

Analysis of EPA and DOE WIPP Air Sampling Data

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1.0 Summary

During the U.S. Environmental Protection Agency's (EPA) April 2014 visit to the Waste Isolation Pilot Plant (WIPP), EPA co-located four ambient air samplers with existing Department of Energy (DOE) ambient air samplers to independently corroborate DOE's reported air sampling results. The air samplers were located at the WIPP Far Field, WIPP East and WIPP South locations. Three sets of samples were run for week-long time in April 2014. The air filters were analyzed for americium-241 (Am-241), plutonium-238 (Pu-238) and plutonium-239/240 (Pu-239/240) with the primary purpose of determining whether DOE and EPA data were comparable.

Data for each radionuclide were fit separately using an analysis of variance model that accounted for potential effects on measured activity concentration of monitor location, time period and data source (DOE or EPA). Results provided no evidence of a systematic difference between the DOE and EPA activity concentration measurements, although it should be noted that the statistical power for detecting such a difference is limited. For both DOE and EPA data, the variation observed among the activity concentration measurements was consistent with the combined standard uncertainty estimates (CSU) that had been included with the data. The data provided no evidence of positive concentrations for any of the monitored radionuclides.

2.0 Monitoring Data

Both DOE and EPA provided spreadsheets with common data fields; these included sampling location, time period sampled, activity in disintegrations per minute (dpm), air volume of the sample collected (m^3), the derived measured activity concentration (Bq m^{-3}), an estimate of the activity concentration CSU, and a "Z-value" for determining whether a sample measurement is a non-detect. The reported Z-value is equal to the measured activity concentration divided by the CSU. The CSU is a measure of uncertainty, essentially an estimate of the standard deviation, which accounts for "error" in measurements associated with identified sources of uncertainty (e.g., random variation in the number of disintegrations within the monitoring period, uncertainty about the counting efficiency of the instrument, and uncertainty associated with corrections necessary to subtract out effects from sources of radiation other than the analyte). For many types of data (e.g., for which random variation follows a normal distribution), about 5% of measurements can be expected to differ from the value being measured by at least 2 times the CSU. It follows that a Z-value greater than 2 should not be an especially rare occurrence – even if the concentrations being measured all equal 0. A measurement with a Z-value less than 2 is typically regarded as being too small—relative to its CSU—to establish the presence of the radionuclide. A listing of selected data fields is given in the Attachment I.

The ambient air monitoring data covers three, week-long time periods in April. Both DOE and EPA collected four samples each week; each organization collected two samples at the Far Field location and one each at WIPP East and WIPP South. A measurement from a blank sample was used to account for the effects of background radiation, that is to say, measurements of activity from field samples were "blank corrected" by subtracting out results obtained from the blanks. The EPA spreadsheet included the original results from the blank samples whereas the DOE spreadsheet only included the blank-corrected results. EPA used a common blank sample for each of the four samples collected each week, and a reasonable assumption—based on conversations with DOE—is that the same practice was used at DOE, that is to say, each week a common blank sample was used for each of the four samples. Thus, for both datasets, blank-corrected results from samples collected within the same time period are correlated.

3.0 Methodology

3.1 Descriptive Statistics

Descriptive statistics were first calculated for the main variables of interest: activity concentration, and the corresponding CSU estimate and Z- values. Pairwise comparisons were also made between the blank-adjusted activity measurements from DOE and EPA. For each time period and location, the normalized difference between the average EPA activity measurement and average DOE measurement was calculated according to the equation:

$$Z_d = \frac{\text{mean}(Y_{DOE}) - \text{mean}(Y_{EPA})}{\sqrt{k(\text{mean}(CSU_{DOE}^2) + \text{mean}(CSU_{EPA}^2))}}$$

$k = 1$ for WIPP East or WIPP South and $k = 0.75$ for Far Field.

For a pairwise comparison, an absolute value of Z_d greater than 2 would indicate a difference between the DOE and EPA measurements that cannot be attributed to random error inherent in the sampling and laboratory measurement process. Note that if there is no such difference, then about 2.5% of the Z_d values would be expected to be greater than 2 and about 2.5% less than -2. That is, 5% would be false positives. This assumed false positive rate of 5% is only approximate, in part, because calculated values for Z_d often depend on imprecise estimates of the CSU.

3.2 Main Analysis

For the main analysis, an Analysis of Variance (ANOVA) model – with modification to account for correlations among results for samples collected within the same time period – was used to test the (null) hypotheses that: 1) there is no systematic difference between the DOE and EPA activity concentration measurements, and 2) activity concentration at all monitoring stations for all time periods are equal to 0. For each radionuclide, blank corrected activity concentration measurements were assumed to depend on data source ($i = 1, 2$: DOE or EPA), time period sampled ($j = 1, 2, \text{ or } 3$), sampling location ($k = 1, 2, \text{ or } 3$ for Far Field, WIPP East or WIPP South), and replicate number ($m = 1 \text{ or } 2$ for Far Field; 1 for other locations) according to the equation:

$$Y_{ijk(m)} = \mu + \alpha_i + \beta_j + \gamma_{ij} + \lambda_k + \delta_{ij} + \epsilon_{ijk(m)},$$

$$\delta_{ij}, \epsilon_{ijk(m)} \sim N(0, \sigma^2)$$

Here, μ = mean activity concentration (averaged over all locations and time periods). The parameters α_i (data source), β_j (time period sampled) and λ_k (location) are main effect terms and the γ_{ij} are interaction terms for data source and time period. Note that the variance of a blank-corrected measurement is $2\sigma^2$.

The software package, Stata, was used to calculate the ANOVA statistics and simple adjustments were applied to F-value statistics (for testing hypotheses at the 0.05 significance level), to account for the correlations in the blank-corrected measurements. An advantage of using this approach is that—under the null hypotheses—the mean squared error (MSE) value generated from the standard ANOVA is an

unbiased estimate of σ^2 . The MSE (divided by 2) was then compared to the squared CSU estimates (included in the dataset).

Essentially, the ANOVA compares the amount of observed variation in the activity concentration measurements associated with factors such as data source, monitor location and sampling period to an estimate of the amount of variation that would be expected if these factors would have no effect on the measurements. The F-statistic (with adjustment), is the ratio of the observed variation associated with the factors being tested to an estimate of variation that would be expected if the (tested) factors have no effect on the measurements. A ratio that is sufficiently large indicates that the amount of variation may be too large to be attributed to chance alone, i.e., the results are statistically significant. Statistically significant results are typically interpreted as evidence that the factors being tested have a noticeable effect on measurement results.

4.0 Results

4.1 Descriptive Statistics

Table 1 summarizes the 72 activity concentration measurements for Am-241, Pu-238, and Pu-239/240 (24 measurements each). Measurements (Bq m^{-3}) ranged from (-4.14E-7 to 6.22E-07) for Am-241, (-4.32E-7 to 2.47E-07) for Pu-238 and (-3.35 E-07 to 2.41 E-07) for Pu-239/240. Average measurements were positive for Am-241 and negative for Pu-238. For Pu-239/240 the average was negative for data from EPA. The mean DOE measurement was greater than the corresponding mean from EPA for Am-241 and Pu-239/240, but smaller for Pu-238. For all radionuclides tested, the absolute value of the mean was smaller than the standard deviation.

Table 1: Summary of derived activity concentration measurements (Bq m^{-3})

Radionuclide	Data Source	Mean	Standard deviation	Minimum ¹	Maximum ¹
Am-241	DOE (n=12)	1.29E-07	1.99E-07	-1.08E-07 (1.91E-07)	6.22E-07 (4.58E-07)
	EPA (n=12)	2.20E-08	2.47E-07	-4.14E-07 (2.47E-07)	3.68E-07 (3.44E-07)
	Combined	7.53E-08	2.26E-07	-4.14E-07	6.22E-07
Pu-238	DOE (n=12)	-3.88E-08	1.10E-07	-1.80E-07 (9.01E-08)	2.47E-07 (2.54E-07)
	EPA (n=12)	-2.69E-08	2.24E-07	-4.32E-07 (2.20E-07)	2.26E-07 (3.42E-07)
	Combined	-3.29E-08	1.73E-07	-4.32E-07	2.47E-07
Pu-239/240	DOE (n=12)	9.75E-09	1.13E-07	-1.51E-07 (6.88E-08)	2.00E-07 (2.31E-07)
	EPA (n=12)	-1.01E-07	1.99E-07	-3.35E-07 (2.08E-07)	2.41E-07 (2.80E-07)
	Combined	-4.54E-08	1.68E-07	-3.35E-07	2.41E-07

¹ Minimum and maximum reported Combined Standard Uncertainties given in parentheses.

As seen in Table 2, Z-values for the activity measurements are all smaller than 2. Thus, all samples were designated “non-detects.” It can be easily shown that if the CSU can be accurately determined, the standard deviation of Z-values should be about 1, i.e., a measurement might typically deviate from its measurand by an amount approximately equal to 1 standard deviation. However, for all radionuclides the standard deviations of the Z-values were less than 1. This could be an indication of a bias in the CSU estimates (the CSU estimates having a tendency to be too large), although there are alternate explanations. Z-values associated with paired-comparisons (not shown) were also all less than 2.

Table 2: Summary of Z-values for activity measurements from EPA and DOE

Radionuclide	Data Source	Mean	Standard deviation	Minimum	Maximum
Am-241	DOE*	0.38	0.63	-0.53	1.89
	EPA	0.09	0.81	-1.31	1.26
	Combined	0.24	0.72	-1.31	1.89
Pu-238	DOE*	-0.35	0.58	-1.03	0.97
	EPA	-0.09	0.84	-1.73	0.84
	Combined	-0.22	0.72	-1.73	0.97
Pu-239/240	DOE*	-0.13	0.83	-1.43	0.87
	EPA	-0.47	0.86	-1.47	0.97
	Combined	-0.30	0.84	-1.47	0.97

*Z- values were incorrectly reported by DOE. Spreadsheet values were multiplied by a factor of 2.

4.2 Main Analysis

The fitting of the data to the ANOVA model allowed for a more thorough examination as to whether: 1) there is a systematic difference between DOE and EPA measurements, and 2) there are positive activity concentrations for any of the locations or time periods sampled. Results for the three (sets of) radionuclides are summarized in Tables 3 to 5. The tables include F-statistics with corresponding p-values (statistically significant results are those for which the p-value < 0.05). F-statistics are the ratios of the observed variation associated with tested factors to the estimated expected variation if the factors had no effect on the activity concentration measurements. More specifically, F-statistics are given which were used to determine:

- 1) The statistical significance of variation associated with data source, i.e., whether the data is from DOE or EPA. (A sufficiently large value for this F-statistic would indicate that the difference between DOE and EPA average measurements could not be just a consequence of random variation inherent in the measurement process).
- 2) The statistical significance of variation associated with any of the factors included in the model, (i.e., variation associated with data source, sampling location or time period).
- 3) Whether average measured activity concentrations were significantly > 0 .

There were no significant results. For each radionuclide

- 1) There was no evidence of a systematic difference between DOE and EPA measurements (F-statistic associated with Data Source is not significant).
- 2) There was insufficient evidence to conclude that activity concentrations depended on data source, time period or sampling location (F-statistic associated with the model is not significant).
- 3) There was insufficient evidence (Am-241) or no evidence (Pu-238, Pu-239/240) that activity concentrations for any of the locations or time periods sampled was > 0 .

Finally, Table 6 compares the MSE values (Tables 3 to 5) to the CSU estimates calculated by DOE and EPA. If underlying assumptions for the ANOVA model and CSU calculations are correct, the MSE would be (on average) about half as large as the average of the squared CSU value. Given the small sample sizes (and the variation in data used to calculate the MSE and the CSU), results shown in Table 6 are consistent (ratios in the last column are sufficiently close to 1).

Table 3: Summary of ANOVA results for Am-241 activity concentration measurements (Bq m⁻³)

Source	Hypothesis test question	Df ¹	Mean Square	F -Statistic ²
All in Model	Do activity concentrations depend on data source, time period sampled or location?	7	9.92E-14	1.32 (p = 0.3)
Error		16	3.03E-14	
Data Source (DOE vs. EPA)	Is there a systematic difference between measurements from DOE vs. EPA?	1	6.82E-14	0.90 ³ (p = 0.36)
Constant term	Are activity concentrations for any of the locations or time periods sampled > 0? ⁴	1	1.36E-13	1.79 ³ (p = 0.1)

¹ Degrees of freedom

² Adjusted to account for correlations induced from the use of common blank samples.

³ F-statistic from Stata calculations divided by 2.5.

⁴ Assumes no overall bias in measurements from DOE and EPA.

Table 4: Summary of ANOVA results for Pu-238 activity concentration measurements (Bq m⁻³)

Source	Hypothesis test question	Df	Mean Square	F-Statistic ¹
All in Model	Do activity concentrations depend on data source, time period sampled or location?	7	2.96E-14	0.43 p>0.5
Error		16	3.00E-14	
Data Source (DOE vs. EPA)	Is there a systematic difference between measurements from DOE vs. EPA?	1	9.0 E-16	0.01 ² p>0.5
Constant term	Are activity concentrations for any of the locations or time periods sampled > 0? ³	1	2.58E-14	0.34 ² (mean < 0) p>0.5

¹ Adjusted to account for correlations induced from the use of common blank samples.

² F-statistic from Stata calculations divided by 2.5

³ Assumes no overall bias in measurements from DOE and EPA.

Table 5: Summary of ANOVA results, Pu-239/240 activity concentration measurements (Bq m^{-3})

Source	Hypothesis test question		Df	Mean Square	F-Statistic ¹
All in Model	Do activity concentrations depend on data source, time period sampled or location?		7	6.06E-14	1.80 (p=0.16)
Error			16	1.38E-14	
Data Source (DOE vs. EPA)	Is there a systematic difference between measurements from DOE vs. EPA?		1	7.28E-14	2.11 ² (p=0.17)
Constant term	Are activity concentrations for any of the locations or time periods sampled > 0 ? ³		1	4.95E-14	1.43 ² (mean < 0) (p > 0.5)

¹ Adjusted to account for correlations induced from the use of common blank samples.

² F-statistic from Stata calculations divided by 2.5.

³ Assumes no overall bias in measurements from DOE and EPA.

Table 6: Comparison of MSE values from ANOVA to estimates of CSU

Radionuclide	MSE	(Mean Square CSU)/2	Ratio
Am-241	3.03E-14	4.38E-14	0.69
Pu-238	3.00E-14	2.71E-14	1.11
Pu-239/240	1.38E-14	2.00E-14	0.69

5.0 Limitations of the Analysis

The data provided no evidence of a systematic difference between the DOE and EPA measurements. Although this is an indication that systematic differences between DOE and EPA measurements may be negligible, it should be noted that the statistical power for detecting such differences is limited. The dataset is small (for each radionuclide, measurements from DOE and EPA were each based on only 12 field samples and 3 blank samples), and average differences in measurement less than the CSUs for individual blank-corrected activity concentration measurements were not detectable. A potentially more important issue relates to the possibility that the magnitude of systematic differences, if they exist, might be expected to increase with the level of concentrations being measured. However, this issue could not be investigated, since the data were consistent with zero concentrations for each of the radionuclides.

Table 7: Table of Units

Unit	Abbreviation	Quantity/Description
Becquerel	Bq	International System of Units (SI unit) of radioactivity = 1 disintegration per second
Disintegrations per minute	dpm	Radioactivity
Becquerel per meter cubed	Bq m^{-3} or Bq/m^3	Activity concentration
Meter cubed	m^3	Air volume of the sample collected

Attachment I: Data Listings**Table I.1: DOE Supplied Data for Selected Variables1****Am-241**

Sample	Location	Sampling Period	Blank-	Volume	Activity	CSU	Z-value
			corrected		Concentration		
			Activity	m ³	(Bq m ⁻³)	(Bq m ⁻³)	
1	WFF	4/8-4/15	1.20E-02	571.20	3.50E-07	4.11E-07	0.85
2	WFF	4/8-4/15	1.84E-04	574.06	5.34E-09	3.32E-07	0.02
3	WEE	4/8-4/15	5.53E-03	568.60	1.62E-07	3.11E-07	0.52
4	WSS	4/8-4/15	3.71E-03	570.74	1.08E-07	4.58E-07	0.24
5	WFF	4/15-4/22	1.89E-04	551.76	5.71E-09	2.10E-07	0.03
6	WFF	4/15-4/22	1.57E-03	555.76	4.71E-08	2.44E-07	0.19
7	WEE	4/15-4/22	-3.60E-03	557.36	-1.08E-07	2.05E-07	-0.53
8	WSS	4/15-4/22	4.40E-03	543.32	1.35E-07	2.64E-07	0.51
9	WFF	4/22-4/29	2.10E-02	562.80	6.22E-07	3.29E-07	1.89
10	WFF	4/22-4/29	5.12E-04	577.01	1.48E-08	1.91E-07	0.08
11	WEE	4/22-4/29	-1.13E-03	568.83	-3.31E-08	2.17E-07	-0.15
12	WSS	4/22-4/29	8.14E-03	579.86	2.34E-07	2.51E-07	0.93

Pu-238

1	WFF	4/8-4/15	8.47E-03	571.20	2.47E-07	2.54E-07	0.97
2	WFF	4/8-4/15	-3.57E-04	574.06	-1.04E-08	1.70E-07	-0.06
3	WEE	4/8-4/15	-1.88E-03	568.60	-5.51E-08	9.01E-08	-0.61
4	WSS	4/8-4/15	-2.21E-03	570.74	-6.45E-08	9.93E-08	-0.65
5	WFF	4/15-4/22	-4.27E-03	551.76	-1.29E-07	1.47E-07	-0.88
6	WFF	4/15-4/22	-6.01E-03	555.76	-1.80E-07	1.75E-07	-1.03
7	WEE	4/15-4/22	-7.68E-04	557.36	-2.30E-08	2.30E-07	-0.10
8	WSS	4/15-4/22	-4.53E-03	543.32	-1.39E-07	1.55E-07	-0.90
9	WFF	4/22-4/29	-8.83E-04	562.80	-2.61E-08	1.70E-07	-0.15
10	WFF	4/22-4/29	1.84E-03	577.01	5.31E-08	2.05E-07	0.26
11	WEE	4/22-4/29	-2.93E-03	568.83	-8.58E-08	1.15E-07	-0.75
12	WSS	4/22-4/29	-1.83E-03	579.86	-5.26E-08	1.75E-07	-0.30

Pu-239/240

Sample	Location	Sampling Period	Blank-corrected Activity (dpm)	Volume m³	Activity Concentration (Bq m⁻³)	CSU (Bq m⁻³)	Z-value
1	WFF	4/8-4/15	2.76E-03	571.20	8.05E-08	1.95E-07	0.41
2	WFF	4/8-4/15	-5.20E-03	574.06	-1.51E-07	1.57E-07	-0.96
3	WEE	4/8-4/15	-1.03E-04	568.60	-3.02E-09	1.64E-07	-0.02
4	WSS	4/8-4/15	6.86E-03	570.74	2.00E-07	2.31E-07	0.87
5	WFF	4/15-4/22	-3.71E-04	551.76	-1.12E-08	1.38E-07	-0.08
6	WFF	4/15-4/22	4.21E-03	555.76	1.26E-07	1.78E-07	0.71
7	WEE	4/15-4/22	-3.30E-03	557.36	-9.87E-08	6.88E-08	-1.43
8	WSS	4/15-4/22	-4.51E-03	543.32	-1.38E-07	1.08E-07	-1.28
9	WFF	4/22-4/29	-2.78E-03	562.80	-8.23E-08	8.74E-08	-0.94
10	WFF	4/22-4/29	3.50E-03	577.01	1.01E-07	1.69E-07	0.60
11	WEE	4/22-4/29	-9.04E-05	568.83	-2.65E-09	1.44E-07	-0.02
12	WSS	4/22-4/29	3.34E-03	579.86	9.60E-08	1.54E-07	0.62

¹ Does not include data on quality control blanks

Table I.2: EPA Data for Selected Variables¹**Am-241**

Sample	Location	Sampling period	Blank-corrected Activity (dpm)	Volume (m ³)	Activity Concentration (Bq m ⁻³)	CSU (Bq m ⁻³)	Z-value
1	WEE	4/7-4/15	-7.91E-03	589.20	-2.24E-07	3.06E-07	-0.73
2	WFF	4/7-4/15	-8.41E-03	575.00	-2.44E-07	3.21E-07	-0.76
3	WFF	4/7-4/15	-1.47E-02	592.50	-4.14E-07	3.15E-07	-1.31
4	WSS	4/7-4/15	-8.52E-03	602.00	-2.36E-07	2.94E-07	-0.80
5	WFF	4/15-4/22	2.04E-03	557.00	6.11E-08	3.06E-07	0.20
6	WFF	4/15-4/22	6.18E-03	616.30	1.67E-07	2.67E-07	0.63
7	WSS	4/15-4/22	4.27E-03	540.70	1.32E-07	2.89E-07	0.46
8	WEE	4/15-4/22	1.11E-02	541.60	3.40E-07	3.44E-07	0.99
9	WEE	4/22-4/29	5.91E-03	527.90	1.87E-07	2.85E-07	0.65
10	WSS	4/22-4/29	2.06E-03	525.59	6.53E-08	2.47E-07	0.26
11	WFF	4/22-4/29	1.15E-02	521.60	3.68E-07	2.92E-07	1.26
12	WFF	4/22-4/29	2.06E-03	563.40	6.09E-08	2.66E-07	0.23

Pu-238

1	WEE	4/7-4/15	7.97E-03	589.20	2.26E-07	2.72E-07	0.83
2	WFF	4/7-4/15	4.17E-03	575.00	1.21E-07	2.71E-07	0.45
3	WFF	4/7-4/15	2.12E-03	592.50	5.96E-08	3.42E-07	0.17
4	WSS	4/7-4/15	4.24E-03	602.00	1.17E-07	2.20E-07	0.53
5	WFF	4/15-4/22	-8.33E-03	557.00	-2.49E-07	2.92E-07	-0.85
6	WFF	4/15-4/22	7.78E-03	616.30	2.11E-07	2.50E-07	0.84
7	WSS	4/15-4/22	-1.00E-02	540.70	-3.09E-07	2.74E-07	-1.13
8	WEE	4/15-4/22	-4.26E-03	541.60	-1.31E-07	3.32E-07	-0.39
9	WEE	4/22-4/29	-5.87E-03	527.90	-1.85E-07	2.84E-07	-0.65
10	WSS	4/22-4/29	1.86E-03	525.59	5.90E-08	2.40E-07	0.25
11	WFF	4/22-4/29	5.95E-03	521.60	1.90E-07	3.17E-07	0.60
12	WFF	4/22-4/29	-1.46E-02	563.40	-4.32E-07	2.49E-07	-1.73

Pu-239/240

Sample	Location	Sampling period	Blank-	Volume	Activity	CSU	Z-value
			corrected		Concentration		
			Activity	(m ³)	(Bq m ⁻³)	(Bq m ⁻³)	
1	WEE	4/7-4/15	-9.58E-03	589.20	-2.71E-07	2.13E-07	-1.27
2	WFF	4/7-4/15	-1.16E-02	575.00	-3.35E-07	2.43E-07	-1.38
3	WFF	4/7-4/15	-1.16E-02	592.50	-3.25E-07	2.21E-07	-1.47
4	WSS	4/7-4/15	-9.50E-03	602.00	-2.63E-07	2.26E-07	-1.16
5	WFF	4/15-4/22	-6.57E-03	557.00	-1.96E-07	2.08E-07	-0.94
6	WFF	4/15-4/22	-8.63E-03	616.30	-2.33E-07	2.10E-07	-1.11
7	WSS	4/15-4/22	9.20E-04	540.70	2.84E-08	2.42E-07	0.12
8	WEE	4/15-4/22	-1.61E-03	541.60	-4.96E-08	2.80E-07	-0.18
9	WEE	4/22-4/29	7.64E-03	527.90	2.41E-07	2.48E-07	0.97
10	WSS	4/22-4/29	-2.16E-03	525.59	-6.85E-08	2.39E-07	-0.29
11	WFF	4/22-4/29	3.97E-03	521.60	1.27E-07	2.37E-07	0.53
12	WFF	4/22-4/29	4.69E-03	563.40	1.39E-07	2.59E-07	0.54

¹ Does not include data on blanks