PNNL-15220



# Statistical Analysis of the Critical Mass Laboratory (209-E) Filter Data

T. L. Welsh

June 2005



Prepared for the U.S. Department of Energy under Contract DE-AC05-76RL01830

#### DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor Battelle Memorial Institute, nor any of their employees, makes **any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or Battelle Memorial Institute. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.** 

#### PACIFIC NORTHWEST NATIONAL LABORATORY operated by BATTELLE for the UNITED STATES DEPARTMENT OF ENERGY under Contract DE-AC05-76RL01830

#### Printed in the United States of America

Available to DOE and DOE contractors from the Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831-0062; ph: (865) 576-8401 fax: (865) 576-5728 email: reports@adonis.osti.gov

Available to the public from the National Technical Information Service, U.S. Department of Commerce, 5285 Port Royal Rd., Springfield, VA 22161 ph: (800) 553-6847 fax: (703) 605-6900 email: orders@ntis.fedworld.gov online ordering: http://www.ntis.gov/ordering.htm



PNNL-15220

## Statistical Analysis of the Critical Mass Laboratory (209-E) Filter Data

T. L. Welsh

June 2005

Prepared for the U.S. Department of Energy under Contract DE-AC05-76RL01830

Pacific Northwest National Laboratory Richland, Washington 99352

### **Summary**

The potential-to-emit (PTE) calculation for the Critical Mass Laboratory (209-E) exhaust stack, which was developed around 1992, was questioned by the Washington Department of Health (WDOH). WDOH issued a notice of assurance of discontinuance and a commitment was made in the subsequent reply to develop a new PTE. Samples were obtained from the pre-filters and the first bank of HEPA filters in the 209-E exhaust system and analyzed per the sampling and analysis plan (LeBaron 2005). The sample information from the exhaust pre-filters and the first bank of HEPA filters provided in Table 5 will be used to develop a PTE. If the PTE shows that the stack emissions are greater than 0.1 mrem/year, the stack will be designated as a major stack. If the PTE shows that the stack emissions are less than 0.1 mrem/year, the stack will be designated as a minor stack.

## Contents

Report	1
References	4

## Tables

1	Pre-Filter Sample Results	5
2	HEPA Filter Sample Results	6
3	Mean Concentrations and the Associated Variability	13
4	Pre-filter versus HEPA filter Mean Concentrations	13
5	Final Mean Concentrations and the Associated Variability	14
6	Systematic and Total Measurement Uncertainty Estimates	15

# Figures

Total Alpha Analytical Results	.7
Americium-241 Analytical Results	.7
Total Beta Analytical Results	.8
Plutonium-239/240 Analytical Results	.8
Plutonium/Americium-241 Analytical Results	.9
Plutonium/Americium-242 Analytical Results	.9
Uranium-234 Analytical Results	10
Uranium-235 Analytical Results	10
Uranium-236 Analytical Results	11
Uranium-238 Analytical Results	12
	Total Alpha Analytical Results

## Statistical Analysis of the Critical Mass Laboratory (209-E) Filter Data

#### Summary

The potential-to-emit (PTE) calculation for the Critical Mass Laboratory (209-E) exhaust stack, which was developed around 1992, was questioned by the Washington Department of Health (WDOH). WDOH issued a notice of assurance of discontinuance and a commitment was made in the subsequent reply to develop a new PTE. Samples were obtained from the pre-filters and the first bank of HEPA filters in the 209-E exhaust system and analyzed per the sampling and analysis plan (LeBaron 2005). The sample information from the exhaust pre-filters and the first bank of HEPA filters provided in Table 5 will be used to develop a PTE. If the PTE shows that the stack emissions are greater than 0.1 mrem/year, the stack will be designated as a major stack. If the PTE shows that the stack emissions are less than 0.1 mrem/year, the stack will be designated as a minor stack.

#### Purpose

The purpose of the statistical analysis of the data is to determine the mean concentration for each radionuclide used in determining the PTE.

#### Sampling Design

In order to obtain samples from the two banks of filters (the pre-filters and the first bank of HEPA filters), a sampling design was developed using the methodology outlined in SW-846 (EPA, 2005). The number of samples was determined using Equation 8 from Table 9-1 varying the mean, variability, and the difference from the threshold (see Appendix B in LeBaron 2005). Equation 8 assumes that the data are from a normal distribution. It was determined that ten samples would be obtained from each bank of filters.

Each of the two banks of filters is a two-by-two array consisting of four filters each. The filter media of all four filters is considered the filter face (filter bank) through which the exhaust air flows. An imaginary grid was superimposed over each filter face and a series of consecutive numbers was assigned to each cell of the grid. Ten random numbers for each filter face (each number representing a cell) were selected using a random number generator from SPLUS (Statistical Sciences, 1993). A sample core (a full penetration sample; from front to back) was obtained for each selected cell. All samples were sent to 222-S Analytical Laboratory for dissolution and analysis.

#### Analytical Results

The twenty samples were analyzed according to the sampling and analysis plan (LeBaron 2005) at 222-S Analytical Laboratory. The data were reported in Bushaw 2005. The data are listed in Table 1 for the pre-filter sample results and Table 2 for the HEPA filter sample results. The data are illustrated in Figures 1- 10.

To compare the results for analytes measured using two different analytical methods some of the data were converted from  $\mu$ g/sample to  $\mu$ Ci/sample (or vice versus) using the specific activities reported in DOE-STD-1027-92 (Chow et.al., 1995). As can be seen in Figure 2, the two analytical methods (GEA versus TRU SPEC) for americium-241 provide equivalent results within the total measurement uncertainty. In addition, after summing the plutonium-239 and plutonium-240 results obtained using ICP/MS, the results are equivalent (within the total

measurement uncertainty of the two methods) to the plutonium-239/240 results obtained by the TRU SPEC analytical method (see Figure 4).

#### Statistical Analysis

If the reported analytical results were a mixture of "real" values and "less than" values, then three methodologies were used to determine a mean concentration. The first method did not use the "less than" values in the statistical analysis. The second method uses the upper bound of the "less than" value (e.g., use 6.51E-04 for <6.51E-04). The third method uses half of the upper bound of the "less than" value. All three methods produce (1) a bias (typically positive) of unknown magnitude in the mean concentration and (2) a bias (direction not known) of unknown magnitude in the variance associated with the mean. The method which produced the largest mean concentration (so as not to underestimate the PTE) is reported.

#### Normal Distribution

Two independent tests were performed to test the hypothesis that the random sample comes from a normal distribution with unspecified mean and variance versus the alternative that the distribution is not normal. The two tests are called the Lilliefors Test for Normality and the Shapiro-Wilk Test for Normality (Bowen 1988, chapter 9). The normality tests were performed for each set of filter face analytical results.

The hypothesis that the random sample comes from a normal distribution was not rejected using either test at the 0.05 level of significance for all analytes except for the Total Beta results from the HEPA filter samples. If one result (S05051-20) was deleted from the HEPA filter data set for Total Beta, the hypothesis that the random sample comes from a normal distribution would not be rejected. However, the following two statements (Ostle, 1988) indicate that using the mean concentration for the HEPA filter Total Beta result is reasonable. "That is, unless the population distribution is extremely different from a normal distribution and the sample size is extremely small, the sampling distribution of  $\overline{X}$  will be approximately normal. Therefore we can obtain reasonably good approximation by using normal theory." "However, for many reasonable distributions, (see chapter 4), the Central Limit Theorem will allow us to use a normal approximation with a sample size as small as n=5."

#### Mean Concentration and the Variability Associated with the Mean

For each analyte for each data set (filter bank), the data were statistically evaluated using one-way analysis of variance (ANOVA): the data are identified by one variable (the sample). The one-way ANOVA statistical model used to describe the structure of the data is

$$\begin{split} Y_{ij} &= \mu + S_i + A_{ij}, \\ i &= 1, 2, \dots, a, \quad j {=} 1, 2, \dots, n_i, \end{split}$$

where

$\mathbf{Y}_{ij}$	=	concentration from the j <sup>th</sup> analytical result from the i <sup>th</sup> sample
μ	=	the grand mean
$\mathbf{S}_{i}$	=	the effect of the i <sup>th</sup> sample
A <sub>ij</sub>	=	the effect of the j <sup>th</sup> analytical result from the i <sup>th</sup> sample
a	=	the number of samples (10)
ni	=	the number of analytical results from the $i^{th}$ sample (1 or 2).

The variable  $S_i$  is assumed to be a random effect (the samples were randomly chosen from the filter face). This variable, as well as Aij, are assumed to be normally distributed with means zero and variances  $\sigma^2(S)$  and  $\sigma^2(A)$ , respectively. Estimates of  $\sigma^2(S)$  and  $\sigma^2(A)$  were obtained using Restricted Maximum Likelihood Estimation (REML) techniques. This method applied to variance component estimation is described in Harville, 1977. The results using the REML techniques were obtained using the statistical analysis package S-PLUS (Statistical Sciences, 1993).

The estimator,  $\hat{\mu}$ , is the maximum likelihood estimate of the mean. This estimator was determined by the structure of the data reflected by the statistical model. The estimate of  $\hat{\mu}$  was obtained using REML techniques in S-PLUS. The estimated standard deviation of the mean (where the mean is the maximum likelihood estimate),  $\hat{\sigma}_{\hat{\mu}}$ , is the square root of a function of the variance estimates  $\sigma^2(S)$  and  $\sigma^2(A)$ . Since the data are unbalanced,  $\hat{\sigma}_{\hat{\mu}}$ , is a complicated function of these two variances.

The mean analyte concentration ( $\hat{\mu}$ ) and the relative standard deviation (RSD) associated with

the mean  $(\hat{\sigma}_{\hat{\mu}}/\hat{\mu} \times 100)$  are listed in Table 3 for both the Pre-filter sample results and the HEPA

filter sample results.

The comparisons of the Pre-filter results to the HEPA filter results indicate a fairly constant ratio. The data, the ratio, and the summary statistics for the ratio data are provided in Table 4. Based on these comparisons, the HEPA filter mean concentration for analytes with all data reported as "less than" values, was determined by multiplying the Pre-filter mean concentration by 0.5. This replaces using an upper value from a "less than" value for the mean concentration which produces a positive bias; the mean concentration is overstated.

The final mean concentrations for the Pre-filter sample results and the HEPA filter sample results are listed in Table 5. If the HEPA filter mean concentration was determined from the Pre-filter mean concentration then the uncertainty for this HEPA filter result was calculated using both the variability in the Pre-filter result and the variability in the mean ratio result.

Reference standards and/or spike analyses were performed as specified in the analytical procedures. All reference standards were within statistical process control limits. The reference standards and the spike recoveries provide assurances that the analytical procedures are producing valid results. The reference standard or spike results provide estimates of the "systematic" uncertainty associated with the analytical procedures for the sample matrix. The systematic uncertainties ranged from 3% (1 RSD) to 14%. The total measurement uncertainty for a mean concentration was determined by adding in quadrature the variance associated with the mean (1  $RSD^2$ ) and the systematic variance estimate (1  $RSD^2$ ). The systematic uncertainty along with the total measurement uncertainty are provided for each analyte per filter type in Table 6. Except for cesium-137 and strontium-89/90 the total measurement uncertainty ranges from approximately 5% to approximately 20%.

#### Conclusion

The mean concentrations from which the PTE will be developed are listed in Table 5.

## References

Bowen, W.Michael and Carl A. Bennett (ed.). 1988. *Statistical Methods for Nuclear Material Management*, NUREG/CR-4604.

Bushaw, R.A. 2005. *Final Report for the Critical Mass Laboratory (209-E) Filter Samples Collected in March 2005*, CH2M-0501330. CH2M Hill Hanford Group, Inc, Richland, WA.

Chow, Jim, et.al.. 1995. *Table of DOE-STD-1027-92 Hazard Category 3 Threshold Quantities for the ICRP-30 List of 757 Raionucldes*. Los Alamos National Laboratory, Los Alamos, New Mexico.

Environmental Protection Agency. 2005. *Test Methods for Evaluating Solid Waste, Physical/Chemical Methods* (SW-846).

Harville, D.A. 1977. "Maximum Likelihood Approaches to Variance Component Estimation and to Related Problems," *Journal of the American Statistical Association*, pp. 320-340.

LeBaron, G.J. 2005. *Sample Analysis Plan for the Critical Mass Laboratory (209-E)*, HNF-23373 Revision 0. Fluor Hanford, Richland, WA.

Ostle, Bernard and Linda C. Malone. 1988. *Statistics in Research, Fourth Edition.*, Iowa State University Press, Ames, Iowa.

*S-PLUS Reference Manual*, Version 3.3. 1993. Statistical Sciences, Inc., StatSci, a division of MathSoft, Inc. Seattle, Washington.

Table 1. Pre-Filter Sample Results							
	Alpha Env				Pu239		
					ICP/MS		
Analysis			· · · · · · · · · · · · · · · · · · ·		µg/sample		
1					2.73E-01		
2		1.29E-02			2.70E-01		
1		1.98E-02		4.17E-03	4.29E-01		
1		1.11E-02		2.09E-03	2.51E-01		
1	4.49E-02		1.49E-02	2.93E-03	3.28E-01		
1	4.87E-02	1.59E-02	1.55E-02	3.43E-03	3.60E-01		
1	4.89E-02	1.60E-02	1.54E-02	3.16E-03	3.60E-01		
2	4.65E-02	n/a	n/a	3.20E-03	n/a		
1	1.97E-02	6.45E-03	6.03E-03	1.33E-03	1.41E-01		
1	2.81E-02	9.22E-03	8.49E-03	1.96E-03	1.91E-01		
1	6.40E-02	2.12E-02	2.08E-02	4.50E-03	4.77E-01		
1	6.34E-02	2.02E-02	1.95E-02	4.38E-03	4.55E-01		
	Pu240	Pu244/Cm244	Pu/Am242	Pu/Am241	Pu238		
	ICP/MS	ICP/MS	ICP/MS	ICP/MS	TRU-SPEC		
Analysis	µg/sample	µg/sample	µg/sample	µg/sample	µCi/sample		
1	2.97E-02	1.96E-05	9.70E-04	4.37E-03	2.46E-03		
2	2.91E-02	1.90E-05	9.52E-04	4.33E-03	2.62E-03		
1	4.57E-02	2.27E-05	1.47E-03	6.92E-03	3.65E-03		
1	2.63E-02	1.56E-05	8.27E-04	3.88E-03	<2.39E-03		
1	3.48E-02	2.00E-05	1.12E-03	5.35E-03	3.44E-03		
1	3.91E-02	3.82E-05	1.30E-03	5.80E-03	4.06E-03		
1	3.91E-02	2.32E-05	1.25E-03	5.76E-03	3.84E-03		
2	n/a	n/a	n/a	n/a	n/a		
1	1.49E-02	2.79E-05	4.78E-04	2.30E-03	<1.70E-03		
1	2.06E-02	3.46E-05	6.69E-04	3.20E-03	<2.13E-03		
1	5.10E-02	3.50E-05	1.61E-03	7.54E-03	<5.26E-03		
1	4.90E-02	1.72E-05	1.56E-03	7.41E-03	4.93E-03		
	•			•			
	Pu239/240	U233	U234	U235	U236		
		ICP/MS Acid	ICP/MS	ICP/MS	ICP/MS Acid		
	TRU-SPEC	Add	Acid Add	Acid Add	Add		
Analysis	µCi/sample	µg/sample	µg/sample	µg/sample	µg/sample		
1	2.07E-02	1.91E-05	5.97E-05	1.70E-03	1.35E-04		
2	2.13E-02	2.18E-05	6.35E-05	1.76E-03	1.20E-04		
1	3.23E-02	2.83E-05	1.03E-04	2.95E-03	2.29E-04		
1	2.23E-02	1.74E-05	5.84E-05	1.54E-03	1.11E-04		
1	3.04E-02	2.39E-05	7.96E-05	2.31E-03	1.62E-04		
1	3.41E-02	2.06E-05	8.46E-05	2.08E-03	1.56E-04		
1	3.21E-02	2.46E-05	7.34E-05	2.05E-03	1.60E-04		
2	n/a	n/a	n/a	n/a	n/a		
1	1.41E-02	1.03E-05	4.15E-05	1.35E-03	6.45E-05		
1	1.83E-02	1.39E-05	5.02E-05	1.35E-03	9.93E-05		
1	4.70E-02	3.04E-05	1.21E-04	3.30E-03	2.26E-04		
1	4.25E-02	3.36E-05	1.15E-04	3.58E-03	2.15E-04		
	2 1 1 1 1 1 2 1 1 1 1 1 1 1 1 1 1 1 1 1	Alpha Env Solid/MiscAnalysis $\mu$ Ci/sample13.82E-0223.61E-0215.86E-0213.18E-0214.49E-0214.87E-0214.87E-0214.89E-0224.65E-0211.97E-0212.81E-0216.40E-0216.34E-0212.97E-0222.91E-0213.91E-0213.91E-0213.91E-0213.91E-0215.10E-0214.90E-0213.23E-0213.23E-0213.04E-0213.04E-0213.21E-0213.21E-0213.21E-0213.41E-0213.21E-02	Alpha Env         Am-241           Solid/Misc         TRU-SPEC $\mu$ Ci/sample $\mu$ Ci/sample           1         3.82E-02         1.28E-02           2         3.61E-02         1.29E-02           1         5.86E-02         1.98E-02           1         3.18E-02         1.11E-02           1         4.49E-02         1.55E-02           1         4.87E-02         1.59E-02           1         4.89E-02         1.60E-02           2         4.65E-02         n/a           1         1.97E-02         6.45E-03           1         2.81E-02         9.22E-03           1         6.40E-02         2.12E-02           1         6.40E-02         2.02E-02           1         6.40E-02         2.02E-03           1         6.40E-02         2.02E-02           1         6.34E-02         2.02E-02           1         2.97E-02         1.96E-05           2         2.91E-02         1.90E-05           1         2.63E-02         1.56E-05           1         3.91E-02         2.32E-05           1         3.91E-02         3.23E-05           1 <td><math display="block">\begin{array}{ c c c c c c c c c c c c c c c c c c c</math></td> <td><math display="block"> \begin{array}{ c c c c c c c c c c c c c c c c c c c</math></td>	$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$		

Table 1. Pre-Filter Sample Results

		U238	Cs137	Cs137	
		ICP/MS			
		Acid Add	GEA	GEA	
Customer ID	Analysis	µg/sample	µCi/sample	µCi/sample	
S05051-01	1	1.50E-01	<6.14E-05	<6.14E-05	
S05051-01	2	1.57E-01	<6.24E-05	<6.24E-05	
S05051-02	1	2.46E-01	<6.39E-05	<6.39E-05	
S05051-03	1	1.36E-01	<6.39E-05	<6.39E-05	
S05051-04	1	1.92E-01	<6.51E-05	<6.51E-05	
S05051-05	1	1.71E-01	<6.35E-05	<6.35E-05	
S05051-06	1	1.76E-01	<6.35E-05	<6.35E-05	
S05051-06	2	n/a	n/a	n/a	
S05051-07	1	9.38E-02	<6.39E-05	<6.39E-05	
S05051-08	1	1.20E-01	<6.42E-05	<6.42E-05	
S05051-09	1	2.30E-01	<6.38E-05	<6.38E-05	
S05051-10	1	2.49E-01	<6.47E-05	<6.47E-05	

Table 1. Pre-Filter Sample Results (continued)

 Table 2. HEPA Filter Sample Results

		Alpha Env	Am-241	Am-241	Beta in Env	Pu239
		Solid/Misc	TRU-SPEC	GEA	Samples	ICP/MS
Customer ID	Analysis	µCi/sample		µCi/sample	µCi/sample	µg/sample
S05051-11	1	2.54E-02	7.30E-03	7.99E-03	2.15E-03	1.80E-01
S05051-11	2	2.53E-02	7.98E-03	6.36E-03	2.51E-03	1.86E-01
S05051-12	1	1.68E-02	5.41E-03	5.96E-03	1.49E-03	1.16E-01
S05051-13	1	1.71E-02	5.55E-03	5.44E-03	1.49E-03	1.14E-01
S05051-14	1	2.20E-02	8.07E-03	6.28E-03	1.82E-03	1.80E-01
S05051-15	1	2.46E-02	7.89E-03	8.74E-03	2.06E-03	1.73E-01
S05051-16	1	2.28E-02	7.88E-03	7.86E-03	2.14E-03	1.75E-01
S05051-16	2	2.33E-02	n/a	n/a	2.19E-03	n/a
S05051-17	1	2.16E-02	7.04E-03	7.36E-03	2.19E-03	1.57E-01
S05051-18	1	1.74E-02	5.91E-03	5.52E-03	1.53E-03	1.28E-01
S05051-19	1	2.21E-02	7.90E-03	8.40E-03	1.93E-03	1.70E-01
S05051-20	1	1.85E-02	6.83E-03	6.58E-03	4.48E-03	1.42E-01
		Pu240	Pu244/Cm244	Pu/Am242	Pu/Am241	Pu238
		ICP/MS	ICP/MS	ICP/MS	ICP/MS	TRU-SPEC
Customer ID	Analysis	µg/sample	µg/sample	µg/sample	µg/sample	µCi/sample
S05051-11	1	1.92E-02	<4.50E-05	5.54E-04	2.36E-03	<4.65E-03
S05051-11	2	1.96E-02	<2.50E-05	5.63E-04	2.47E-03	<3.56E-03
S05051-12	1	1.30E-02	<4.50E-05	4.56E-04	1.71E-03	<2.52E-03
S05051-13	1	1.28E-02	<4.50E-05	4.74E-04	1.58E-03	<2.68E-03
S05051-14	1	1.92E-02	<4.50E-05	6.20E-04	2.53E-03	<2.68E-03
S05051-15	1	1.85E-02	<4.50E-05	6.00E-04	2.41E-03	<3.15E-03
S05051-16	1	1.90E-02	<4.50E-05	6.50E-04	2.49E-03	<3.02E-03
S05051-16	2	n/a	n/a	n/a	n/a	n/a
S05051-17	1	1.73E-02	<4.50E-05	5.71E-04	2.26E-03	<4.49E-03
S05051-18	1	1.40E-02	<4.50E-05	4.74E-04	1.74E-03	<2.17E-03
	1	1.045.00	4 505 05	C 10E 04	2 405 02	2 15E 02
S05051-19	1	1.84E-02	<4.50E-05	6.12E-04	2.40E-03	<2.15E-03

$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$		10	$D_{12}220/240$	U233	U234	U235	U236
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$			Fu239/240				
$\begin{array}{c c} \mbox{Customer ID} & \mbox{Analysis} & \mbox{$\mu$Ci/sample} & \mbox{$\mu$g/sample} & \m$			TDU SDEC				
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	Customer ID	Amolycic					
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$		Analysis					
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $		1					
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$							
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$							
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $		-					
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $		_					
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $							
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	S05051-16		1.54E-02	<4.50e-05	5.45E-05	1.29E-03	7.10E-05
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	S05051-16						
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	S05051-17	1	1.40E-02	<4.50e-05	4.95E-05	1.60E-03	6.20E-05
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	S05051-18	1	1.14E-02	<4.50e-05	2.70E-05	1.16E-03	6.15E-05
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	S05051-19	1	1.46E-02	<4.50e-05	3.70E-05	1.44E-03	7.45E-05
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	S05051-20	1	1.30E-02	<4.50e-05	5.30E-05	1.39E-03	6.75E-05
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$		-					•
$\begin{array}{c c c c c c c c c c c c c c c c c c c $			U238	Cs137	Sr89/90		
$\begin{array}{c c c c c c c c c c c c c c c c c c c $			ICP/MS				
S05051-11         1         1.04E-01         <3.95E-04         <4.93E-05           S05051-11         2         1.07E-01         <3.56E-04			Acid Add	GEA	Env. Misc		
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Customer ID	Analysis	µg/sample	µCi/sample	µCi/sample		
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	S05051-11	1	1.04E-01	<3.95E-04	<4.93E-05		
S05051-13         1         8.30E-02         <3.87E-04         <5.07E-05           S05051-14         1         1.30E-01         <3.72E-04	S05051-11	2	1.07E-01	<3.56E-04	<3.18E-05		
S05051-14         1         1.30E-01         <3.72E-04         <5.50E-05           S05051-15         1         1.22E-01         <3.87Ee-04	S05051-12	1	8.68E-02	<3.71E-04	<3.20E-05		
S05051-15         1         1.22E-01         <3.87Ee-04         <3.77E-05           S05051-16         1         1.08E-01         <3.44E-04	S05051-13	1	8.30E-02	<3.87E-04	<5.07E-05		
S05051-16         1         1.08E-01         <3.44E-04         <3.32E-05           S05051-16         2         n/a         n/a         n/a           S05051-17         1         1.52E-01         <3.60E-04	S05051-14	1	1.30E-01	<3.72E-04	<5.50E-05		
S05051-16         2         n/a         n/a         n/a           S05051-17         1         1.52E-01         <3.60E-04	S05051-15	1	1.22E-01	<3.87Ee-04	<3.77E-05		
S05051-17         1         1.52E-01         <3.60E-04         <3.11E-05           S05051-18         1         1.03E-01         <3.40E-04	S05051-16	1	1.08E-01	<3.44E-04	<3.32E-05		
S05051-18         1         1.03E-01         <3.40E-04         <3.29E-05           S05051-19         1         1.14E-01         <3.87E-04	S05051-16	2	n/a	n/a	n/a		
S05051-18         1         1.03E-01         <3.40E-04         <3.29E-05           S05051-19         1         1.14E-01         <3.87E-04	S05051-17	1	1.52E-01	<3.60E-04	<3.11E-05		
		1					
	S05051-19	1	1.14E-01	<3.87E-04	<4.67E-05		
		1					

Table 2. HEPA Filter Sample Results (continued)

Alpha Env Solid/Misc (each)







Am241

Figure 2. Americium 241 Analytical Results









Pu239/240

Figure 4. Plutonium-239/240 Analytical Results

Pu/Am241 ICP/MS



Figure 5. Plutonium/Americium-241 Analytical Results



Pu/Am242 ICP/MS

Figure 6. Plutonium/Americium-242 Analytical Results

Uranium-234 ICP/MS Acid Add







Uranium-235 ICP/MS Acid Add

Figure 8. Uranium-235 Analytical Results

Uranium-236 ICP/MS Acid Add







Uranium-238 ICP/MS Acid Add

Figure 10. Uranium-238 Analytical Results

	loncentration			Random	Uncertainty associated
		Mean Cor	ncentration		mean (%)
Analyte	Units	Pre-Filter	HEPA Filter	Pre-Filter	HEPA Filter
Alpha Env: Solid/Misc (Each)	µCi/sample	4.44E-02	2.09E-02	10.8	4.8
Am-241 by TRU-SPEC Resin IonEx	µCi/sample	1.48E-02	7.01E-03	10.5	4.7
Americium-241 by GEA	µCi/sample	1.42E-02	6.93E-03	10.9	5.3
Beta in Env. Samples (Each)	µCi/sample	3.05E-03	2.15E-03	11.3	12.9
Cesium-137 by GEA	µCi/sample	<6.51E-05 <sup>a</sup>	<3.95E-04 <sup>a</sup>	NA	NA
Plutonium-239 by ICP/MS	µg/sample	3.26E-01	1.54E-01	10.9	5.5
Plutonium-240 by ICP/MS	µg/sample	3.50E-02	1.67E-02	10.9	5.0
Pu 244/Cm 244 by ICP/MS	µg/sample	2.54E-05	<4.50E-05 <sup>a</sup>	10.1	NA
Pu/Am 242 ICP/MS	µg/sample	1.12E-03	5.54E-04	10.8	3.9
Pu/Am-241 by ICP/MS	µg/sample	5.25E-03	2.15E-03	10.8	5.4
Pu-238 by TRU-SPEC Resin IonEx	µCi/sample	3.74E-03 <sup>b</sup>	<4.65E-03	8.5	NA
Pu-239/240 by TRU-SPEC Resin	µCi/sample	2.94E-02	1.37E-02	11.3	5.1
Pu239+Pu240 by ICP/MS by addition	µCi/sample	2.83E-02	1.34E-02	10.9	5.3
Sr-89/90 Env. Misc. (ea.)	µCi/sample	1.32E-04 °	<5.50E-05 <sup>a</sup>	45.0	NA
Uranium-233 by ICP/MS Acid Add	µg/sample	2.24E-05	<4.50E-05 <sup>a</sup>	10.4	NA
Uranium-234 by ICP/MS Acid Add	µg/sample	7.88E-05	3.92E-05	10.9	8.7
Uranium-235 by ICP/MS Acid Add	µg/sample	2.22E-03	1.34E-03	11.4	5.3
Uranium-236 by ICP/MS Acid Add	µg/sample	1.55E-04	6.60E-05	11.4	3.2
Uranium-238 by ICP/MS Acid Add	µg/sample	1.77E-01	1.16E-01	9.5	6.5

Table 3. Mean Concentrations and the Associated Variability

the largest "less than" value a

b

"less than" values deleted from the statistical analysis only result that was not a "less than" value; RSD is from the counting statistics с

not available with the stated data NA

Table 4. Fre-inter versus filler A finter ivitian Concentrations						
		Mean Cond	centration	HEPA m	ean/ Pre-filter mean	
Analyte	Units	Pre-Filter	HEPA Filter	All Data	Exclude Total Beta	
Alpha Env: Solid/Misc (Each)	µCi/sample	4.44E-02	2.09E-02	0.470	0.470	
Am-241 by TRU-SPEC Resin IonEx	µCi/sample	1.48E-02	7.01E-03	0.473	0.473	
Americium-241 by GEA	µCi/sample	1.42E-02	6.93E-03	0.489	0.489	
Beta in Env. Samples (Each)	µCi/sample	3.05E-03	2.15E-03	0.705		
Plutonium-239 by ICP/MS	µg/sample	3.26E-01	1.54E-01	0.471	0.471	
Plutonium-240 by ICP/MS	µg/sample	3.50E-02	1.67E-02	0.478	0.478	
Pu/Am 242 ICP/MS	µg/sample	1.12E-03	5.54E-04	0.493	0.493	
Pu/Am-241 by ICP/MS	µg/sample	5.25E-03	2.15E-03	0.409	0.409	
Pu-239/240 by TRU-SPEC Resin	µCi/sample	2.94E-02	1.37E-02	0.467	0.467	
Pu239+Pu240 by ICP/MS by addition	µCi/sample	2.83E-02	1.34E-02	0.473	0.473	
Uranium-234 by ICP/MS Acid Add	ug/sample	7.88E-05	3.92E-05	0.497	0.497	
Uranium-235 by ICP/MS Acid Add	ug/sample	2.22E-03	1.34E-03	0.603	0.603	
Uranium-236 by ICP/MS Acid Add	ug/sample	1.55E-04	6.60E-05	0.426	0.426	
Uranium-238 by ICP/MS Acid Add	ug/sample	1.77E-01	1.16E-01	0.654	0.654	
Mean				0.508	0.493	
Standard Deviation				0.085	0.066	
Standard Deviation of the Mean				0.023	0.018	

Table 4. Pre-filter versus HEPA filter Mean Concentrations

		Random Uncertainty				
				RSD associated		
		Mean Conc	entration	with the	mean (%)	
Analyte	Units	Pre-Filter	HEPA Filter	Pre-Filter	HEPA Filter	
Alpha Env: Solid/Misc (Each)	µCi/sample	4.44E-02	2.09E-02	10.8	4.8	
Am-241 by TRU-SPEC Resin IonEx	µCi/sample	1.48E-02	7.01E-03	10.5	4.7	
Americium-241 by GEA	µCi/sample	1.42E-02	6.93E-03	10.9	5.3	
Beta in Env. Samples (Each)	µCi/sample	3.05E-03	2.15E-03	11.3	12.9	
Cesium-137 by GEA	µCi/sample	6.51E-05 <sup>a</sup>	3.26E-05 <sup>b</sup>	50.0 °	50.2	
Plutonium-239 by ICP/MS	µg/sample	3.26E-01	1.54E-01	10.9	5.5	
Plutonium-240 by ICP/MS	µg/sample	3.50E-02	1.67E-02	10.9	5.0	
Pu 244/Cm 244 by ICP/MS	µg/sample	2.54E-05	1.27E-05 <sup>b</sup>	10.1	11.0	
Pu/Am 242 ICP/MS	µg/sample	1.12E-03	5.54E-04	10.8	3.9	
Pu/Am-241 by ICP/MS	µg/sample	5.25E-03	2.15E-03	10.8	5.4	
Pu-238 by TRU-SPEC Resin IonEx	µCi/sample	3.74E-03 <sup>d</sup>	1.87E-03 <sup>b</sup>	8.5	9.6	
Pu-239/240 by TRU-SPEC Resin	µCi/sample	2.94E-02	1.37E-02	11.3	5.1	
Pu239+Pu240 by ICP/MS by addition	µCi/sample	2.83E-02	1.34E-02	10.9	5.3	
Sr-89/90 Env. Misc. (ea.)	µCi/sample	1.32E-04 <sup>e</sup>	6.60E-05 <sup>b</sup>	45.0	45.2	
Uranium-233 by ICP/MS Acid Add	µg/sample	2.24E-05	1.12E-05 <sup>b</sup>	10.4	11.3	
Uranium-234 by ICP/MS Acid Add	µg/sample	7.88E-05	3.92E-05	10.9	8.7	
Uranium-235 by ICP/MS Acid Add	µg/sample	2.22E-03	1.34E-03	11.4	5.3	
Uranium-236 by ICP/MS Acid Add	µg/sample	1.55E-04	6.60E-05	11.4	3.2	
Uranium-238 by ICP/MS Acid Add	µg/sample	1.77E-01	1.16E-01	9.5	6.5	

Table 5. Final Mean Concentrations and the Associated Variability

a the upper value of the largest "less than" (e.g., 6.51E-05 for <6.51E-05)

b assumes 50% of the Pre-filter result

c assumption; the less than implies that the result could be anywhere below the stated number. using 50% as 1 RSD and 100% as 2 RSD allows the result to be as low as zero

d "less than" values were deleted from the statistical analysis (6 samples instead of 10)

e only result that was not a "less than" value; RSD is from the counting statistics

	Systematic	Total Measurement Uncertainty associated with the mean (%)			
Analyte	Uncertainty	Pre-Filter	HEPA Filter		
Alpha Env: Solid/Misc (Each)	8	13.4	9.3		
Am-241 by TRU-SPEC Resin IonEx	9	13.8	10.2		
Americium-241 by GEA	8	13.5	9.6		
Beta in Env. Samples (Each)	9	14.4	15.7		
Cesium-137 by GEA	8	50.6 <sup>a</sup>	50.8		
Plutonium-239 by ICP/MS	3	11.3	6.3		
Plutonium-240 by ICP/MS	3	11.3	5.8		
Pu 244/Cm 244 by ICP/MS	3	10.5	11.4		
Pu/Am 242 ICP/MS	3	11.2	4.9		
Pu/Am-241 by ICP/MS	9	14.0	10.5		
Pu-238 by TRU-SPEC Resin IonEx	7	11.0	11.9		
Pu-239/240 by TRU-SPEC Resin	7	13.3	8.7		
Pu239+Pu240 by ICP/MS by addition	3	11.3	6.1		
Sr-89/90 Env. Misc. (ea.)	5	45.3	45.5		
Uranium-233 by ICP/MS Acid Add	14	17.4	18.0		
Uranium-234 by ICP/MS Acid Add	14	17.8	16.5		
Uranium-235 by ICP/MS Acid Add	14	18.1	15.0		
Uranium-236 by ICP/MS Acid Add	14	18.1	14.4		
Uranium-238 by ICP/MS Acid Add	8	12.4	10.3		

Table 6. Systematic and Total Measurement Uncertainty Estimates

а assumes a random uncertainty of 50%

"less than" values were deleted from the statistical analysis (6 samples instead of 10) only result that was not a "less than" value; RSD is from the counting statistics b

с

## Distribution

#### No. of <u>Copies</u>

### OFFSITE

### ONSITE

No. of

**Copies** 

### 3 Fluor Hanford

D.L. Dyekman	H8-13
G. LeBaron	S2-42
M. Wright	S2-42

### 2 Pacific Northwest National Laboratory

B.A. Pulsipher	K6-08
TL Welsh	K6-08