

EEG-88



APPLICATION OF BERYLLIUM-7 AND LEAD-210 CONCENTRATION MEASUREMENTS TO MONITORING THE VALIDITY OF EFFLUENT AIR SAMPLING AT THE WASTE ISOLATION PILOT PLANT

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Environmental Evaluation Group
New Mexico

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FOREWORD

The purpose of the New Mexico Environmental Evaluation Group (EEG) is to conduct an independent technical evaluation of the Waste Isolation Pilot Plant (WIPP) Project to ensure the protection of the public health and safety and the environment of New Mexico. The WIPP Project, located in southeastern New Mexico, became operational in March 1999 for the disposal of transuranic (TRU) radioactive wastes generated by the national defense programs. The EEG was established in 1978 with funds provided by the U.S. Department of Energy (DOE) to the State of New Mexico. Public Law 100-456, the National Defense Authorization Act, Fiscal Year 1989, Section 1433, assigned the EEG to the New Mexico Institute of Mining and Technology and continued the original contract DE-AC04-79AL10752 through DOE contract DE-AC04-89AL58309. The National Defense Authorization Act for Fiscal Year 1994, Public Law 103-160, and the National Defense Authorization Act for Fiscal Year 2000, Public Law 106-65, continued the authorization.

EEG performs independent technical analyses on a variety of issues. Now that the WIPP is operational, these issues include facility modifications and waste characterization for future receipt and emplacement of remote-handled waste, generator site audits, contact-handled waste characterization issues, the suitability and safety of transportation systems, mining of new panels, analysis of new information as part of the five year recertification cycles as mandated by the WIPP Land Withdrawal Act. Review and comment is also provided on the annual Safety Analysis Report and Proposed Modifications to the Hazardous Waste Facility Permit. The EEG also conducts an independent radiation surveillance program which includes a radiochemical laboratory.



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ACRONYMS AND SYMBOLS

Be	Beryllium
Bq	Becquerel
CEMRC	Carlsbad Environmental Monitoring and Research Center
DOE	US Department of Energy
EEG	Environmental Evaluation Group
NESHAPS	National Emissions Standards for Hazardous Airborne Pollutants
Pb	Lead
TRU	Transuranic
TSP	Total Suspended Particulate
WIPP	Waste Isolation Pilot Plant
WTS	Washington TRU Solutions

EXECUTIVE SUMMARY

The Waste Isolation Pilot Plant (WIPP) is an underground repository in southeast New Mexico owned and operated by the U.S. Department of Energy (DOE) for the purpose of safely disposing of transuranic (TRU) waste materials generated by the national defense programs. During normal operations, the underground rooms and drifts at WIPP are ventilated by three large induced draft exhaust fans. These fans pull approximately 425,000 cu ft/min down an intake shaft, through the repository, up a 4.3-meter-diameter exhaust shaft and out one or more exhaust vents to the atmosphere. A sampling station, called Station A, has been established near the top of the exhaust shaft so that a sample of the effluent air may be extracted immediately before the effluent air is discharged to the environment.

Over the past ten years two observations have generated concern regarding the validity of the aerosol sampling regime at Station A. First, as early as April 1993, there were indications that moisture in the exhaust shaft was interfering with the operation of hot-wire flow sensors (MVS 1993). Subsequently, video inspections of the probes and shaft liner revealed a build-up of salt deposits on the probes, water flowing into the shaft through several cracks in the liner and water droplets entrained in the exhaust airflow being carried up toward the probes. The probes were pulled on a regular basis for inspection and cleaning and were often found to be wet. One concern has been that the actual sampling conditions resulting from wet probes and sample lines at Station A have not been approximated by the test conditions used to characterize the operation of the sampling system and, therefore, the ability of the system to extract a representative sample from the exhaust airstream under all conditions was unproven.

Second, because of the moisture in the exhaust shaft, salt build-up on the sampling probe might reduce the efficacy of the sampling regime.

In September 2003 the Environmental Evaluation Group (EEG) began to reexamine archived data from analysis by gamma spectrometry of filter samples from Skid A-3 of Station A and from a more recently established sampling station, called Station D. Station D, located in the

E300 drift near the bottom of the exhaust shaft, samples air coming down the E300 drift leading from the active repository rooms. The E300 airflow accounts for about 70% of the air discharged from the exhaust shaft during normal operations. Station D conforms to the requirements of ANSI N13.1-1999 (Gadbury 2001) for representative sampling. Furthermore, Station D uses the same sample head design and the same sample media (filters) as are used at Station A, but is unaffected by water inflow to the exhaust shaft.

The purpose of the reexamination was to compare the efficacy of the sampling regime at Skid A-3 with that of the uncompromised sampling regime at Station D by tracking and comparing the measured concentrations of ^7Be and ^{210}Pb at the two sampling locations. Both ^7Be and ^{210}Pb are naturally-occurring radionuclides found in ambient air and are associated with the smaller (<2 micron diameter) aerosol particles. The results of the EEG analysis show that the <2 micron aerosol concentrations, using ^7Be and ^{210}Pb as tracers, were approximately the same for the samples of air drawn through the sampling filters at both Skid A-3 and Station D. Statistical examination of these data confirmed the two sampling stations to be highly correlated for each radionuclide, with correlation coefficients of 0.913 ($p = 0.004$) for ^7Be and 0.866 ($p = 0.000$) for ^{210}Pb .

This report assumes uniform mixing of the effluent aerosol across the exhaust shaft. With that caveat, these results support a conclusion that the presence of water in the exhaust shaft and salting up of the sampling probe during the period from September 2001 to the present had little effect on the collection of <2 micron aerosol particles at Skid A-3 and, consequently, for that period and for that aerosol size fraction, Station A measurements should be considered to be representative of the effluent airstream. Further work is needed and planned to determine whether this conclusion can be extended to include particles in the 2-10 micron range.

INTRODUCTION

The U.S. Department of Energy's (DOE) Waste Isolation Pilot Plant (WIPP) in southeast New Mexico is the nation's first nuclear waste repository dedicated to the permanent safe disposal of waste materials contaminated with transuranic (TRU) radionuclides from the national defense programs. The WIPP was designed and constructed to bury TRU waste 650 meters (2150 feet) underground in a 600-meter-thick (2000-foot-thick) layer of bedded salt called the Salado formation. At closure it is estimated the WIPP will contain over 6 million curies of radioactivity, with about half of the total from isotopes of plutonium and americium. The WIPP received its first shipment of waste in March 1999 and as of October 2, 2003, contained 15,299 m³ of waste materials (DOE 2003).

During normal operations the underground rooms and drifts at WIPP are ventilated by large induced draft exhaust fans which move air at a rate of approximately 425,000 cu ft/min through the repository, up a 4.3-meter-diameter exhaust shaft, and out one or more exhaust vents to the atmosphere. A sampling station, called Station A, has been established at the top of the exhaust shaft so that a sample of the effluent air may be extracted immediately before it is released to the environment. Station A consists of three independent sampling lines and instrument platforms called skids, supporting pumps, filter holders, and flow-monitoring equipment. The sampled air is split into three equal fractions which are each passed through sample filters at the rate of 2 cu ft/min to collect suspended aerosol particles. Efforts have been aimed at insuring that all three filter samples are true replicates. Each day the filters are collected by personnel from Washington TRU Solutions (WTS), the DOE's management and operating contractor for WIPP; the Carlsbad Environmental Monitoring and Research Center; and the New Mexico Environmental Evaluation Group (EEG), a group of scientists and engineers mandated by Congress to carry out independent technical oversight of DOE activities at WIPP. The filters are then analyzed for various alpha-, beta-, and gamma-emitting radionuclides. The EEG uses the results of its analyses to verify WIPP's compliance with federal standards limiting the public's annual committed effective dose from WIPP operations, codified in 40 CFR 61 Subpart H, the National Emissions Standards for Hazardous Airborne Pollutants (NESHAPS).

STATION A

Figure 1 depicts the top of the exhaust shaft and the sampling location at Station A. As shown, three independent sampling lines have been established. These are labeled A-1, A-2, and A-3. The lines are located at the east, west, and south sides of the exhaust shaft, approximately 2 feet from the shaft liner. Currently, the line on the south side of the shaft (A-3) is the designated “skid of record” for determination of compliance with NESHAPS. Each sample line delivering sampled air to the splitter block in the enclosure extends into the exhaust airstream 21 feet below ground surface. The lines themselves are 2-inch diameter steel tubes. The lower end of the lines support a shrouded probe assembly (see Figure 2) designed and tested by Texas A&M University (McFarland et al. 1988) to extract a representative sample of the effluent air. The suspended aerosol particles are collected on Versapore[®] filters having a 3-micron absolute pore diameter.

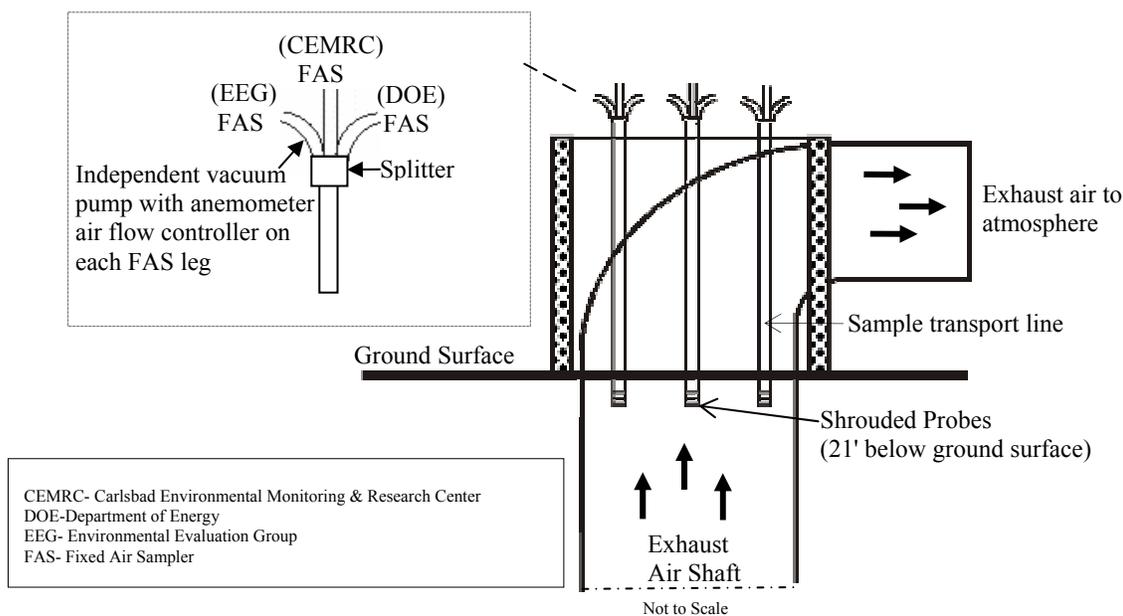


Figure 1. Exhaust Shaft and Station A

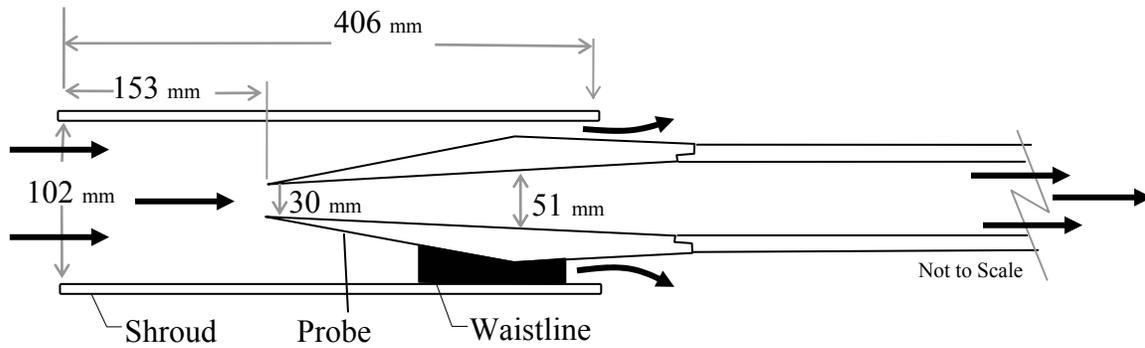


Figure 2. Station A Probe

In operation, the shrouded probe can accumulate deposits of salt and other minerals, as well as diesel soot produced by underground equipment, which may partially block the probe opening and the waistline area shown in Figure 2. Studies (Chandra et al. 1993) at Texas A&M have suggested that a 6-mm-thick buildup on the inner surface of the probe tip could reduce the probe's sampling efficiency for 10-micron particles, expressed as the ratio of the aerosol concentration at the exit of a partially-blocked probe to that of a clean unrestricted probe, by as much as 50%, given the same conditions of mean exhaust-stream airflow and aerosol concentration. These same experiments indicated that blockage of as much as one-third of the waistline had only a minimal effect on sampling efficiency.

WATER INFLOW

The above characterization tests were conducted under dry conditions. As early as April 1993, there were indications that moisture in the exhaust shaft was interfering with the operation of hot-wire flow sensors (MVS 1993). A few months later (Chandra et al. 1993), video inspections of the in-shaft probes revealed a build-up of salt deposits. Subsequently, the probes were pulled on a regular (approximately quarterly) basis for inspection and cleaning and were often found to be wet. By 1998 video inspections of the exhaust shaft liner and the probes clearly showed water flowing into the shaft through several cracks in the liner at about 85 feet below the shaft collar. These inspections showed water droplets entrained in the exhaust airflow being carried upward toward the probes. In early 2000, WTS initiated monthly probe pulls for inspection and cleaning.

The EEG first expressed concern that the presence of water in the exhaust shaft may invalidate effluent air monitoring at Station A in a letter to DOE (Neill 1995) in May 1995 and published that concern in an EEG report (Kenney et al. 1999) in October 1999. The basis of the concern has been that the actual sampling conditions resulting from wet probes and lines at Station A have not been approximated by the test conditions used to characterize the operation of the sampling system and, therefore, the ability of the system to extract a representative sample from the exhaust airstream under all conditions was unproven. Given that concern, it would not have been possible for EEG, based on measurements at Station A alone, to independently assess compliance with NESHAPS, with confidence.

ATMOSPHERIC TRACERS

In September 2003 the EEG began to reexamine archived data from analysis by gamma spectrometry of filter samples from the Station A “skid of record” (A-3) and from a more recently established sampling station, called Station D. Figure 3 shows an idealized schematic of the repository.

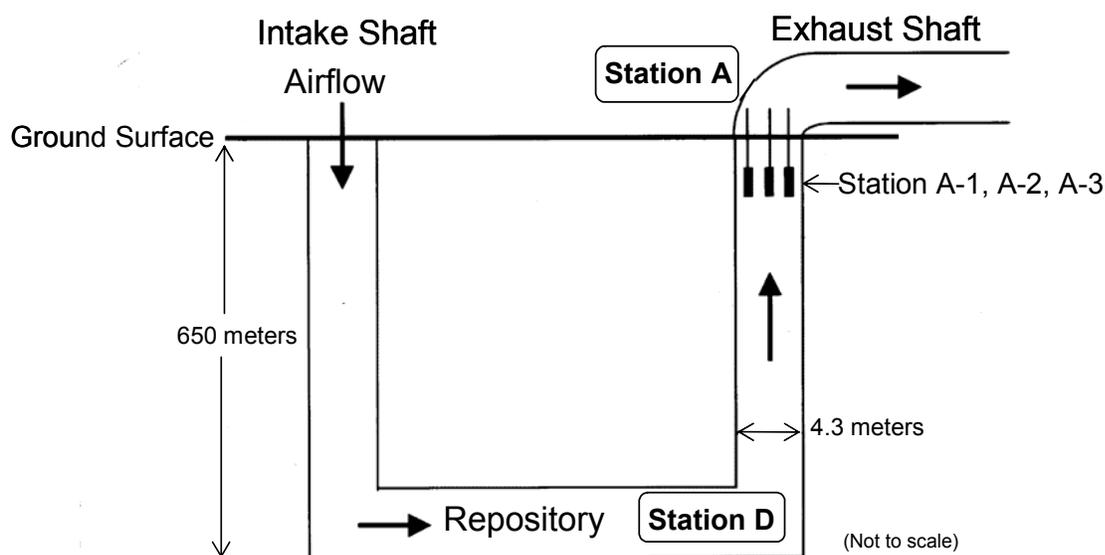


Figure 3. Idealized Schematic of WIPP Repository Airflow

Air flows from the active repository rooms, through the E300 drift, past Station D, and up the exhaust shaft. Hence, the sampling at Station D takes place at the underground level and is not subjected to moisture in the exhaust shaft. The E300 airflow accounts for about 70% of the air discharged from the exhaust shaft during normal operations. Station D conforms to the requirements of ANSI N13.1-1999 (Gadbury 2001) for representative sampling. Furthermore, Station D uses the same sample head design and the same sample media (filters) as those used at Station A.

The purpose of the reexamination was to compare the efficacy of the sampling regime at Skid A-3 with that of the uncompromised sampling regime at Station D by tracking and comparing the measured concentrations of ^7Be and ^{210}Pb at the two sampling locations. Both ^7Be and ^{210}Pb

are naturally-occurring radionuclides. The action of cosmic rays on nitrogen and oxygen in the stratosphere produces ^7Be and once formed it becomes attached to submicron size particles (NCRP 1987). Measurements of ^7Be in samples collected near the surface of the earth are used as an indicator of meteorological processes which cause the atmosphere to “turn over”. The radon daughter ^{210}Pb is produced by decay of natural uranium in the surface of the earth and, in the atmosphere, is associated predominantly with particles of <2 micron diameter (Marley 2000). Both ^7Be and ^{210}Pb are widely used to monitor processes, such as precipitation, which remove aerosol particles from the air (Dibb 1990). The radiological properties of each are shown in Table 1 (Lederer 1978).

Table 1. Radiological Properties of ^7Be and ^{210}Pb

Radionuclide	Half-life	Gamma Energy	Gamma Intensity (Abs)
^7Be	53.4 days	478 keV	10.4%
^{210}Pb	22.3 years	46.5 keV	4.05%

The reexamination included data files resulting from gamma spectrometry of filter samples collected since September 2001. The record has been continuous for Skid A-3, but not for Station D. The filters from both stations were grouped into monthly composites. For samples collected in 2002 and before, in most cases, the Station D composites were not assayed until many months after collection. Because of the relatively short radiological half-life of ^7Be , it was not possible to determine a ^7Be activity in those samples.

During sample collection at both stations, an automatic flow recorder kept a minute-by-minute record of airflow through the sample filters. Integration of these data produced measurements of the total volume of air sampled for each composite. The measured volumes were then used to calculate ^7Be and ^{210}Pb concentrations for each composite in millibecquerels (10^{-3} Bq) per cubic meter (mBq/m^3). The results of these measurements are summarized in Table 2, which also includes a ratio of the concentration measured at Skid A-3 to the concentration measured at Station D.

Table 2. Measured Activity Concentration (mBq/m³) of ⁷Be and ²¹⁰Pb at Skid A-3 and Station D

Coll. Date	Skid A-3 ⁷ Be	Sta. D ⁷ Be	(A/D) ⁷ Be	Skid A-3 ²¹⁰ Pb	Sta. D ²¹⁰ Pb	(A/D) ²¹⁰ Pb
Sep-01	4.40			0.79	0.94	0.840
Oct-01	6.00			1.16	1.16	1.000
Nov-01	4.92			0.94	1.09	0.862
Dec-01	5.63			1.08	0.97	1.113
Jan-02	4.81			1.09	1.08	1.009
Feb-02	3.75			0.89	0.99	0.890
Mar-02	5.38			0.79	0.82	0.963
Apr-02	5.57			0.82	0.72	1.139
May-02	5.41			0.65		
Jun-02	4.72			0.70		
Jul-02	4.20			0.50		
Aug-02	4.66			0.63		
Sep-02	4.62			1.10	0.87	1.264
Oct-02	3.82			0.55		
Nov-02	3.12			0.57		
Dec-02	2.54	2.01	1.264	0.65	0.73	0.890
Jan-03	2.61			0.79		
Feb-03	3.71			0.71		
Mar-03	4.97			0.57		
Apr-03	5.00	5.36	0.933	0.65	0.62	1.048
May-03	5.40	6.45	0.837	0.68	0.72	0.932
Jun-03	5.38	4.95	1.087	0.57	0.49	1.164
Jul-03	4.64	4.03	1.151	0.65	0.61	1.066
Aug-03	3.80	4.28	0.888	0.71	0.73	0.973
Sep-03	3.81	3.44	1.108	0.69	0.66	1.045
		Mean ± 1SD	1.04 ± 0.16		Mean ± 1SD	1.01 ± 0.12

The results of the concentration measurements for ⁷Be and ²¹⁰Pb are presented graphically in Figures 4 and 5 respectively.

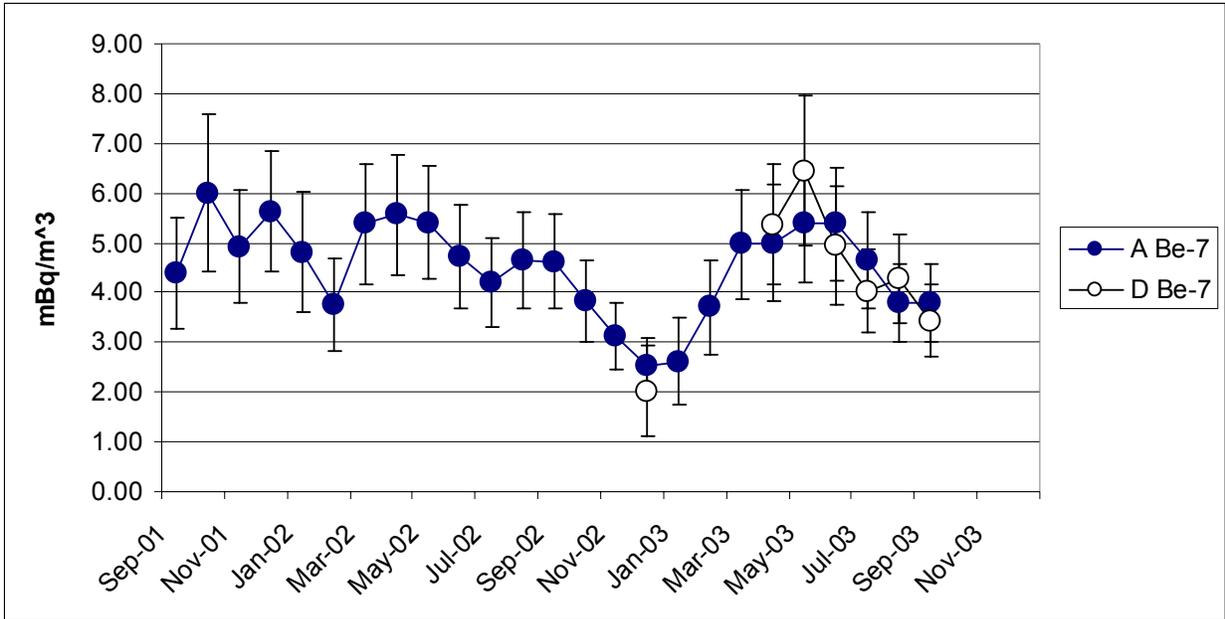


Figure 4. Measured Concentrations of ⁷Be at Skid A-3 and Station D

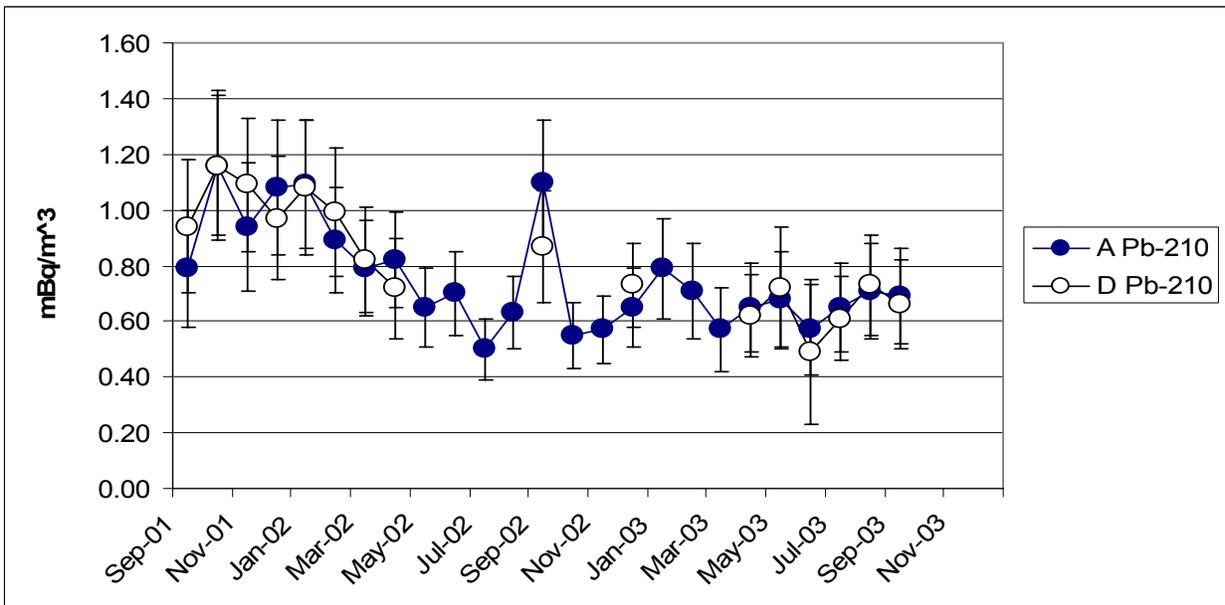


Figure 5. Measured Concentrations of ²¹⁰Pb at Skid A-3 and Station D

The vertical bars in Figures 4 and 5 depict the 95% confidence intervals of the measurements. Statistical examination of these data confirmed the two sampling stations to be highly correlated for each radionuclide, with correlation coefficients (r) of 0.913 ($p = 0.004$) for ⁷Be and 0.866 ($p = 0.000$) for ²¹⁰Pb.

CONCLUSIONS AND RECOMMENDATIONS

It should be noted that the concentrations of ^7Be and ^{210}Pb reported here are similar to those reported by other investigators (NCRP 1987) in surface air. This report assumes that the aerosol is uniformly mixed across the 4.3-meter-diameter exhaust shaft, as required by ANSI N13.1 1999. During the period covered by this report, video inspections of the condition of the sampling probes and of the exhaust shaft liner continued to show evidence of water inflow. Probe pulls during which the sample probes were hoisted out of the exhaust shaft for inspection and cleaning continued to show that the probes were often wet. However, the results reported here show that the aerosol concentrations, using ^7Be and ^{210}Pb as tracers, were approximately the same for the samples of air drawn through the filters at both Skid A-3 and Station D. These results support a conclusion that the presence of liquid water in the sampling system and salting up of the sampling probes during the period from September 2001 to the present had little effect on the collection of <2-micron-diameter aerosol particles at Skid A-3 and, consequently, for that period and for that aerosol size fraction, Skid A-3 measurements should be considered to be representative of the effluent airstream under the assumption of uniform mixing across the exhaust shaft.

Further work is needed to determine whether this conclusion holds for larger aerosol particles (2 to 10 micron diameter). Particles of 10 micron diameter are specifically benchmarked in ANSI N13.1 1999. The EEG (Rodgers 1987) suggested that at least 50% of 10-micron particles should be delivered by the sampling system to the collection media. Because uranium (^{238}U and ^{234}U) in the local environment comes predominantly from soil resuspension which generally produces particles in the larger size ranges (Seinfeld 1975, p89), ^{238}U and ^{234}U concentration measurements at Skid A-3 and Station D should correlate with the concentration of larger-sized particles. Also, the total suspended particulate (TSP) mass is typically bimodal, with one maximum occurring between 0.1 and 1.0 micron and another between 1 and 30 microns (Seinfeld 1975, p89). Rodgers (1987) has stated that particles as large as 100 microns could be carried up the exhaust shaft, even under low flow conditions. Consequently, TSP mass should be significantly influenced by larger particles. Therefore, the EEG will include measurements of

isotopic uranium and TSP mass in the suite of analyses for Station A and Station D samples beginning with the 2003 sampling year to attempt to determine the magnitude of larger particle losses, if any, occurring at Skid A-3.

The EEG will continue to regularly monitor ^7Be and ^{210}Pb in the samples collected at Stations A and D. As long as there continues to be little statistical difference between the concentrations at Stations A and D, we will conclude, with the qualifications stated above regarding uniform mixing and particle size, that the sampling processes at Stations A and D are equally valid. However, continuation of the regular inspection and maintenance programs already implemented by Washington TRU Solutions is needed to support this conclusion.

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