WASH ANALYSES		
		ZINC, $\mu\text{g}/\text{cm}^2$			ZINC, μg		
		1	2	3	1	2	3
R1	SAMPLING TIME - 102 MIN. -- GRAMS INJECTED - 29				WASH CODE - 1		
	B1	1.56	1.6	1.56	5.54	4.94	5.49
	B4	1.92	1.97	1.96			
	B6	1.67	1.71	1.69			
	ARHCO	0.29	0.33	0.28			
	BACKGROUND	0.015	0.0043	0.0027			
R2	SAMPLING TIME - 150 MIN. -- GRAMS INJECTED - 31.1				WASH CODE - 1		
	B1	1.67	1.7	1.6	9.53	9.53	9.53
	B4	2.15	2.17	2.15			
	B6	1.99	2.09	2.03			
	ARHCO	0.15	0.19	0.2			
	BACKGROUND	0.016	0.016	0.0095			
R3	SAMPLING TIME - 106 MIN. -- GRAMS INJECTED - 30.2				WASH CODE - 1		
	B1	1.47	1.49	1.55	7.31	7.31	7.31
	B4	2.02	2.04	2.01			
	B6	1.84	1.9	1.99			
	ARHCO	0.19	0.21	0.22			
	BACKGROUND	0.0089	0.0058	0.0018			
R4	SAMPLING TIME - 113 MIN. -- GRAMS INJECTED - 32.3				WASH CODE - 1		
	B1	1.79	1.95	1.3	3.68	3.68	3.68
	B4	2.17	2.24	2.2			
	B6	2	2.16	2.05			
	ARHCO	0.21	0.23	0.25			
	BACKGROUND	0.023	0.03	0.02			
R5	SAMPLING TIME - 107 MIN. -- GRAMS INJECTED - 30.2				WASH CODE - 1		
	B1	1.49	1.38	1.54	1	0.94	0.93
	B4	1.38	1.8	1.98			
	B6	1.76	1.52	1.79			
	ARHCO	0.23	0.21	0.22			
	BACKGROUND	0.1	0.11	0.11			
R6	SAMPLING TIME - 105 MIN. -- GRAMS INJECTED - 29				WASH CODE - 1		
	B1	1.5	1.39	1.46	0.78	0.75	0.77
	B4	1.84	1.76	1.83			
	B6	1.74	1.66	1.74			
	ARHCO	0.35	0.32	0.32			
	BACKGROUND	0.015	0.022	0.027			
R7	SAMPLING TIME - 101 MIN. -- GRAMS INJECTED 30.6				WASH CODE - 3		
	B1	1.52	1.53	1.51	0.59	0.8	0.68
	B4	1.89	1.94	1.91	0.91	0.99	0.88
	B6	1.83	1.93	1.94	0.66	0.78	0.95
	ARHCO	0.42	0.42	0.43			
	BACKGROUND	0.026	0.04	0.044			
R10	SAMPLING TIME - 105 MIN. -- GRAMS INJECTED - 31				WASH CODE - 3		
	B1	1.56	1.62	1.56	0.4	0.48	0.4
	B4	1.86	1.95	1.91	0.53	0.49	0.48
	B6	1.84	1.92	1.96	0.7	0.56	0.64
	ARHCO	0.093	0.098	0.089			
	BACKGROUND	0.04	0.033	0.038			
R11	SAMPLING TIME - 111 MIN. -- GRAMS INJECTED - 29				WASH CODE - 3		
	B1	1.35	1.44	1.4	0.14	0.19	0.11
	B4	1.83	1.86	1.82	0.4	0.54	0.54
	B6	1.63	1.65	1.65	0.47	0.49	0.47
	ARHCO	0.061	0.039	0.058			
	BACKGROUND	0.015	0.03	0.028			
R12	SAMPLING TIME - 101 MIN. -- GRAMS INJECTED - 31.6				WASH CODE - 3		
	B1	1.57	1.52	1.51	0.34	0.34	0.37
	B4	2.12	2.1	2.09	0.89	0.93	1.06
	B6	1.92	1.91	1.88	0.44	0.33	0.47
	ARHCO	0.27	0.28	0.29			
	BACKGROUND	0.033	0.03	0.025			
NUMBER OF RUNS - 10		SAMPLER			FLOW SCFM	DEPOSIT cm^2	
STACK FLOWRATE - 300,000 SCFM		B1			0.92	10.02	
BACKGROUND FILTER DEPOSIT AREA - 17.35 cm^2		B4			1.04	10.29	
AVERAGE WASH BACKGROUND - 0.157 μg		B6			1.06	10.2	
		ARHCO			1.3	14.37	

APPENDIX F.4 Comparison Sampling, 296-Z-3 Stack, ZnS Tracer Data

RUN	SAMPLER LOCATION	FILTER ANALYSES			WASH ANALYSES		
		ZINC, $\mu\text{g}/\text{cm}^2$			ZINC, μg		
		1	2	3	1	2	3
R1	SAMPLING TIME - 62 MIN. -- GRAMS INJECTED - 5.1				WASH CODE - 3		
	B1	67.3	68.7	68.5	29.5	28.9	24.9
	B2	68.8	70.3	69.8	33.5	33.9	33.7
	B3	59.1	60.5	60.2	28.3	28.8	26.8
	ARMCO	0.45	0.43	0.46			
	BACKGROUND	0.015	0.002	0.006			
R2	SAMPLING TIME - 49 MIN. -- GRAMS INJECTED - 3.4				WASH CODE - 3		
	B1	45.7	46.7	46.3	83.8	85.2	81.9
	B2	53.2	54.7	54.1	73.3	71.1	71.8
	B3	41.9	42.9	42.2	119	116	105
	ARMCO	0.22	0.22	0.22			
	BACKGROUND	0.037	0.033	0.022			
R3	SAMPLING TIME - 56 MIN. -- GRAMS INJECTED - 3.5				WASH CODE - 3		
	B1	43.9	45.1	44.4	45.3	45	44.4
	B2	52.5	53.8	52.8	46.3	46.6	45.9
	B3	47.2	48.4	47.7	31.8	31.9	31.2
	ARMCO	0.24	0.21	0.21			
	BACKGROUND	0.01	0.017	0.011			
R4	SAMPLING TIME - 52 MIN. -- GRAMS INJECTED - 3.5				WASH CODE - 3		
	B1	40.7	41.5	41	70.3	70.5	69.2
	B2	52.1	53.3	52.8	60.8	61.7	61
	B3	40	41.1	40.5	50	50	48.6
	ARMCO	0.2	0.19	0.21			
	BACKGROUND	0.009	0.016	0.008			
R6	SAMPLING TIME - 50 MIN. -- GRAMS INJECTED - 3.5				WASH CODE - 3		
	B1	50	49.7	49.8	52.4	52.1	50.9
	B2	52.8	52.5	52.5	83.7	83.7	83
	B3	47.6	47.2	47.3	57.3	57.2	55.9
	ARMCO	0.28	0.29	0.27			
	BACKGROUND	0.021	0.016	0.024			
R7	SAMPLING TIME - 46 MIN. -- GRAMS INJECTED - 3.5				WASH CODE - 3		
	B1	49.2	48.9	49.1	60.5	60.2	60.9
	B2	51.2	50.5	51.1	46.4	45.8	46.7
	B3	40.8	40.1	40.7	52.1	60.9	60.9
	ARMCO	0.29	0.31	0.29			
	BACKGROUND	0.012	0.013	0.009			
R8	SAMPLING TIME - 55 MIN. -- GRAMS INJECTED - 3.7				WASH CODE - 1		
	B1	42.5	43.2	43.4	279	279	282
	B2	49.6	50.6	51.1			
	B3	36.3	36.5	36.8			
	ARMCO	0.28	0.27	0.28			
	BACKGROUND	0.034	0.033	0.024			
R9	SAMPLING TIME - 65 MIN. -- GRAMS INJECTED - 3.9				WASH CODE - 1		
	B1	47.4	48.1	48.3	231	238	242
	B2	55.2	56	56.5			
	B3	39.5	40.5	40.8			
	ARMCO	0.31	0.31	0.3			
	BACKGROUND	0.019	0.0093	0.011			
R10	SAMPLING TIME - 55 MIN. -- GRAMS INJECTED - 3.6				WASH CODE - 1		
	B1	42.6	43.7	43.9	303	301	308
	B2	52	53.1	53.8			
	B3	39.3	40.1	40.3			
	ARMCO	0.32	0.34	0.32			
	BACKGROUND	0.035	0.03	0.032			
R11	SAMPLING TIME - 46 MIN. -- GRAMS INJECTED - 3				WASH CODE - 1		
	B1	25.2	25.6	26	216	215	221
	B2	32.8	33.3	33.4			
	B3	25.8	25.8	26.1			
	ARMCO	0.2	0.21	0.23			
	BACKGROUND	0.025	0.023	0.009			
R12	SAMPLING TIME - 55 MIN. -- GRAMS INJECTED - 3.2				WASH CODE - 1		
	B1	39.2	37.2	39.1	236	227	235
	B2	48.6	46.3	49			
	B3	41.1	39	42.1			
	ARMCO	0.14	0.14	0.14			
	BACKGROUND	0.026	0.039	0.026			
		SAMPLER	FLOW SCFM	DEPOSIT cm^2			
NUMBER OF RUNS - 11		B1	1.38	9.8			
STACK FLOWRATE - 1983 SCFM		B2	1.34	10.36			
BACKGROUND FILTER DEPOSIT AREA - 17.35 cm^2		B3	1.41	10.25			
AVERAGE WASH BACKGROUND - 0.157 μg		ARMCO	1.53	57.17			

APPENDIX F.5

Analyses of Wash Solution Blanks

Blank Analyses - Micrograms

<u>Analysis Replicate</u>	<u>Zinc</u>				<u>Titanium</u>
	<u>#1</u>	<u>#2</u>	<u>#3</u>	<u>#4</u>	<u>#5</u>
1	.091	.21	.068	.25	.54
2	.018	.25	.17	--	.55
3	.11	.27	.13	--	.54

APPENDIX F.6

Tracer Concentration and Recovery, TiO₂, 291-Z-1 Stack

<u>Run</u>	<u>Location</u>	<u>Titanium Concentration μg/std. ft³</u>	<u>Percent Recovered</u>	<u>Ratio Battelle/ ARHCO</u>
R3	Battelle	.382686	97.1679	1.08007
	ARHCO	.354315	89.9641	
R4	Battelle	.347458	89.8191	1.05367
	ARHCO	.32976	85.2441	
R5	Battelle	.376704	97.3792	.92857
	ARHCO	.405682	104.87	
R6	Battelle	.408062	99.9358	.949964
	ARHCO	.429555	105.2	
R7	Battelle	.335279	89.5988	1.22085
	ARHCO	.274627	73.3903	
R8	Battelle	.187463	84.2046	1.31042
	ARHCO	.143056	64.2579	
R9	Battelle	.274036	78.3675	1.03792
	ARHCO	.264024	75.5044	
R11	Battelle	.292819	73.7477	.997889
	ARHCO	.293438	73.9037	
R12	Battelle	.27573	72.7937	1.0825
	ARHCO	.254716	67.246	

Average of Ratios = 1.07354, $S_x = .123$, $S_{\bar{x}} = .041$
 Battelle Average = 87.0016
 ARHCO Average = 82.1756
 Ratio of Averages = 1.05873

APPENDIX F.7

Tracer Concentration and Recovery,
TiO₂, 296-Z-3 Stack

<u>Run</u>	<u>Location</u>	<u>Titanium Concentration μg/std. ft³</u>	<u>Percent Recovered</u>	<u>Ratio Battelle/ ARHCO</u>
R1	Battelle	7.00092	81.2158	5.84361
	ARHCO	1.19805	13.8982	
R2	Battelle	9.30059	79.6091	15.1482
	ARHCO	.613975	5.25536	
R3	Battelle	8.58661	80.0445	5.38307
	ARHCO	1.59511	14.8697	
R4	Battelle	9.96951	80.5444	213.811
	ARHCO	4.66277E-2	.376709	
R5	Battelle	11.1951	86.806	5.46846
	ARHCO	2.0472	15.8739	
R6	Battelle	9.26559	79.656	5.75649
	ARHCO	1.60959	13.8376	
R7	Battelle	6.35845	66.3864	3.64511
	ARHCO	1.74438	18.2125	
R8	Battelle	11.6559	77.2966	7.94684
	ARHCO	1.46674	9.72671	
R9	Battelle	8.64538	70.7422	3.18955
	ARHCO	2.71053	22.1794	
R10	Battelle	8.60167	64.7647	2.88746
	ARHCO	2.97898	22.4297	
R11	Battelle	8.57009	66.4522	3.30082
	ARHCO	2.59636	20.132	

Average of Ratios = 24.7619, $S_x = 62.8$, $S_{\bar{x}} = 18.9$

Battelle Average = 75.7744

ARHCO Average = 14.2538

Ratio of Averages = 5.31608

APPENDIX F.8

Tracer Concentration and Recovery, ZnS, 291-Z-1 Stack

<u>Run</u>	<u>Location</u>	<u>Zinc Concentration $\mu\text{g}/\text{std. ft}^3$</u>	<u>Percent Recovered</u>	<u>Ratio Battelle/ ARHCO</u>
R1	Battelle ARHCO	.188323 2.29064E-2	35.0464 4.26283	8.22139
R2	Battelle ARHCO	.151251 9.02117E-3	38.5983 2.30214	16.7663
R3	Battelle ARHCO	.193711 1.51508E-2	35.9742 2.81366	12.7855
R4	Battelle ARHCO	.189698 1.45301E-2	35.1138 2.68958	13.0555
R5	Battelle ARHCO	.152462 8.45587E-3	28.5809 1.58516	18.0303
R6	Battelle ARHCO	.159535 2.34685E-2	30.5623 4.49587	6.79786
R7	Battelle ARHCO	.178378 3.05633E-2	31.1516 5.3375	5.83636
R10	Battelle ARHCO	.169768 4.28312E-3	30.4243 .767585	39.6364
R11	Battelle ARHCO	.147904 2.03779E-3	29.9533 .412688	72.5808
R12	Battelle ARHCO	.186147 1.98134E-2	31.4796 3.35067	9.39501

Average of Ratios = 20.3106, $S_x = 20.8$, $S_{\bar{x}} = 6.57$

Battelle Average = 32.6885

ARHCO Average = 2.80177

Ratio of Averages = 11.6671

APPENDIX F.9

Tracer Concentration and Recovery, ZnS, 296-Z-3 Stack

<u>Run</u>	<u>Location</u>	<u>Zinc Concentration μg/std. ft³</u>	<u>Percent Recovered</u>	<u>Ratio Battelle/ ARHCO</u>
R1	Battelle ARHCO	8.17184 .264575	34.7441 1.12489	30.8866
R2	Battelle ARHCO	8.47329 .14438	42.708 .72772	58.6873
R3	Battelle ARHCO	6.90397 .138343	38.633 .774136	49.9047
R4	Battelle ARHCO	7.19003 .135811	37.3599 .705684	52.9414
R6	Battelle ARHCO	8.27985 .194054	41.3679 .969537	42.6677
R7	Battelle ARHCO	8.37618 .22907	38.5013 1.05293	36.5661
R8	Battelle ARHCO	7.03325 .167354	36.5643 .870037	42.0261
R9	Battelle ARHCO	6.32538 .16876	36.8702 .983691	37.4815
R10	Battelle ARHCO	7.42192 .199965	39.6567 1.06845	37.1161
R11	Battelle ARHCO	5.6667 .157858	30.3883 .84653	35.8975
R12	Battelle ARHCO	6.70631 7.45056E-2	40.3122 .447859	90.0109

Average of Ratios = 46.7442, $S_x = 16.6$, $S_{\bar{x}} = 5.00$
 Battelle Average = 37.9187
 ARHCO Average = .870133
 Ratio of Averages = 43.5781

APPENDIX F.10

Comparison Sampling Experiment Calculations

A computer program, COMEXP, was written to perform the calculations described below. The tabulated data were printed in computer format, hence the excessive number of significant digits in most cases.

MEMBRANE FILTERS

The triplicate analyses of the blank membrane filter analyzed with each run (BKGR) were first averaged. This value was then subtracted from each membrane filter analyzed from that run. The background corrected triplicate analyses of each membrane filter were then averaged. This averaged analysis was then multiplied by the filter deposit area to yield the total tracer in μg (titanium or zinc) on each membrane filter. In the case of the control samples the totals from each of the three filters were added to yield the total tracer collected on control sample membrane filters.

CONTROL SAMPLE NOZZLE WASHES

The tracer contents of blank wash solutions (see Appendix F.5) were averaged. This average was subtracted from the wash analyses. The triplicate analyses were then averaged. In cases when the three nozzle washes from a run were analyzed separately (WASH CODE = 3), the averaged analyses were added to yield the total tracer collected in nozzles during a run. In cases where the washes were combined before analysis (WASH CODE = 1) the averaged analysis yielded the total per run. In two instances when it was intended to analyze the nozzle washes separately, one wash solution was lost (WASH CODE = 2).

In these two instances an estimated average analysis was calculated as a flowrate weighted average of the averaged analyses of the two remaining washes. Calculations then proceeded as for samples with WASH CODE = 3.

TRACER CONCENTRATION

For the control samples, the total amounts of tracer in the nozzle washes and on the membrane filters were added, yielding the total tracer collected by the control samplers. The total tracer on the membrane filter from ARHCO's existing sampler constituted the total collected by that sampler, as the probe was not washed. Tracer concentration in $\mu\text{g}/\text{std. ft}^3$ was then calculated by dividing the total tracer collected by the respective total flowrate and sampling time. The total flowrates were:

	<u>Control</u>	<u>ARHCO</u>
291-Z-1 Stack	3.02 scfm	1.80 scfm
296-Z-3 Stack	4.13 scfm	1.53 scfm

TRACER RECOVERY

The percent tracer recovered was calculated from the total tracer collected by the control or ARHCO samplers as follows:

$$\% \text{ Recovered} = \frac{\text{Total } \mu\text{g} \times \text{Stack Flowrate scfm}}{\text{Total Sample Flowrate scfm} \times \text{Injected } \mu\text{g} \times P} \times 100 \quad (\text{F-1})$$

where P is the weight fraction of zinc (0.567) or titanium (0.547) in the tracer material as determined by x-ray fluorescence analyses. The total sample flowrates are given in the table above and the stack flowrates were taken as 300,000 scfm and 1983 scfm for the 291-Z-1 and 296-Z-3 stacks respectively.

The tabulated ratios of recovered tracer are exactly equivalent to the ratios of concentrations.

APPENDIX G

PARTICLE SIZE DATA

APPENDIX G.1

Data for High Flowrate Cascade Impactor, 291-Z-1 Stack

				SAMPLE COMPONENT	d/min Pu	AERODYNAMIC CUTOFF DIAMETER, μm	Pu CONCENTRATION $\mu\text{Ci}/\text{mi}^3$ *
<u>RUN 1</u>							
	<u>MONTH</u>	<u>DAY</u>	<u>TIME</u>	INLET	12,500		
ON	10	20	1203	COLLECTION SURFACE NO. 1	7,030	7.0	
				COLLECTION SURFACE NO. 2	4,290	3.3	
OFF	11	3	0900	COLLECTION SURFACE NO. 3	1,300	2.0	
				COLLECTION SURFACE NO. 4	970	1.1	
ELAPSED TIME = 19,977 min				BACK-UP FILTER	970		
				TOTAL	27,060		$1.1 (10^{-12})$
				% IN INLET	46%		
				AMAD = $6.8 \mu\text{m}$ (15)**			
<u>RUN 2</u>							
	<u>MONTH</u>	<u>DAY</u>	<u>TIME</u>	INLET	3,500		
ON	11	4	0910	COLLECTION SURFACE NO. 1	1,400	7.0	
				COLLECTION SURFACE NO. 2	620	3.3	
OFF	11	18	1025	COLLECTION SURFACE NO. 3	200	2.0	
				COLLECTION SURFACE NO. 4	100	1.1	
ELAPSED TIME = 20,235 min				BACK-UP FILTER	81		
				TOTAL	5,901		$2.3 (10^{-13})$
				% IN INLET	59%		
				AMAD = $8.6 \mu\text{m}$ (26)			
				ARHCO	24		$1.1 (10^{-14})$
	<u>MONTH</u>	<u>DAY</u>	<u>TIME</u>				
ON	11	4	0800				
OFF	11	18	0830				
ELAPSED TIME = 20,190 min							
<u>RUN 3</u>							
	<u>MONTH</u>	<u>DAY</u>	<u>TIME</u>	INLET	5,390		
ON	1	21	1055	COLLECTION SURFACE NO. 1	(6,000) [†]	7.0	
				COLLECTION SURFACE NO. 2	3,480	3.3	
OFF	2	4	0855	COLLECTION SURFACE NO. 3	2,410	2.0	
				COLLECTION SURFACE NO. 4	1,340	1.1	
ELAPSED TIME = 20,040 min				BACK-UP FILTER	5,240		
				TOTAL	23,860		$9.5 (10^{-13})$
				% IN INLET	23%		
				AMAD = $3.5 \mu\text{m}$ (6.2)			
				ARHCO	74		$4.3 (10^{-14})$
	<u>MONTH</u>	<u>DAY</u>	<u>TIME</u>				
ON	1	21	1000				
OFF	2	4	0900				
ELAPSED TIME = 19,620 (EIGHT HOUR SAMPLE MISSING)							

*VOLUME IS MEASURED AT 70°F, 1 atm WHICH IS APPROXIMATELY THE EFFLUENT CONDITIONS. CONCENTRATION IS CALCULATED AS FOLLOWS:

$$\text{CONCENTRATION } \mu\text{Ci}/\text{mi}^3 = \frac{d}{\text{min}} \times \frac{\mu\text{Ci}-\text{min}}{2.22(10^6)\text{d}} \times \frac{\text{min}}{\text{SF ft}^3} \times \frac{1}{\Delta t \text{ min}} \times \frac{\text{ft}^3}{28,320 \text{ mi}}$$

WHERE SF IS THE SAMPLE FLOWRATE IN SCFM. FOR THE IMPACTOR SF = 20 SCFM AND FOR ARCHO'S SAMPLE SF = 1.8 SCFM FOR RUNS 1 AND 2 AND 1.4 SCFM FOR RUN 3.

**THE AMAD'S WERE OBTAINED FROM LOG NORMAL PLOTS OF IMPACTOR DATA. SAMPLE INTERMEDIATE CALCULATIONS ARE SHOWN ON THE FOLLOWING PAGES. THE INLET LOSSES WERE IGNORED ON THE ASSUMPTION THAT PARTICULATES LOST IN THE INLET WERE OF THE SAME SIZE DISTRIBUTION AS THOSE THAT REACHED THE IMPACTOR. IT MAY BE MORE REASONABLE TO ASSUME THAT THE PARTICULATES DEPOSITED IN THE INLET ARE LARGER THAN THOSE NOT DEPOSITED. THE AMAD'S SHOWN IN THE PARENTHESES WERE OBTAINED WHEN THE ACTIVITY DEPOSITED IN THE INLET WAS ADDED TO THE ACTIVITY ON THE FIRST COLLECTION SURFACE (THE SURFACE WITH THE LARGEST CUTOFF DIAMETER) AND THE PERCENT PER STAGE WAS RECALCULATED.

†AN ESTIMATE BECAUSE PART OF THE SAMPLE WAS LOST DURING ANALYSIS

APPENDIX G.2 Data For Cascade Impactors, 296-Z-3 Stack

				SAMPLE COMPONENT	d/min Pu	AEROODYNAMIC CUTOFF DIAMETER, μ m	Pu CONCENTRATION μ Ci/ml *
<u>HIGH FLOWRATE IMPACTOR</u>							
<u>RUN 1</u>				INLET	59.4		
				COLLECTION SURFACE NO. 1	162	7.0	
				COLLECTION SURFACE NO. 2	211	3.3	
<u>ON</u>				COLLECTION SURFACE NO. 3	122	2.0	
				COLLECTION SURFACE NO. 4	121	1.1	
<u>OFF</u>				BACK-UP FILTER	160		
ELAPSED TIME - 10.010 min				TOTAL	835		$5.9 (10^{-14})$
				% IN INLET	7%		
				AMAD - 3.1 (3.5) μ m			
ARHCO					100		$9.1 (10^{-14})$
				<u>MONTH DAY TIME</u>			
<u>ON</u>				4 21 1010			
<u>OFF</u>				4 28 0900			
ELAPSED TIME - 10.080 min							
<u>RUN 2</u>				INLET (NOZZLE WASH LOST)	671		
				COLLECTION SURFACE NO. 1	1060	7.0	
				COLLECTION SURFACE NO. 2	749	3.3	
<u>ON</u>				COLLECTION SURFACE NO. 3	784	2.0	
				COLLECTION SURFACE NO. 4	512	1.1	
<u>OFF</u>				BACK-UP FILTER	1077		
ELAPSED TIME - 33.090 min				TOTAL	4953		$1.0 (10^{-13})$
				% IN INLET	14%		
				AMAD - 2.8 μ m (3.6)			
<u>RUN 3</u>				INLET	124.8		
				COLLECTION SURFACE NO. 1	142	7.0	
				COLLECTION SURFACE NO. 2	154	3.3	
<u>ON</u>				COLLECTION SURFACE NO. 3	182	2.0	
				COLLECTION SURFACE NO. 4	59.9	1.1	
<u>OFF</u>				BACK-UP FILTER	127		
ELAPSED TIME - 19.914 min				TOTAL	789.4		$2.7 (10^{-14})$
				% IN INLET	16%		
				AMAD - 3.0 μ m (3.7)			
<u>LOW FLOWRATE IMPACTOR</u>							
<u>RUN 1</u>				INLET	16.5 ± 1.3		
				COLLECTION SURFACE NO. 0	6.41 ± 0.86	15	
				COLLECTION SURFACE NO. 1	7.65 ± 0.95	7.9	
<u>ON</u>				COLLECTION SURFACE NO. 2	1.04 ± 0.41	3.6	
				COLLECTION SURFACE NO. 3	8.52 ± 1.00	2.2	
<u>OFF</u>				COLLECTION SURFACE NO. 4	3.44 ± 0.64	1.1	
				COLLECTION SURFACE NO. 5	3.02 ± 0.61	0.56	
ELAPSED TIME - 25.940 min				BACK-UP FILTER	9.82 ± 1.95		
				TOTAL	56.4		$3.4 (10^{-14})$
				% IN INLET	29%		
				AMAD - 2.7 μ m (9.6)			
ARHCO					151		$5.1 (10^{-14})$
				<u>MONTH DAY TIME</u>			
<u>ON</u>				1 31 1030			
<u>OFF</u>				2 19 0800			
ELAPSED TIME - 27.210 min							

* VOLUME MEASURED AT EFFLUENT CONDITIONS -- 150°F, 1 atm
CONCENTRATION IS CALCULATED AS FOLLOWS:

$$\text{CONCENTRATION } \mu\text{Ci/ml} = \frac{d}{\text{min}} \times \frac{\mu\text{Ci-min}}{2.2210^{10} \text{ d}} \times \frac{\text{min}}{\text{SF ft}^3} \times \frac{1}{\Delta t \text{ min}} \times \frac{\text{ft}^3}{28.320 \text{ ml}}$$

WHERE SF IS THE SAMPLE FLOWRATE IN CFM AT 150°F, 1 atm

	SF, CFM
HIGH FLOWRATE IMPACTOR	23
LOW FLOWRATE IMPACTOR	1.01
ARHCO SAMPLER	1.73

* THE AMAD'S WERE OBTAINED FROM LOG-NORMAL PLOTS OF IMPACTOR DATA. SAMPLE INTERMEDIATE CALCULATIONS ARE SHOWN ON THE FOLLOWING PAGE. THE INLET LOSSES WERE IGNORED ON THE ASSUMPTION THAT PARTICULATES LOST IN THE INLET WERE OF THE SAME SIZE DISTRIBUTION AS THOSE THAT REACHED THE IMPACTOR. IT MAY BE MORE REASONABLE TO ASSUME THAT THE PARTICULATES DEPOSITED IN THE INLET ARE LARGER THAN THOSE NOT DEPOSITED. THE AMAD'S SHOWN IN PARENTHESES WERE OBTAINED WHEN THE ACTIVITY DEPOSITED IN THE INLET WAS ADDED TO THE ACTIVITY ON THE FIRST COLLECTION SURFACE (THE SURFACE WITH THE LARGEST CUTOFF DIAMETER) AND THE PERCENT PER STAGE WAS RECALCULATED.

APPENDIX G.3

Particle Size Distribution Sample Calculations

Example 1. Run 1 291-Z-1 Stack

<u>Sample</u>	<u>d/min</u>	<u>%</u>	<u>Cumulative %</u>	<u>Aerodynamic Equivalent Cutoff Diam. μm</u>
Collection Surface #1	7,030	48.3	100.1	7.0
Collection Surface #2	4,290	29.5	51.8	3.3
Collection Surface #3	1,300	8.9	22.3	2.0
Collection Surface #4	970	6.7	13.4	1.1
Back-up Filter	<u>970</u>	6.7	6.7	
Total	14,560			

This distribution is plotted in Figure 25. The cumulative percent is plotted versus the cutoff diameter of the next stage, i.e., 6.7% is plotted versus 1.1 μm . The AMAD (6.8 μm) is taken from the plot where the plotted curve intersects the 50% abscissa. This is an example where inlet losses are ignored. Alternatively it can be reasoned that the material lost in the inlet was of a particle size larger than the largest size reaching the impactor stages. Following this reasoning, the activity found in the inlet components can be added to the first stage as in the next example.

Example 2. Low Flowrate Impactor, 296-Z-3 Stack

<u>Sample</u>	<u>d/min</u>	<u>%</u>	<u>Cumulative %</u>	<u>Aerodynamic Equivalent Cutoff Diam. μm</u>
Inlet & Collection Surface #0	22.91	40.6	100.0	15
Collection Surface #1	7.65	13.6	59.4	7.9
Collection Surface #2	1.04	1.8	45.8	3.6
Collection Surface #3	8.52	15.1	44.0	2.2
Collection Surface #4	3.44	6.1	28.9	1.1
Collection Surface #5	3.02	5.4	22.8	0.56
Back-up Filter	<u>9.82</u>	17.4	17.4	
Total	56.4			

The data are plotted and the AMAD is found in the same manner as explained above. The AMAD is 9.6 μm , the value in parentheses shown in the data table.

APPENDIX H

DATA ON TOTAL ALPHA AND PLUTONIUM EMISSIONS

Run Name	mo-day-hour Battelle Sample		mo-day-hour ARHCO Sample		Battelle Sample			Battelle Analysis of ARHCO Samples		ARHCO Reported Alpha Ci***
	On	Off	On	Off	dpm Pu	cfm	Pu Ci* Emitted	dpm Pu	Pu Ci** Emitted	
291-BK-1	2-9-1100	2-12-0940	2-9-0800	2-12-0800	10.93	1.2	9.7E-7	--	--	1.3E-6
291-BK-2	2-12-0950	2-19-0920	2-12-0800	2-19-0800	1870	1.2	1.7E-4	--	--	9.0E-6
291-BK-3	2-19-0930	2-27-0940	2-19-0800	2-27-0800	427	1.2	3.8E-5	--	--	5.9E-6
291-BK-4	2-27-0945	3-1-0850	2-27-0800	3-1-0800	70.8	1.2	6.3E-6	--	--	1.3E-6
291-HI-R1	10-20-1203	11-3-0900	10-20-0830	11-2-0800	27,060	20	1.9E-4	--	--	1.3E-5
291-HI-R2	11-4-0910	11-18-1025	11-4-0800	11-18-0830	5,901	20	4.1E-5	24	1.8E-6	2.0E-5
291-HI-R3	1-21-1055	2-4-0855	1-21-1000	2-4-0900	23,860	20	1.6E-4	74	5.6E-6	1.4E-4

$$^* \text{Ci Emitted} = \frac{d}{\text{min.}} \times \frac{\text{Ci min.}}{2.22(10^{12}) d} \times \frac{\text{Stack Flowrate, cfm}}{\text{Sample Flowrate, cfm}} \quad (\text{H-1})$$

All cfm in table are at 70°F, 1 atm. The stack flowrates obtained from ARHCO were 236,552 cfm when background samples were taken and 305,235 cfm when Impactor Samples were taken.

** Same as Equation H-1 except ARHCO's sample flowrate was 1.8 cfm.

*** Same as Equation H-1 but ARHCO's assumed sample flowrate was 2 cfm. Also, ARHCO assumed a sampling efficiency of 70%, thus increasing reported releases by 1/0.7. These release values are from ARHCO's AIRSAM report.

Sample missing in Battelle's analysis. According to ARHCO's analysis that sample accounted for about 1/3 of the total activity released during this run.

NOTE: To save space computer format used for scientific notation, thus E-6 equals 10^{-6} .

APPENDIX H-1. Comparison of Samples Analyzed for Alpha Emissions from 291-Z-1 Stack

Run Name	mo-day-hour Battelle Sample		mo-day-hour ARHCO Sample		Battelle Sample			Battelle Analysis of ARHCO Samples		ARHCO Reported Alpha Ci*** Emitted
	On	Off	On	Off	dpm Pu	cfm	Pu Ci* Emitted	dpm Pu	Pu Ci** Emitted	
241-BK-1	2-9-1030	2-12-0927	2-9-0830	2-12-0900	11.0	1.7 (1.5 scfm)	7.2E-9	--	--	1.8E-8
241-BK-2	2-12-0935	2-19-0910	2-12-0900	2-19-0900	20.1	1.7	1.3E-8	--	--	2.2E-9
241-BK-3	2-19-0920	2-27-0950	2-19-0900	2-27-0900	22.9	1.7	1.5E-8	--	--	5.7E-8
241-BK-4	2-27-0950	3-1-0920	2-27-0900	3-1-0830	8.26	1.7	5.4E-9	--	--	1.0E-8
241-HI-R1	4-21-1010	4-28-0900	4-21-0830	4-28-0830	835	23 (20 scfm)	4.1E-8	100	6.6E-8	9.4E-8
241-HI-R2	4-29-0930	5-21-0900	4-29-0830	5-21-0900	4,853	23	2.4E-7	--	--	4.8E-7
241-HI-R3	6-8-1535	6-22-1129	6-8-0800	6-23-0900	789.4	23	3.8E-8	--	--	1.0E-7
241-LI-R1	1-31-1040	2-18-1100	1-31-1030	2-19-0800	56.4	1.0 (0.87 scfm)	6.3E-8	151	9.9E-8	2.0E-7

$$^* \text{Ci Emitted} = \frac{d}{\text{min.}} \times \frac{\text{Ci min.}}{2.22(10^{12})d} \times \frac{\text{Stack Flowrate, 2481 cfm}}{\text{Sample Flowrate, cfm}}$$

(H-2)

All cfm in table are at 150°F, 1 atm. The stack flowrate was obtained from ARHCO.

** Same as Equation H-2 except ARHCO's sample flowrate was 1.5 scfm or 1.7 cfm.

*** Same as Equation H-2 except ARHCO assumed a sample flowrate of 1.5 cfm. ARHCO also assumed a sampling efficiency of 70%, thus increasing reported releases by 1/0.7. These release values are from ARHCO's AIRSAM report.

NOTE: To save space, computer format used for scientific notation, i.e., E-6 equals 10^{-6} .

APPENDIX H-2. Comparison of Samples Analyzed for Alpha Emissions from 296-Z-3 Stack

APPENDIX I

INTERIM RADIOACTIVE GASEOUS EFFLUENT SAMPLING AND MONITORING
CRITERIA FOR ARHCO FACILITIES

Date: November 12, 1975
To: Distribution
From: B. J. McMurray *BJ McMurray*
Subject: INTERIM RADIOACTIVE GASEOUS EFFLUENT SAMPLING AND
MONITORING CRITERIA FOR ARHCO FACILITIES

In a continuing effort to report radioactive emissions to the environment as accurately as practicable, the performance of ARHCO's gaseous effluent sampling and monitoring systems is being studied by the Particulate and Gaseous Waste Research Section of Battelle-Northwest. This study has revealed several problems with our existing sampling systems.

In order to upgrade the performance of ARHCO's sampling and monitoring systems, positive goals must be set that will ensure that changes proceed in a direction that will result in improvements of the systems. The attached interim gaseous effluent sampling and monitoring criteria have been developed to aid those involved in the design, fabrication, and operation of these systems. It must be recognized that portions of the criteria are not easy to meet unless the sampling and monitoring systems are carefully planned and designed with these goals in mind.

The effectiveness of the attached interim criteria will be evaluated as improvements in existing systems are made, or as new systems are brought on line, to determine if changes in the criteria are necessary.

BJM:DJC:bac

Distribution

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INTERIM RADIOACTIVE
GASEOUS EFFLUENT SAMPLING AND MONITORING CRITERIA
FOR ATLANTIC RICHFIELD HANFORD COMPANY FACILITIES

INTRODUCTION

To ensure that gaseous radioactive releases to the environment are kept as low as practicable, it is necessary to sample all of the radioactive or potentially radioactive gaseous effluent streams in the 200 Areas. Specific gaseous effluent streams that have the potential for release of large quantities of radioactive materials must also be monitored by radiation detection instrumentation.

The criteria listed below are intended to be used as guidelines to aid in the design of new systems or the upgrading of existing systems. It is highly desirable that new or upgraded systems be able to meet or nearly approach as many of the following criteria as is practicable.

SAMPLING CRITERIA

The term "sampling" applies to the extraction, transport and collection of a sample of the radionuclides present in the gaseous effluent stream on a collection media suitable for subsequent laboratory analysis. Gaseous effluent sampling systems shall:

1. extract in a 24-hour period a representative fraction of the effluent stream such that an activity release at least as low as the concentration guides given in ERDA Appendix 0524, Annex A, Table II can be measured with appropriate accuracy upon subsequent laboratory analysis.*
2. be designed, fabricated, and operated so that the particles extracted and transported to the collector will be representative to the degree that particles 10 micrometers and smaller in the sample collected will be at least 90 percent of the 10 micrometer and smaller particles in the same volume of a truly representative sample. At least 90 percent of the radioactive gases withdrawn shall be collected on the media selected or delivered to the activity sensing chamber in flow through monitors.
3. operate in a continuous mode, except for short periods when the sample collection media is being changed, whenever the facility is exhausting to the atmosphere.
4. extract samples of particles and gases that are a representative aliquot of the actual effluent stream.

* If one half of the Class I Action Level in ARH-222, ARHCO Emergency Procedure 1.9 (or the Emergency Procedure that governs gaseous discharges for the specific facility) is less than the Table II concentration guides, use one half of the Class I Action Level from the appropriate Emergency Procedure as the basis for this criterion.

5. collect at least two samples, one of which will be designated and used exclusively as an inventory sample.
6. shall provide particulate and gaseous samples that have not been adversely affected by condensation of moisture.
7. be equipped with a vacuum system for sample extraction that has the same number of back-up power systems as the exhaust fans of the facility. The vacuum system shall be provided with an alarm that is activated upon the loss of vacuum.
8. provide an inventory sample which is an accurate aliquot, within a factor of two, of the absolute radioactive materials of the effluent stream. (Note: This statement relates only to the sample extraction and delivery capability, not to the accuracy of detection and measurement.)
9. provide inventory samples with less than 25 percent variation from the mean of several similar samplings at the 95 percent confidence level.
10. have at least an annual check to ensure that variation in effluent characteristics (particle size, mass activity concentrations, etc.) have not changed significantly.

MONITORING CRITERIA

The term "monitoring" applies to the use of radiation detection instrumentation to measure the amount of radioactivity collected by the collection media. Gaseous effluent monitoring systems shall:

1. monitor the samples collected (except for the designated inventory sample) as detailed in items 1-10 under Sampling Criteria. The monitoring system shall be able to detect an activity release at least as low as one half that required for the Class I Action Level listed in ARH-222, ARHCO Emergency Procedure 1.9 (or the Emergency Procedure that governs gaseous discharges for the specific facility).
2. detect the radiation from radionuclides in the effluent stream with appropriate sensitivity for the radiation emitted.
3. operate in a continuous mode, except for short periods while samples are being changed, whenever the facility is exhausting to the atmosphere.
4. be equipped with the same number of back-up power systems as the exhaust fans of the facility.
5. incorporate a permanent recorder to document the level of radioactivity measured by the radiation detection instrument.

6. be provided with a "fail-safe" system that alarms when a malfunction occurs in the radiation detection instrument or upon loss of vacuum.
7. alarm at a pre-set "trip" point that is consistent with a Class II Action Level as specified in ARH-222, ARHCO Emergency Procedure 1.9 (or the Emergency Procedure that governs gaseous discharges for the specific facility).
8. be able to detect a Class II Action Level as specified in ARH-222, ARHCO Emergency Procedure 1.9 (or the Emergency Procedure that governs gaseous discharges for the specific facility) in less than 24 hours.
9. alarm visually and audibly in a continuously occupied area for both the "fail-safe" alarm(s) and the emergency alarm(s).
10. be equipped with sufficient range to detect activity levels up to 100 times the Class II Action Level in ARH-222, ARHCO Emergency Procedure 1.9 (or the Emergency Procedure that governs gaseous discharges for the specific facility).
11. have the radiation detection instrumentation calibrated on at least a quarterly basis.

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