

Am-241, Pu-238, Pu-239/240, and Sr-90 Decision Levels for the Environmental Air Monitoring Program for the Idaho National Laboratory

April 2021



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April 2021

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ABSTRACT

This report provides a comprehensive analysis of Am-241, Pu-238, Pu-239/240, and Sr-90 radiation data collected from air monitoring sites at or near Idaho National Laboratory from June 2013 through December 2020. These data were used to compute monitoring limits that will be used to assess future measurements of Am-241, Pu-238, Pu-239/240, and Sr-90. Data were analyzed for seasonality, stationarity, and other data issues that may impact the calculation and use of the monitoring limits.

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ACRONYMS AND ABBREVIATIONS

ANLW	Argonne National Laboratory – West
ANOVA	analysis of variance
ARA	Auxiliary Reactor Area
BLKFT	Blackfoot
CFA	Central Facilities Area
CPP	Chemical Processing Plant
CRATERS	Craters of the Moon National Monument & Preserve
EBR 1	Experimental Breeder Reactor I
EFS	Experimental Field Station
EPA	Environmental Protection Agency
IF	Idaho Falls
INL	Idaho National Laboratory
INTEC	Idaho Nuclear Technology and Engineering Center
IRC	INL Research Center
LOC	location
MFC	Materials and Fuels Complex
NRF	Naval Reactors Facility
PBF	Power Burst Facility
RHLLW	Remote-Handled Low-Level Waste
RTC	Reactor Technology Complex
RWMC	Radioactive Waste Management Complex
SMC	Specific Manufacturing Capability
TAN	Test Area North
TRA	Test Reactor Area
UTL	upper tolerance limit

Am-241, Pu-238, Pu-239/240, and Sr-90 Decision Levels for the Environmental Air Monitoring Program for the Idaho National Laboratory

1. INTRODUCTION

Data are collected from air monitors in and around Idaho National Laboratory (INL) and analyzed for certain radionuclides quarterly. This report provides a statistical analysis of the measurements of Am-241, Pu-238, Pu-239/240, and Sr-90 collected from these sampling events. Historical data are used to compute decision limits that are presented at the end of this report. A preliminary data analysis is also included to ensure that the appropriate decision limits were calculated and that information is available to aid in the use of the decision limits.

The R statistical package was used to create the graphs and perform all calculations except where otherwise noted (R Core Team 2021). Data manipulation was done using base R and the dplyr package (Wickham et al. 2021). Graphs were constructed using ggplot2 (Wickham 2016).

2. PRELIMINARY DATA ANALYSIS

Air monitoring data for Sr-90 were collected quarterly from June 2013 through December 2020 are used in this analysis. Data for Am-241, Pu-238, and Pu-239/240 were collected quarterly from September 2014 through December 2020. The data were examined for undesirable traits such as outliers and inconsistencies that indicate possible data quality issues. Data were assessed for seasonality, stationarity, and normality. Each of these traits is discussed in this section. Detection limits are used in place of non-detects for graphs. Normally, Kaplan-Meier estimation would be used for computations to appropriately address non-detects. However, it does not perform well when less than 50% of the observations are detected, which is the case for all four radionuclides.

2.1 Outliers and Quality

Most of the data were non-detects. Outliers are present in the data for each of the four radionuclides. However, all of the outliers in the Am-241, Pu-238, and Sr-90 data are minimum detected activities (MDAs) for non-detect observations. The largest MDAs for all four radionuclides were from the December 29, 2019 sampling event. MDAs from the December 29, 2019 sampling event should not be included in the determination of decision levels because they are so much larger than all other MDAs and observed activities for detected observations. A second unusually large MDA was observed for a non-detect Pu-238 value for the June 27, 2018 sampling event. It was also not used in the analysis of the data in this section. A detected outlier was observed in the Pu-239/240 data for the June 27, 2018 sampling event. The detected outlier was collected from Reactor Technology Complex (RTC) during June 2018. It was retained for the computation of decision levels because it was considered a legitimate measurement. Figure 1 shows the activities observed data by date. Detected data are denoted by solid, blue circles. MDAs for the non-detected values are shown on the graph with hollow orange circles.

Data were also examined for anomalies that indicate dubious data quality. The data appear to be of high quality with no observations reported below 0 that were also not U-flagged.

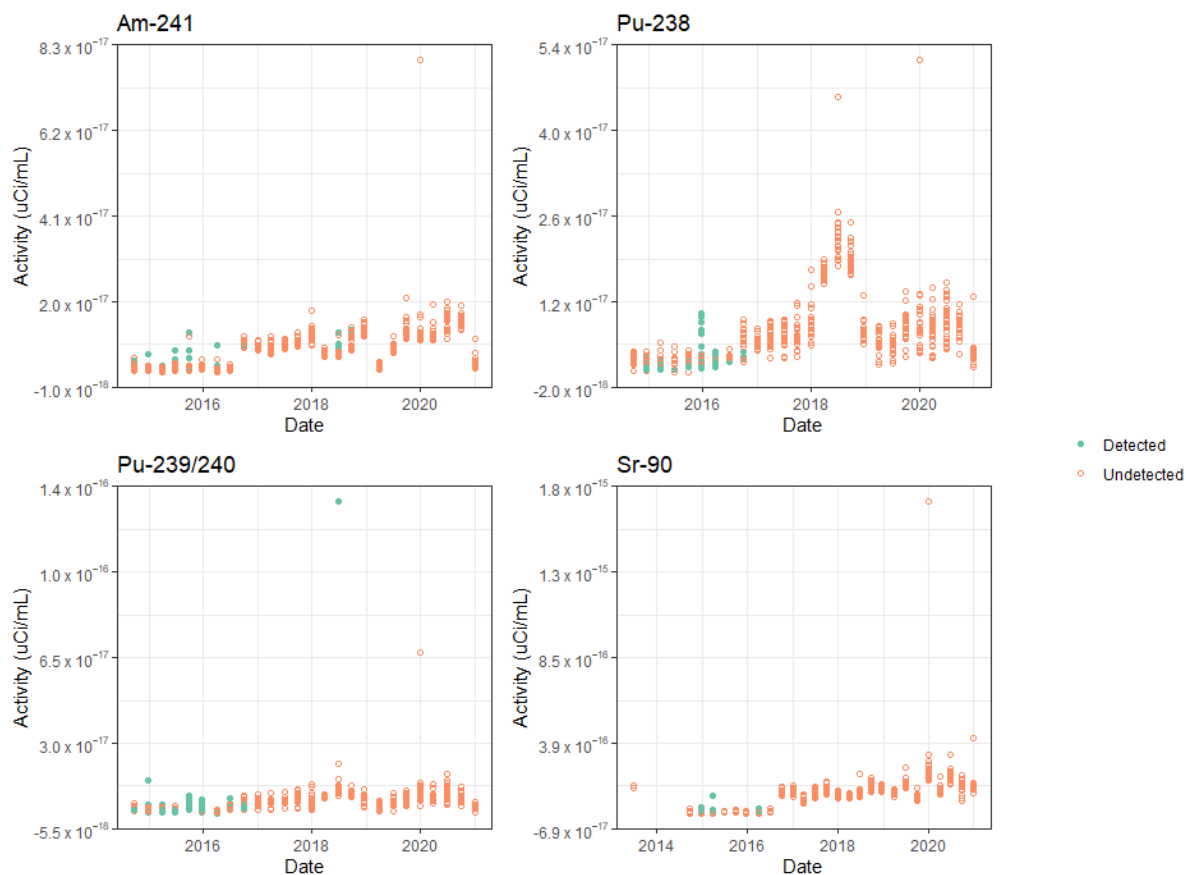


Figure 1. Am-241, Pu-238, Pu239/240, and Sr-90 concentrations measured at INL air monitoring locations, 2013–2020.

2.2 Seasonality

Data were assessed for seasonality, which is a cyclic pattern that occurs over time (EPA 2009 and Gilbert 1987). Figure 2 shows the mean and median activities for each of the radionuclides by month. The concentrations appear to be consistent across all sampling events. The means for the June and December sampling events appear higher than the others, but this is because of the presence of a few outliers. Overall, there does not appear to be seasonality in the data.

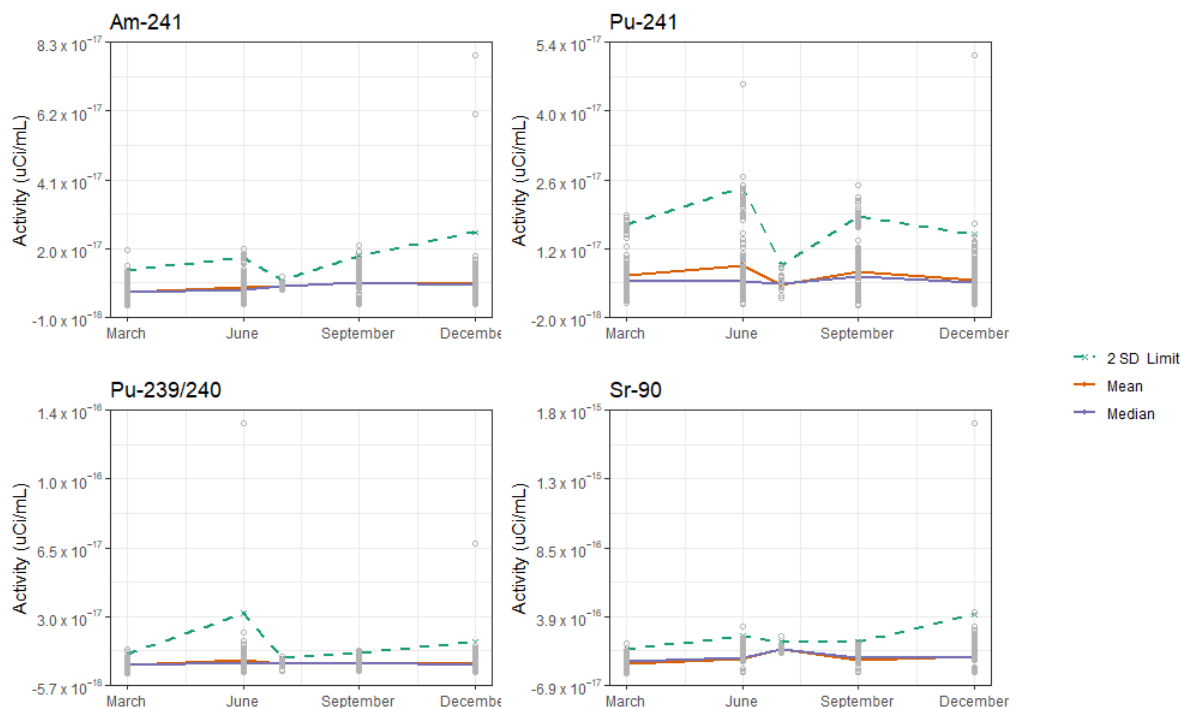


Figure 2. Radionuclide concentrations by month. The average, median, and 2 standard deviation concentration for each month are included for reference.

2.3 Stationarity

Data were also examined for stationarity. Stationarity is when the mean concentration and standard deviation are the same across the area being monitored (EPA 2009 and Gilbert 1987). Analysis of variance (ANOVA) and Kruskal-Wallis were used to determine if the mean or median concentration was different at the different air monitoring sites. ANOVA and Kruskal-Wallis can indicate only if at least one site is different from the others—but cannot identify which sites are different from each other.

The data for Am-241, Pu-238, Pu-239/240, and Sr-90 were examined for normality prior to determine if ANOVA is the appropriate test for evaluating differences between monitoring sites. ANOVA assumes that the residuals of the model are normally distributed. Figures 3-6 shows the distribution of the residuals obtained from the ANOVA model computed using the raw data for each radionuclide and the distribution of the residuals from the model constructed from the natural logarithm of each radionuclide. The first plot in each figure is a residual plot. The points on this plot should look like a random cloud. The second plot is a histogram. It shows the shape, or distribution, of the residuals. If the residuals follow a normal distribution then the histogram will have a bell shape. The histogram can also show if the data are symmetric even when they are not normally distributed. Symmetry can compensate for some degree of non-normality. The third plot is a normal-quantile plot. This is an effective way to determine if the residuals come from a normal distribution. If the residuals are normally distributed the normal-quantile plot follows a straight line. The MDA was used for observations that were non-detects. Figure 3-5 shows that the residuals from the transformed Am-241, Pu-238, and Pu-239/240 data are approximately normally distributed. However, Figure 6 shows that the residuals from the raw and transformed Sr-90 data are not normally distributed. Thus, ANOVA was used to assess stationarity for the Am-241, Pu-238, and Pu-239/240 transformed data and Kruskal-Wallis was used to assess stationarity for the transformed Sr-90 data.

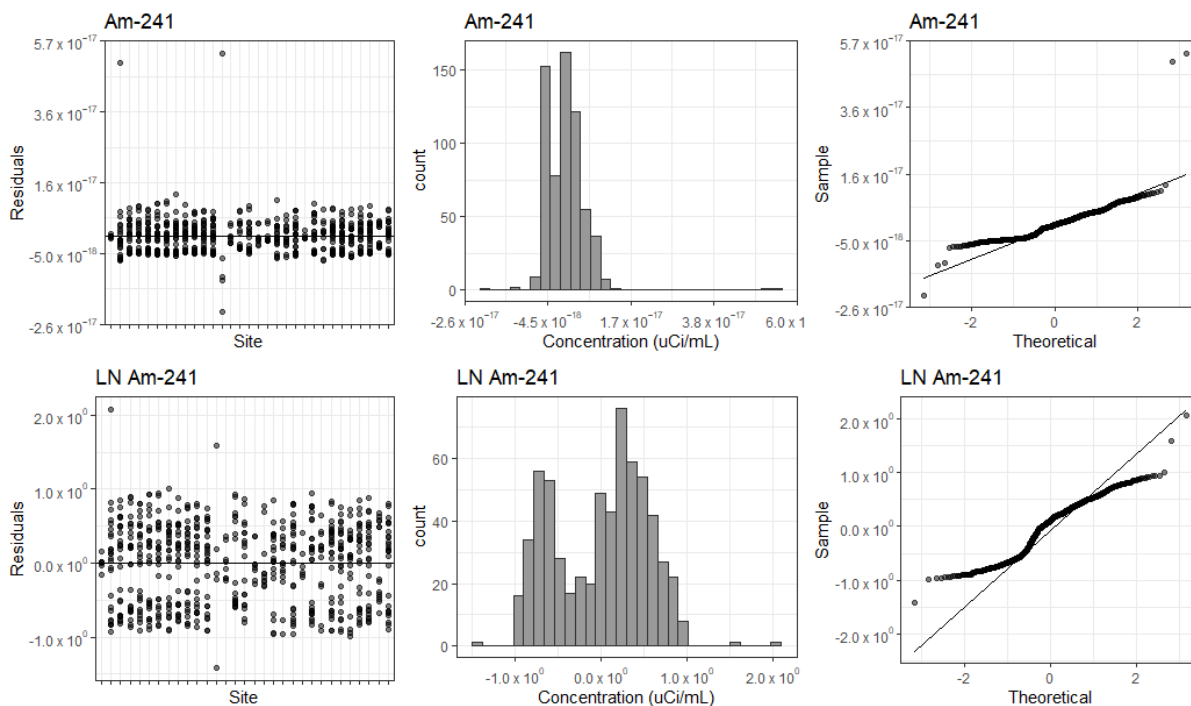


Figure 3. Residual plots, histograms, and normal-quantile plots of residuals from the ANOVA model generated using the raw Am-241 data and the natural logarithm of the Am-241 data.

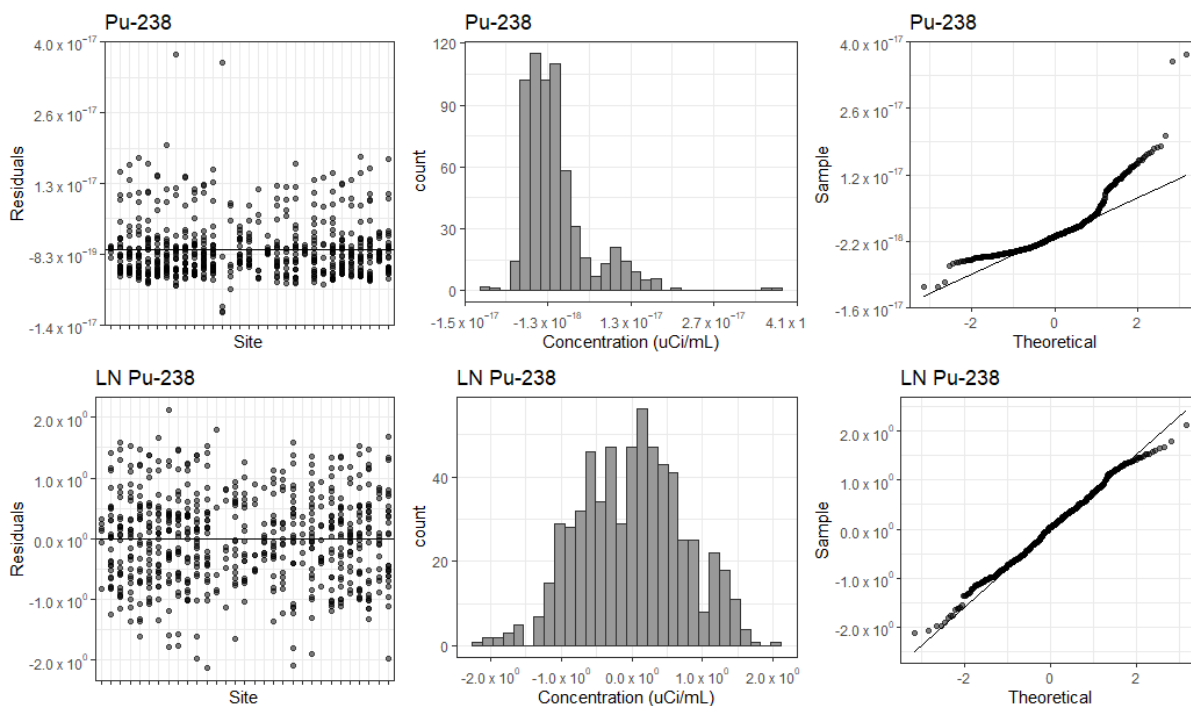


Figure 4. Residual plots, histograms, and normal-quantile plots of residuals from the ANOVA model generated using the raw Pu-238 data and the natural logarithm of the Pu-238 data.

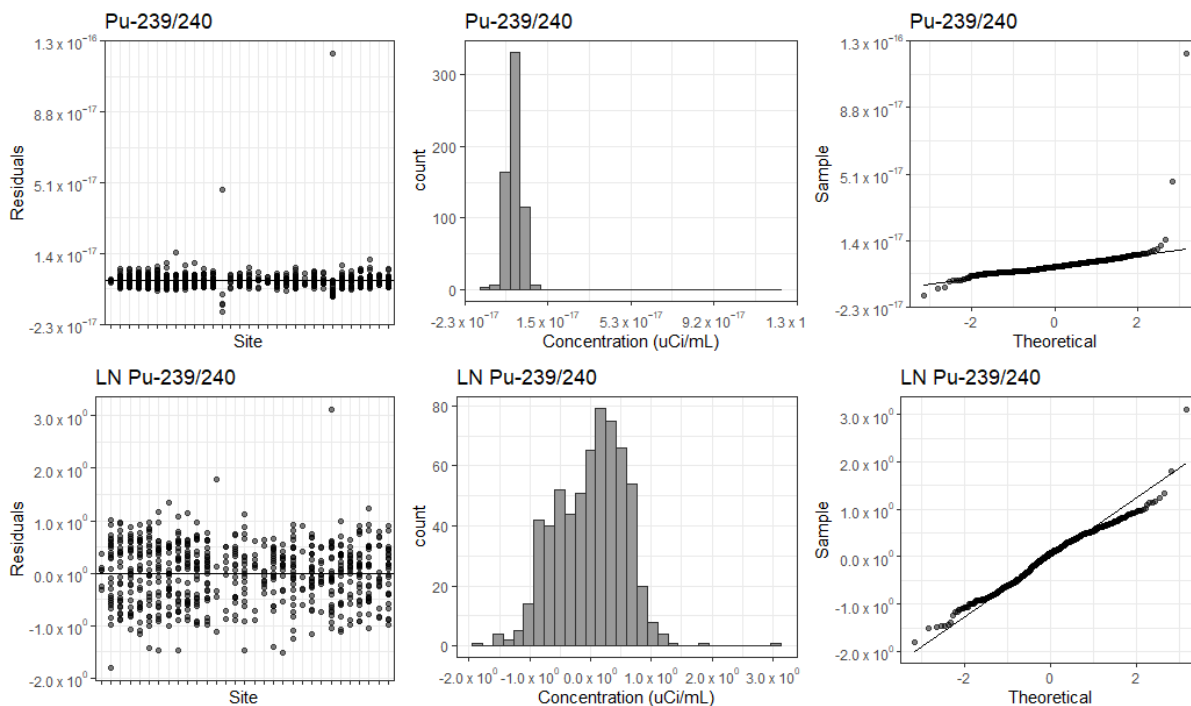


Figure 5. Residual plots, histograms, and normal-quantile plots of residuals from the ANOVA model generated using the raw Pu-239/240 data and the natural logarithm of the Pu-239/240 data.

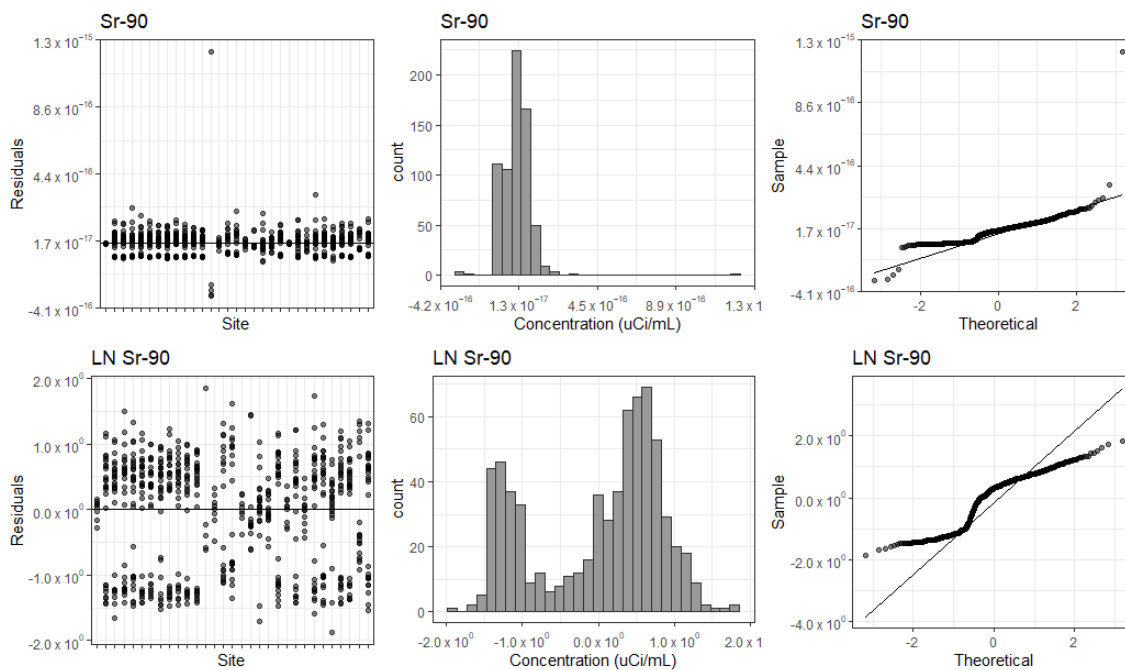


Figure 6. Residual plots, histograms, and normal-quantile plots of residuals from the ANOVA model generated using the raw Sr-90 data and the natural logarithm of the Sr-90 data.

The ANOVA tests for Am-241, Pu-238, and Pu-239/240 and the Kruskal-Wallis test for Sr-90 all had p-values that were less than 0.05. This indicates that the mean activity, or median value in the case of the Kruskal-Wallis test, is different for at least one air monitoring site. However, the ANOVA and Kruskal-Wallis tests cannot identify the site, or sites, that are different. Testing for differences between the individual air monitoring sites was done using the t-test for Am-241, Pu-238, and Pu-239/240 in conjunction with the false discovery rate (Benjamini and Hochberg 1995) to adjust for multiple test comparisons. The Nemenyi test for multiple comparisons (Nemenyi 1963) was done for the Sr-90 data to determine which sites are different. This resulted in 435 comparison tests for each radionuclide. Table 1 shows a summary of the multiple test results for each radionuclide. Although the p-value for Am-241 indicates that at least one air monitoring location is different, none of the pairwise comparison tests were significantly different. This means that the Am-241 concentrations are stationary across the area being monitored. Several pairwise tests indicate that there are differences between air monitors for the other three radionuclides. Table 2 shows the pairs that are significantly different from each other. Pu-238 had the most differences with 8.3% of the tests being significantly different. Most of the contrasts that were significantly different involved air monitors at the Materials Fuels Complex (MFC) which indicates that concentrations of Pu-238 at MFC are different than much of the rest of the monitored area. Pu-239/240 also had many significantly different contrasts with 6.7% of the tests having p-values less than 0.05. Seventy-two percent of the significant contrasts involved Auxiliary Reactor Area (ARA), which indicates that concentrations of Pu-239/240 are different at ARA than much of the rest of the monitored site. Only 1.1% of the contrasts for Sr-90 were significantly different from each other. Most of these involved MFC or Remote-Handled Low-Level Waste (RHLLW) Disposal Facility. However, so few of the test were significantly different the Sr-90 data can be considered stationary. These results indicate that the Pu-238 and Pu-239/240 data are not stationary among sites. Figures 7-10 shows boxplots of each of the radionuclides measured at each site. They are ordered from highest median concentration to lowest median concentration. The boxplots show the relationship between the concentrations of radionuclides at the different monitoring locations. It can be seen in the boxplots that concentrations of all four radionuclides are generally lower at MFC, Power Burst Facility (PBF), and ARA than at the other monitoring sites.

Table 1. Summary of results for ANOVA and Kruskal-Wallis tests for stationarity. A summary of the pairwise contrast tests is also included in the table.

Radionuclide	P-value for ANOVA or Kruskal-Wallis	Number of Contrasts	Number of Different Pairwise Comparisons	Percent of Positive Tests
Am-241	0.0095	435	0	0
Pu-238	<0.0001	435	36	8.3
Pu-239/240	0.0013	435	29	6.7
Sr-90	0.0002	435	5	1.1

Table 2. List of air monitoring sites that were determined to be different based on pairwise statistical contrast tests.

Am-241	Pu-238	Pu-239/240	Sr-90
None	ARA/CPP	ARA/Blackfoot	ARA/RHLLW
	ARA/RWMCS	ARA/CFA	MFC/MFCS
	ARA/RHLLW	ARA/CPP	MFC/RHLLW
	Blackfoot/RHLLW	ARA/Craters of the Moon	MFCS/PBF
	CFA/MFC	ARA/EBR-1	PBF/RHLLW

Am-241	Pu-238	Pu-239/240	Sr-90
	CPP/MFC	ARA/EFS	
	Craters/MFC	ARA/IRC	
	EBR-1/MFC	ARA/IRC	
	Idaho Falls/MFC	ARA/IRC	
	CPP (LOC A)/MFC	ARA/IRC	
	CPP (LOC A)/MFC	ARA/IRC (LOC A)	
	CPP (LOC A)/PBF	ARA/RWMCS (LOC B)	
	CPP (LOC A)/PBF	ARA/MFCN	
	CPP (LOC A)/TRA	ARA/MFCN	
	INTEC/RWMCS (LOC B)	ARA/MFCS	
	INTEC/RHLLW	ARA/NRF	
	INTEC/RHLLW	ARA/REST	
	CFA (LOC B)/RHLLW	ARA/REST	
	RWMCS (LOC B)/MFC	ARA/RHLLW	
	RWMCS (LOC B)/MFC	ARA/RTC	
	RWMCS (LOC B)/PBF	ARA/RWMC	
	RWMCS (LOC B)/TRA	ARA/RWMC	
	MFC/MFCN	ARA/RWMCS	
	MFC/MFCS	ARA/SMC	
	MFC/REST	ARA/Sugar City	
	MFC/RHLLW	ARA/VAN B	
	MFC/RTC	EFS/MFC	
	MFC/RWMCS	CFA (LOC B)/RHLLW	
	MFC/SMC	MFC/MFCS	
	MFC/Sugar City	MFC/RHLLW	
	MFCN/PBF	MFC/RWMC	
	MFCN/TRA	MFC/SMC	
	MFCN/TRA	PBF/RHLLW	
	MFCN/TRA	RHLLW/TRA	
	MFCN/TRA	RWMCS/TRA	
	PBF/RHLLW		
	PBF/RTC		
	PBF/RWMCS		
	RHLLW/TRA		
	RHLLW/VAN B		
	SMC/TRA		

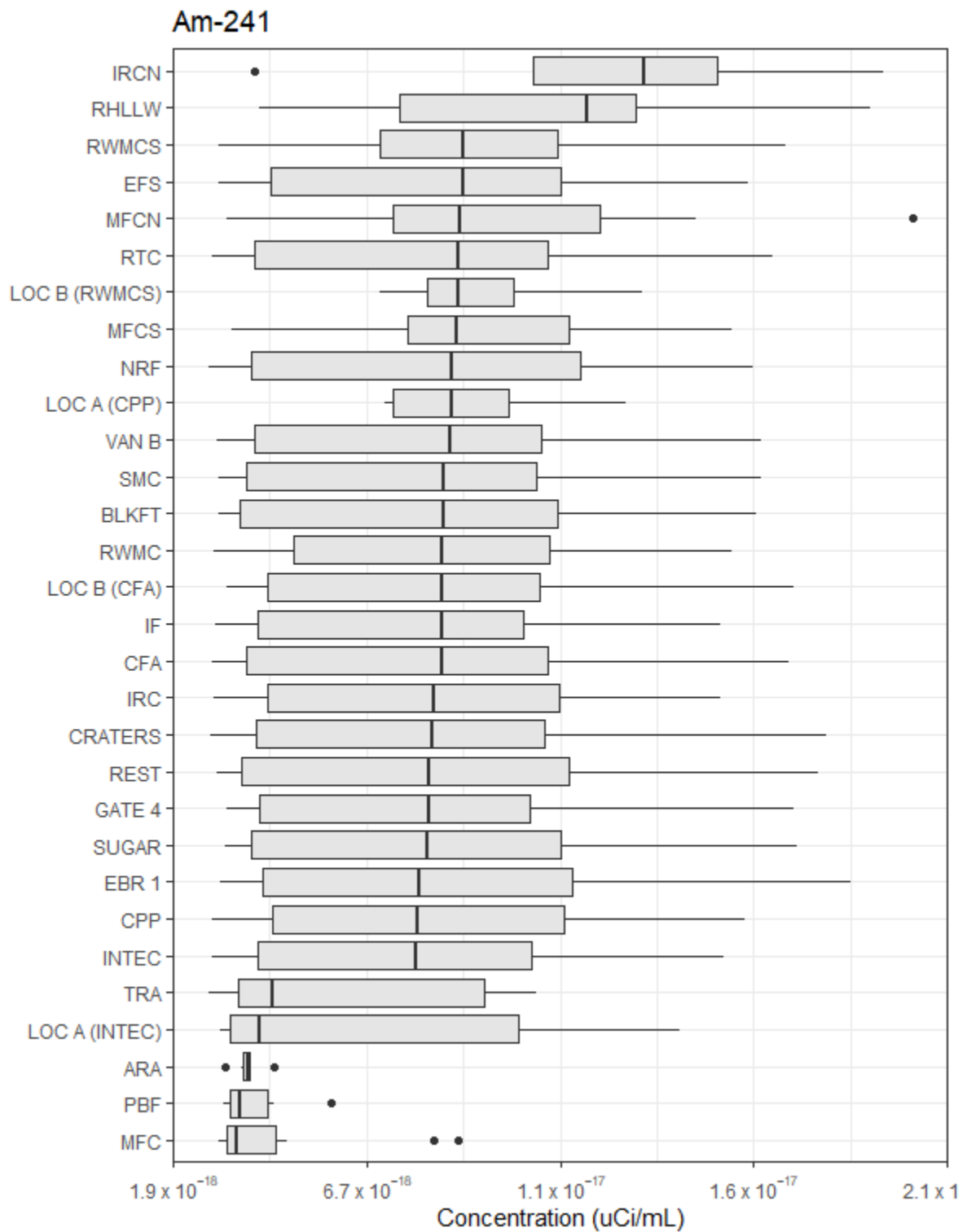


Figure 7. Boxplots showing the concentrations of Am-241 at each monitor location.

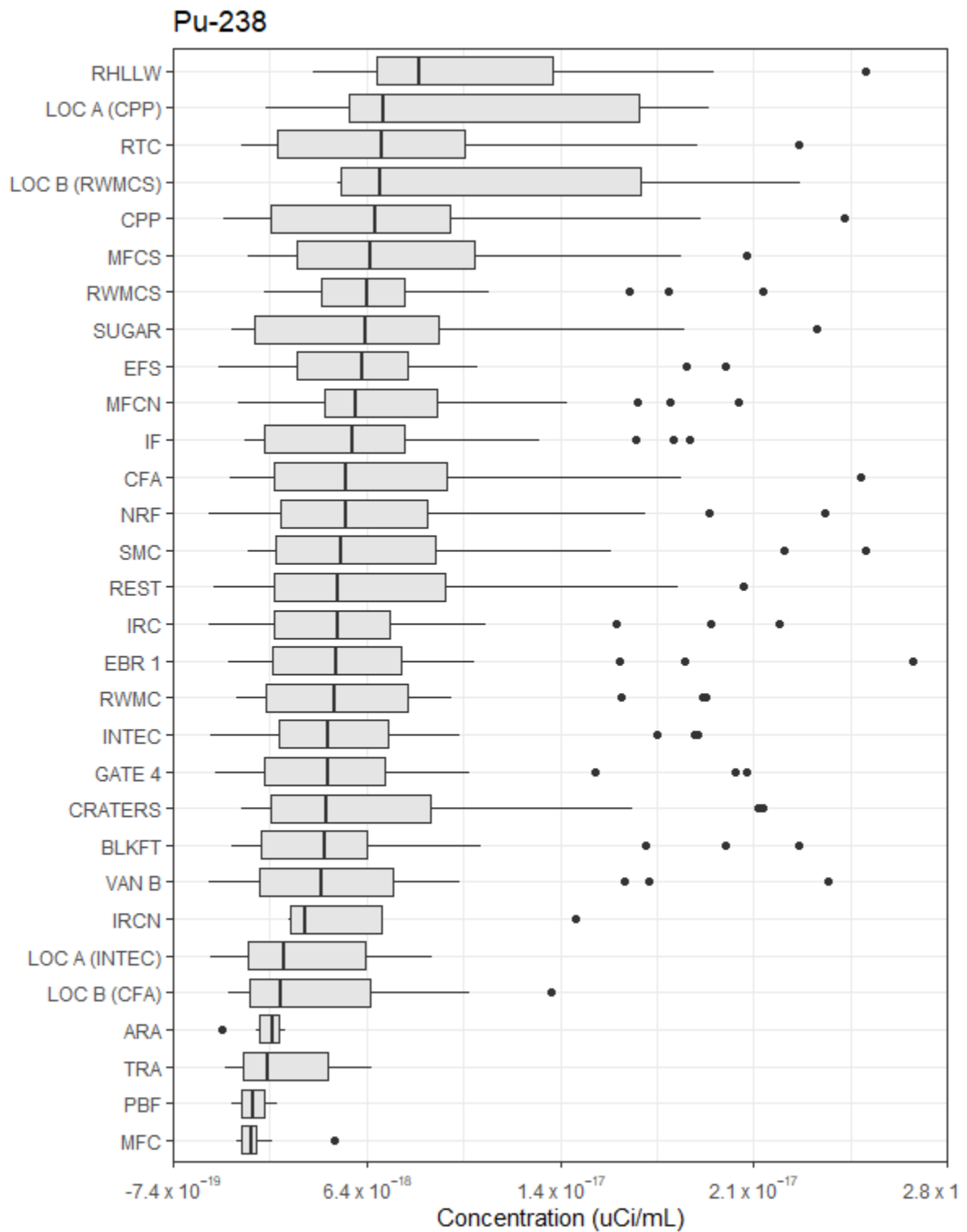


Figure 8. Boxplots showing the concentrations of Pu-238 at each monitor location.

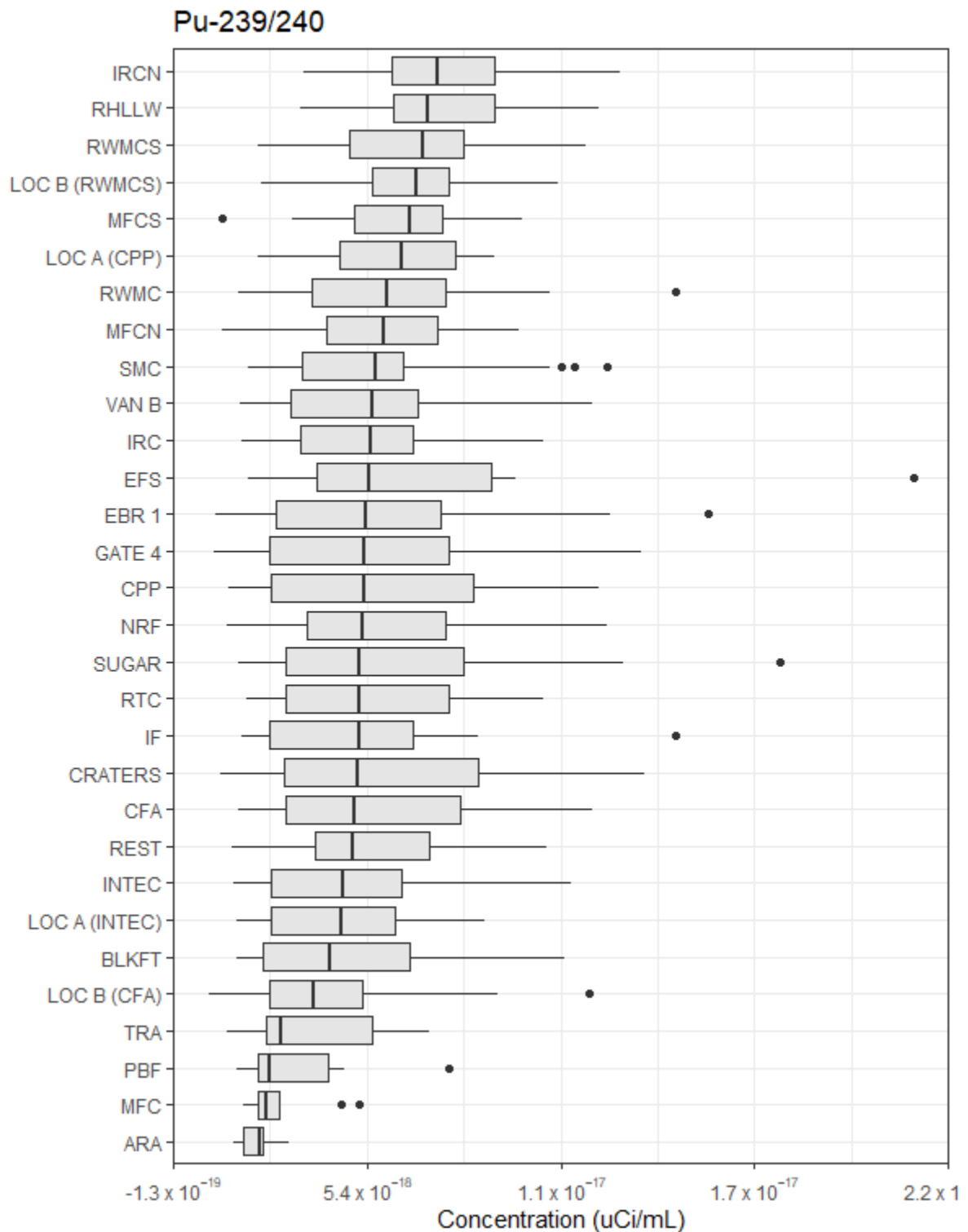


Figure 9. Boxplots showing the concentrations of Pu-239/240 at each monitor location. Note that there is an extreme outlier from Reactor Technology Complex (RTC) that has been omitted from the plot.

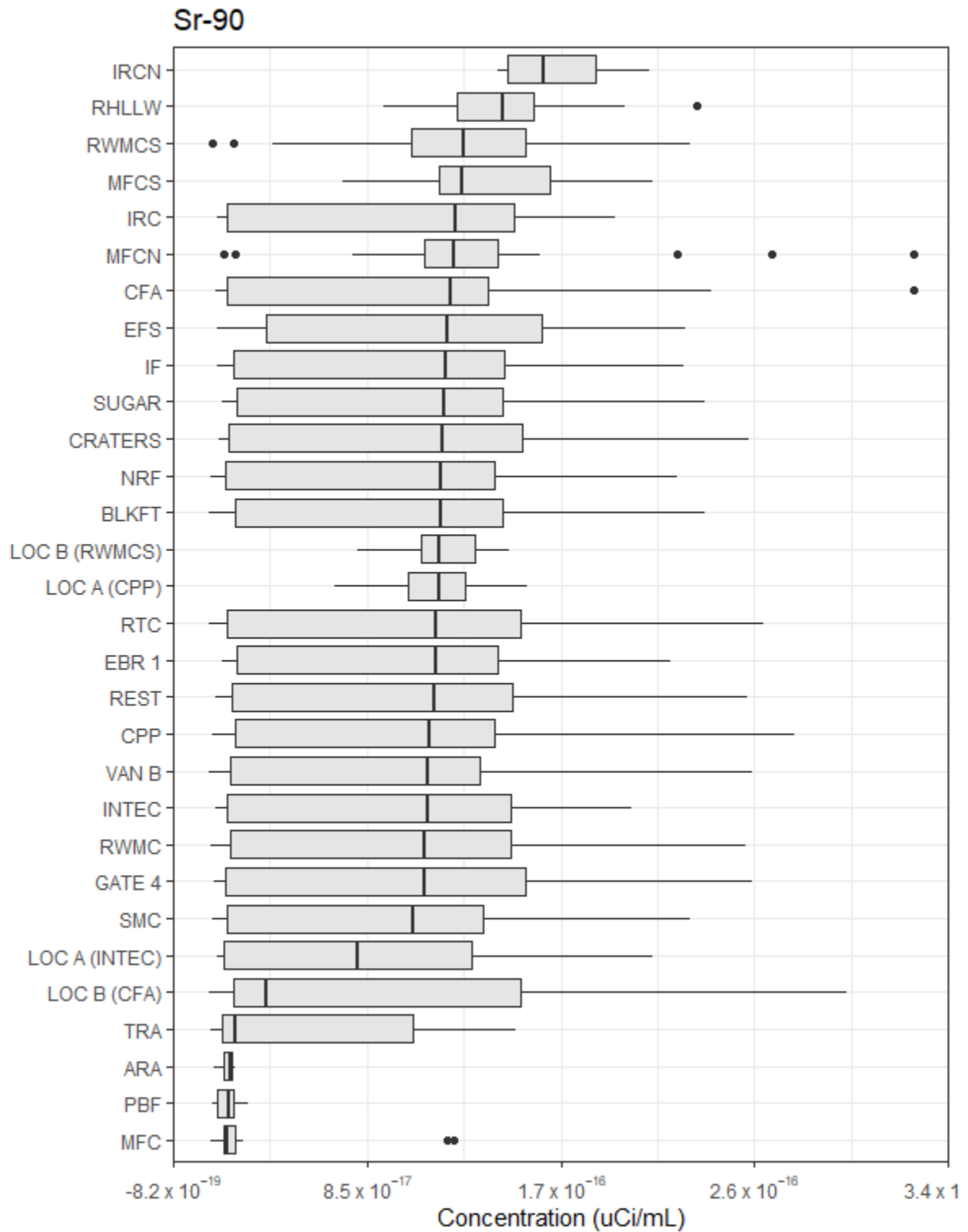


Figure 10. Boxplots showing the concentrations of Sr-90 at each monitor location.

2.4 Summary Statistics

Summary statistics and statistical plots are presented in this section. Plots include a histogram, normal-quantile plot, and boxplot for each radionuclide. The histogram shows the overall shape, or distribution, of the data. It can identify outliers and skewness in the data. The normal-quantile plot illustrates if the data follow a normal distribution. If the points in the graph follow a straight line then the data are considered normally distributed. Deviations from the line indicate that the data are not normally distributed. The boxplot graphs the minimum, first quartile, median, third quartile, and maximum observed values. It also shows potential outliers in the data with points above or below the lines. However, if the data are skewed, points in the tail will be shown as outliers even if they are not actual outliers. Figure 11 shows the plots for the Am-241, Pu-238, Pu-239/240, and Sr-90 concentrations. The outliers mentioned in Section 2.1 were removed prior to making the plots. MDAs were plotted for non-detect values. The graphs show that none of the datasets are normally distributed. They are all right-skewed. The data do not appear to have any outliers after the removal of those cited in Section 2.1 except for the Pu-239/240 outlier observed from the RTC monitor on June 27, 1918. Most of the observations are non-detects so summary statistics cannot be computed on the data. Table 3 shows the number of observations for each site and the number and percent of non-detects for each site.

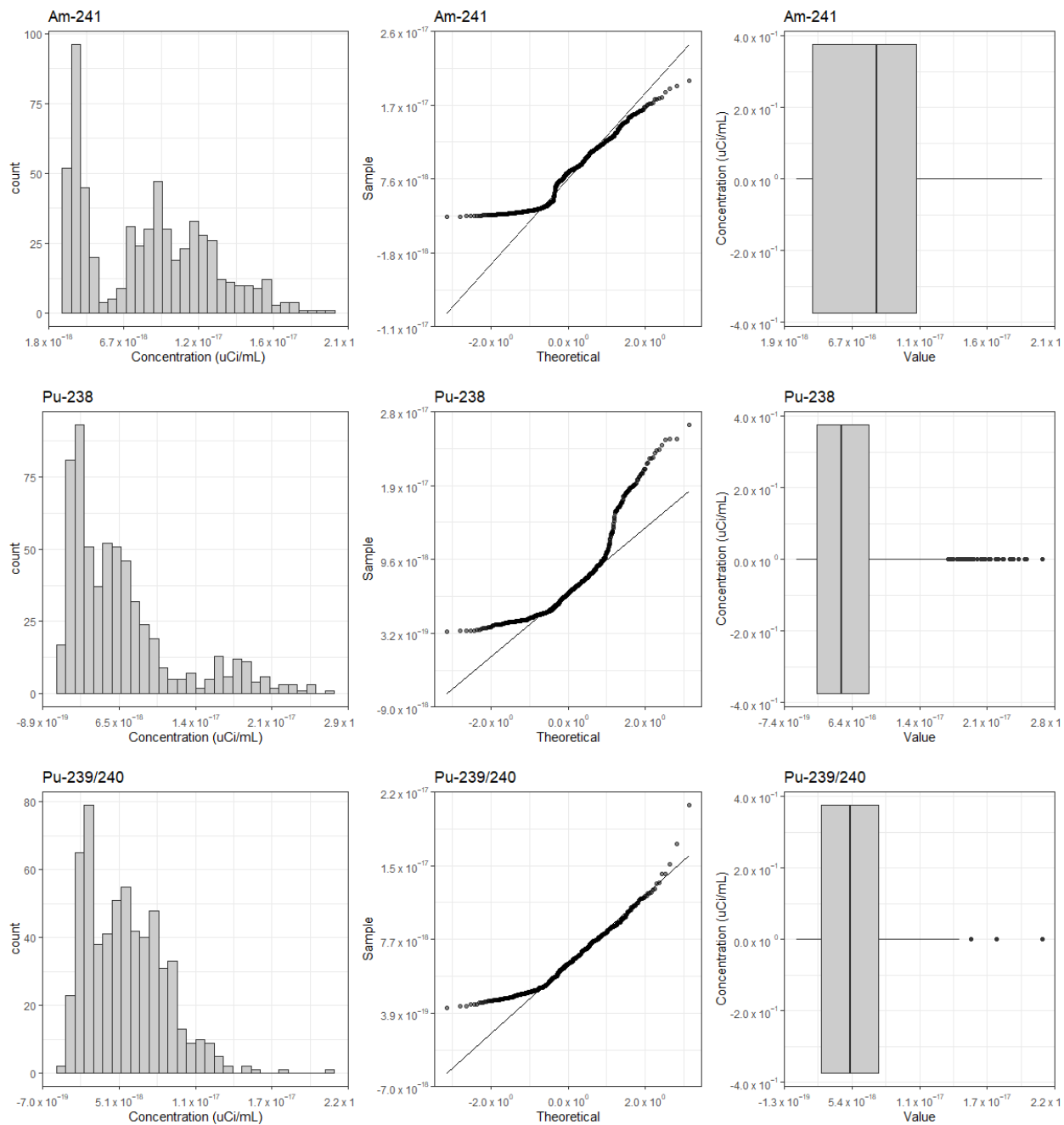


Figure 11. Histograms, normal-quantile plots, and boxplots for Am-241, Pu-238, Pu-239/240, and Sr-90 concentrations.

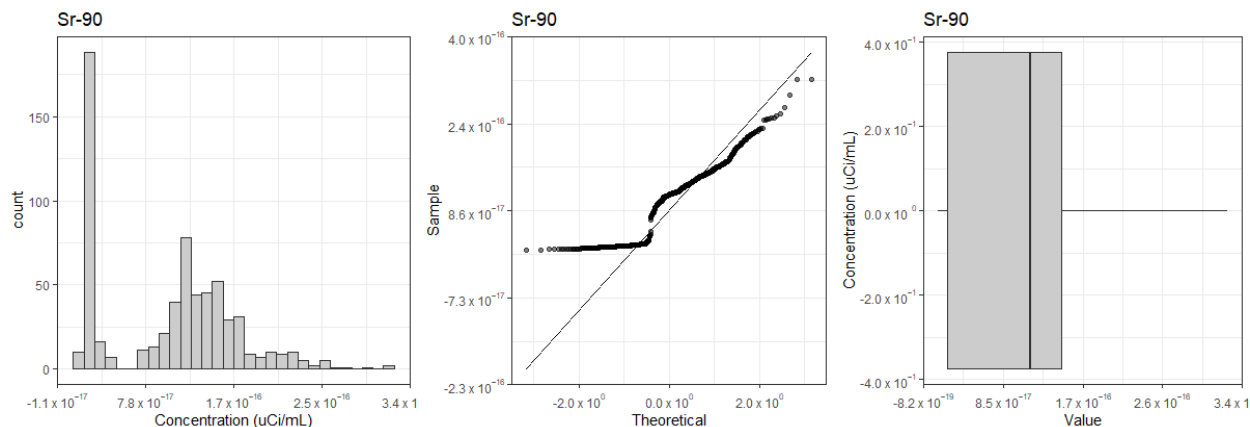


Figure 11 (cont.). Histograms, normal-quantile plots, and boxplots for Am-241, Pu-238, Pu-239/240, and Sr-90 concentrations.

Table 3. Number of samples collected for each radionuclide and the percentage of detects.

Radionuclide	Number of Samples	Percent Detected
Am-241	601	3.49
Pu-238	601	10.3
Pu-239/240	601	17.8
Sr-90	647	4.33

3. DECISION LIMITS

Decision limits were computed for Am-241, Pu-238, Pu-239/240, and Sr-90. Because more than 50% of the observations were non-detects, the larger of the maximum observed detection or the MDA was used as the decision limit for each radionuclide. Note that the outlier MDAs were not used as decision limits.

Table 4. Decision limits for Am-241, Pu-238, Pu-239/240, and Sr-90 air monitoring.

Radionuclide	Decision Limit	Value Type
Am-241	7.90E-17	Detection Limit
Pu-238	2.65E-17	Detection Limit
Pu-239/240	1.29E-16	Observed Value
Sr-90	4.19E-16	Detection Limit

Because there is no seasonality in the data, the decision limits from Table 4 are applicable for measurements collected all year long. There is a lack of stationarity across the site being monitored. However, the boxplots in Figures 7-10 show that the decision limits for each radionuclide and monitoring site. The boxplots demonstrate that the decision limits should be appropriate for all sites.

4. REFERENCES

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