A REVIEW OF RECENT IMPURITY MEASUREMENTS OF LANL MATERIAL

T.B. Edwards

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Applied Computational Engineering and Statistics
Savannah River National Laboratory
Aiken, SC 29808

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REVIEWS AND APPROVALS

AUTHOR:

_____
T. B. Edwards, Applied Computational Engineering and Statistics   Date
Savannah River National Laboratory

TECHNICAL REVIEWER:

_____
E. P. Shine, Applied Computational Engineering and Statistics   Date
Savannah River National Laboratory

APPROVER:

_____
P. L. Lee, Manager, Applied Computational Engineering and Statistics   Date
Savannah River National Laboratory

EXECUTIVE SUMMARY

The Applied Computational Engineering and Statistics (ACES) group of the Savannah River National Laboratory (SRNL) was asked to review recent measurements performed by the Los Alamos National Laboratory (LANL) on material from that facility that is being considered for processing through the Mixed Oxide (MOX) Fuel Fabrication Facility (MFFF) at the Savannah River Site (SRS). There are specification limits for impurities in the feed to the MOX facility: a maximum limit and an exceptional limit. The limits for an impurity apply to the population of concentration values for that impurity for a class of material that is to be processed through MOX. For the purposes of this report, these limits were defined as follows. The concentration of an elemental impurity, expressed as micrograms of the element per gram of plutonium (μg/g Pu), is to be no more than the maximum limit for that element for 98% of the material coming through MOX; that is, 98% of the material processed at MOX is to have a concentration of the given element less than the maximum limit. In addition, the concentration for a given element is to be no more than the exceptional limit for that element for 99.9% of the material processed through MOX.

The measurements evaluated as part of this study included LANL blend lots 1 through 29 and cover carbon (C), chlorine (Cl), fluorine (F), nitrogen (N), phosphorous (P), and sulfur (S). Note that all of the measurements for each impurity were below their respective maximum (and obviously, therefore, their exceptional) limits. Thus, there is no immediate concern regarding the LANL material being suitable for processing through MOX. Two approaches were used to investigate the quantiles of the impurity populations. The first approach used was a nonparametric approach. While the results from this approach did not indicate any problems for any of the impurities, there was too little data available to lead to confident statements about satisfying the maximum and exceptional limits. Impurity data from additional random samples of the LANL material would be needed to increase the confidence level associated with the results from a nonparametric approach for investigating the population quantiles of interest.

For S, F, and Cl, only a nonparametric approach was used. A second approach, a parametric approach, was attempted for C, P, and N. However, the results for the P and N measurements indicated that each of their respective populations was not well modeled by a normal or by a lognormal distribution. Thus, the conclusions for the P and N populations were those provided by the nonparametric approach. From the parametric approach, the results for C indicated no issue in the LANL material meeting the maximum limit for this element assuming that the measurements for this element follow a lognormal distribution.

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1.0 INTRODUCTION

The Applied Computational Engineering and Statistics (ACES) group of the Savannah River National Laboratory (SRNL) was asked to review recent measurements performed by the Los Alamos National Laboratory (LANL) on material from that facility that is being considered for processing through the Mixed Oxide (MOX) Fuel Fabrication Facility (MFFF) at the Savannah River Site (SRS). There are specification limits for impurities in the feed to the MOX facility: a maximum limit and an exceptional limit. For the purposes of this report, these limits were defined as follows. The concentration of an elemental impurity, expressed as micrograms of the element per gram of plutonium (μg/g Pu), is to be no more than the maximum limit for that element for 98% of the material coming through MOX; that is, 98% of the material processed at MOX is to have a concentration of the given element less than the maximum limit. In addition, the concentration for a given element is to be no more than the exceptional limit for that element for 99.9% of the material processed through MOX. For example, the concentration of sulfur (S) must be no more than 250 μg S/g Pu for 98% of the material processed in MOX and no more than 1000 μg S/g Pu for 99.9% of the material processed in MOX.

Thus, these specification limits for an elemental impurity represent a pair of select quantiles (i.e., the 98th and 99.9th quantiles) of the population of concentrations for that impurity for a class of material that is being considered for processing through MOX. One such class of material is that from LANL. The objective of this review was to use recent measurements to assess the performance of the LANL material against these limits. Questions of interest in this assessment included:

- Is there an indication of a problem with the LANL material meeting the specification limit for one or more of the elemental impurities being considered?
- Do the available data on the impurities being considered provide a strong case for limiting the need for additional sampling of the LANL material?

The analyses presented in this report were conducted using JMP Version 9.0.0 [1] and ProUCL Version 4.1.01 [2].

2.0 MEASUREMENTS

The measurements evaluated as part of this study included LANL blend lots 1 through 29 and cover carbon (C), chlorine (Cl), fluorine (F), nitrogen (N), phosphorous (P), and sulfur (S). The measurements are provided in Table 1. A numerical value in Table 1 that is led by a "<"indicates a value below detection, with the numerical value representing the detection limit. Also provided in Table 1 are the maximum and exceptional limits for each of these elements. Note that all of the measurements for each impurity are below their respective maximum (and obviously, therefore, their exceptional) limits. Thus, there was no immediate concern regarding the LANL material being suitable for processing through MOX.

Table 1. LANL Measurements

BLEND LOT		C	N	S		F	Cl	P
1		46	16	< 12		< 35	< 35	71
2		46	< 11	< 12		< 34	< 34	22
3		35	< 12	< 12		< 35	< 35	84
4		69	< 12	< 12		< 35	< 35	68
5		57	49	< 7.6		< 23	< 23	45
6		61	22	< 7.6		< 23	< 23	18
7		49	< 11	< 7.6		< 23	< 23	23
8		76	< 11	< 11		< 23	< 23	130
9		110	25	< 11		< 23	< 23	110
10		88	< 12	< 12		< 23	< 23	130
11		80.2	14.9	< 11		< 23	< 23	94
12		80.2	19.5	< 11		< 23	< 23	130
13		69	< 14	< 11		< 23	< 23	120
14		100	17	< 11		< 23	< 23	110
15		46	16	< 12		< 23	< 23	22
16		99	36	< 11		< 23	< 23	44
17		38	43	< 12		< 23	56.4	31
18		34	43	< 11		< 23	< 23	36
19		54	15	< 11		<23	<23	32
20		127	30.5	< 12		< 23	< 23	44
21		53	29	< 11		< 23	< 23	37
22		56	390	< 12		< 35	< 35	53
23		23	275	< 12		< 35	< 35	31
24		58	255	< 12		< 35	< 35	23
25		53	< 34	< 11		< 34	< 34	44
26		100	< 30	< 12		< 35	< 35	31
27		84	< 30	< 12		< 34	< 34	21
28		31	< 35	< 12		< 35	< 35	27
29		180	< 35	< 12		< 35	< 35	34
Maximum		500	400	250		NA[f]	NA	200
Exceptional		1500	400	1000		500	500	1000
		all elements expressed as μg/g Pu						

[f] Not Available ---- there is no individual maximum limit for this element.

3.0 CONFIDENCE LIMITS FOR POPULATION QUANTILES

As stated above, the maximum and exceptional limits for an impurity apply to the population of concentration values for that impurity for a class of material that is to be processed through MOX. For this investigation, it was not enough to know about the summary statistics of the population, such as its mean and standard deviation. Here the shape of the distribution of values in the impurity population and specifically the values of the pair of upper quantiles linked to the specification limits were of primary interest. Statistical methods can be used to support this investigation if the impurity data provided in Table 1 are considered as random samples from these impurity populations. Thus, the populations of interest in this study were those for the C, P, N, Cl, F, and S, and the statistical methods employed had as their objective the estimation of quantiles of these populations which were to be expressed as confidence limits.

Two approaches were used to investigate the quantiles of each impurity population: a nonparametric (or distribution-free) approach and a parametric approach (i.e., an approach that relies on a specified

probability distribution to represent the population of possible values for the impurity). The goal in each of these investigations was the determination of confidence limits for select quantiles of the population.

3.1 Nonparametric Approach

A nonparametric approach was considered first. There were 29 results reported in Table 1 for each of the indicated elemental concentrations. Two mathematically-equivalent nonparametric methods were applied to each of these sets of measurements. The first utilized an approach presented in [3] that is based on the use of order statistics for the set of measurements from Table 1. These order statistics may be denoted as $X_{(1)}, X_{(2)}, \ldots, X_{(29)}$, where $X_{(1)}$ is the smallest measurement and $X_{(29)}$ is the largest measurement in the set of data. Then, a $(1-\alpha) \times 100\%$ upper confidence limit for the qth quantile, Y_q, of the impurity population Y may be constructed using the binominal probability distribution by determining the smallest value for α between 0 and 1 such that:

$$\text{Probability}[Y_q > X_{(29)}|X_{(29)}] \leq q^{29} \leq \alpha$$

This approach was used to bound various quantiles of the impurity population represented by any of these random samples with the designated confidence. To restate the conclusion provided by this method, there is at least $(1-\alpha) \times 100\%$ confidence that at least $q \times 100\%$ of the population is less than the largest value seen in the random sample (or largest detection limit if all values are below detection). Table 2 presents a listing of the confidence limits that may be assumed for various quantiles of a population of values that are bounded by the largest value in a random sample of 29 observations. For example, consider the shaded row above in Table 2. There is $100\% \times (1-.4)=60\%$ confidence that 96.89% of the population is less than the largest value recorded in the set of 29 measurements randomly selected from the population. The last entry presented in Table 2 provides a confidence statement about the largest sample measurement providing an estimate of the 98th percentile of the population. From this entry, there is only 44% confidence that the largest sample measurement bounds 98% of the population values. Thus, utilizing this nonparametric approach with the limited amount of data (only 29 measurements for each impurity) available for this investigation did not lead to useful confidence statements about the quantiles of interest.

Table 2. Confidence Limits for Quantiles using a Nonparametric Approach

n (sample size)	α (there is probability of at least 1-α that at least q of the population is less than X$_{(29)}$	Confidence (1-α)	q (the proportion of the population)
29	0.01	99%	0.8532
29	0.02	98%	0.8738
29	0.03	97%	0.8861
29	0.04	96%	0.8949
29	0.05	95%	0.9019
29	0.06	94%	0.9075
29	0.07	93%	0.9124
29	0.08	92%	0.9166
29	0.09	91%	0.9203
29	0.10	90%	0.9237
29	0.11	89%	0.9267
29	0.12	88%	0.9295
29	0.13	87%	0.9321
29	0.14	86%	0.9345
29	0.15	85%	0.9367
29	0.16	84%	0.9388
29	0.17	83%	0.9407
29	0.18	82%	0.9426
29	0.19	81%	0.9443
29	0.20	80%	0.9460
29	0.21	79%	0.9476
29	0.22	78%	0.9491
29	0.23	77%	0.9506
29	0.24	76%	0.9520
29	0.25	75%	0.9533
29	0.26	74%	0.9546
29	0.27	73%	0.9559
29	0.28	72%	0.9571
29	0.29	71%	0.9582
29	0.30	70%	0.9593
29	0.31	69%	0.9604
29	0.32	68%	0.9615
29	0.33	67%	0.9625
29	0.34	66%	0.9635
29	0.35	65%	0.9644
29	0.36	64%	0.9654
29	0.37	63%	0.9663
29	0.38	62%	0.9672
29	0.39	61%	0.9681
29	0.40	60%	0.9689
29	0.41	59%	0.9697
29	0.42	58%	0.9705
29	0.43	57%	0.9713
29	0.44	56%	0.9721
29	0.45	55%	0.9728
29	0.46	54%	0.9736
29	0.47	53%	0.9743
29	0.48	52%	0.9750
29	0.49	51%	0.9757
29	0.50	50%	0.9764
29	0.51	49%	0.9770
29	0.52	48%	0.9777
29	0.53	47%	0.9783
29	0.54	46%	0.9790
29	0.55	45%	0.9796
29	0.56	44%	0.9802

The second, mathematically-equivalent, nonparametric approach considered in this investigation is that presented in [4]. For this approach the material from LANL was considered as being produced by a process, and it is the probability, p, of that process producing an unacceptable output, where an unacceptable output is one whose concentration for an impurity exceeds the maximum limit for that impurity, that was investigated. For the LANL material to be successfully processed through MOX the value of p for a given impurity must be less than 2%. This would lead to 98% of the LANL material having concentrations for the given impurity below the maximum limit for that impurity. As already noted above, all of the impurity measurements available for the LANL material were

acceptable. So, with 29 acceptable outcomes for each impurity in a random sample of 29 items what upper confidence limit on the value of p can be determined utilizing the approach presented in [4]? For this approach, an upper $(1-\alpha) \times 100\%$ confidence limit on the value of p, p_u, based upon a random sample of n=29 items with d=0 unacceptable items is given by

$$p_u = (m_1 \, F_{m_1, m_2, \alpha} \, / \, m_2)/(1+(m_1 \, F_{m_1, m_2, \alpha} \, / \, m_2))$$

where $m_1 = 2(d+1) = 2$, $m_2 = 2(n-d) = 2(29) = 58$, and $F_{m_1, m_2, \alpha}$ is the upper α-tail of the F distribution with m_1 and m_2 degrees of freedom for the numerator and denominator, respectively.

Table 3 presents the confidence limits associated with various values for p_u. And from these entries, it is apparent that the results from this nonparametric approach are mathematically equivalent to the results presented in Table 2 for the earlier nonparametric approach. For example, from the shaded row in this table, there is 90% confidence that the probability of an unacceptable item from the sampled process is bounded by 7.63% and from Table 2 there is 90% confidence that 92.37% of the population is less that the largest measurement (an acceptable measurement) seen in the random sample of 29 items.

Table 3. Confidence Intervals for Unacceptable Proportion using a Non-Parametric Approach

				Binomial-Based Approach with All Good in Random Sample		Upper Confidence Limit, p_U, for p, where p=Prob(unacceptable item))
n	d	m_1	m_2	Confidence (1-α)100%	F	Prob(p ≥ p_U) ≤ α, where p_U =
29	0	2	58	50%	0.701	0.0236
29	0	2	58	51%	0.722	0.0243
29	0	2	58	52%	0.743	0.0250
29	0	2	58	53%	0.765	0.0257
29	0	2	58	54%	0.787	0.0264
29	0	2	58	55%	0.810	0.0272
29	0	2	58	56%	0.833	0.0279
29	0	2	58	57%	0.856	0.0287
29	0	2	58	58%	0.881	0.0295
29	0	2	58	59%	0.905	0.0303
29	0	2	58	60%	0.931	0.0311
29	0	2	58	61%	0.957	0.0319
29	0	2	58	62%	0.984	0.0328
29	0	2	58	63%	1.011	0.0337
29	0	2	58	64%	1.040	0.0346
29	0	2	58	65%	1.069	0.0356
29	0	2	58	66%	1.099	0.0365
29	0	2	58	67%	1.130	0.0375
29	0	2	58	68%	1.162	0.0385
29	0	2	58	69%	1.195	0.0396
29	0	2	58	70%	1.229	0.0407
29	0	2	58	71%	1.265	0.0418
29	0	2	58	72%	1.301	0.0429
29	0	2	58	73%	1.339	0.0441
29	0	2	58	74%	1.379	0.0454
29	0	2	58	75%	1.420	0.0467
29	0	2	58	76%	1.463	0.0480
29	0	2	58	77%	1.508	0.0494
29	0	2	58	78%	1.554	0.0509
29	0	2	58	79%	1.603	0.0524
29	0	2	58	80%	1.655	0.0540
29	0	2	58	81%	1.709	0.0557
29	0	2	58	82%	1.767	0.0574
29	0	2	58	83%	1.827	0.0593
29	0	2	58	84%	1.892	0.0612
29	0	2	58	85%	1.961	0.0633
29	0	2	58	86%	2.034	0.0655
29	0	2	58	87%	2.114	0.0679
29	0	2	58	88%	2.200	0.0705
29	0	2	58	89%	2.293	0.0733
29	0	2	58	90%	2.396	0.0763
29	0	2	58	91%	2.511	0.0797
29	0	2	58	92%	2.639	0.0834
29	0	2	58	93%	2.785	0.0876
29	0	2	58	94%	2.954	0.0925
29	0	2	58	95%	3.156	0.0981
29	0	2	58	96%	3.404	0.1051
29	0	2	58	97%	3.727	0.1139
29	0	2	58	98%	4.188	0.1262
29	0	2	58	99%	4.991	0.1468

3.2 Parametric Approach

Since there were so little data above the detection limits for S, F, and Cl, only a nonparametric approach was used, and as seen in the previous discussion, while there was no immediate concern for the LANL material being suitable for processing through MOX regarding the concentrations for these three elements, there was too little data available to lead to useful confidence statements about the quantiles of interest for these three elements.

For the other elements of this study, C, P, and N, there were more measurements recorded as being above the detection limits of the analytical process, and a parametric approach was used to investigate the quantiles of their population of concentration values. The use of a parametric approach relies on employing a probability distribution to represent that population.

3.2.1 *Elements with All Measurements above their Detection Limits*

For those impurities whose measured values were all above detection (i.e., C and P), JMP and its Goodness of Fit test was used to identify candidate probability distributions. First consider the JMP results for P that are provided in Exhibit 1. These results were generated using JMP's Descriptive Statistics platform and include a histogram and summary statistics for the P measurements. The left-side of this exhibit provides these results for the measurements as presented in Table 1, while the right-side of the exhibit provides JMP results for the natural logarithm of the measurements. A normal probability distribution was fit to both sets of data. Thus, while the left-side of the exhibit considered the use of a normal probability to represent the population of P measurements of the LANL material, the right-side explored the use of the lognormal distribution for this population. For each side of the exhibit, the results for a Shapiro-Wilk goodness of fit test for the normal probability are presented. If the p-value for this test is less than 0.05, the null hypothesis of normality is rejected at a 5% significance level. The p-value for the left-side is 0.0002, and the p-value for the right-side is 0.0213. Thus, the hypothesis of the P measurements of Table 1 following a normal distribution was rejected at the 5% significance level and the hypothesis of a natural logarithm of the measurements following a normal distribution (or the original measurements following a lognormal distribution) was rejected at the 5% significance level. Thus, no discernible distribution was discovered for the P measurements, which leads to a reliance on the nonparametric results from Tables 2 and 3 for an interpretation on the bound for the population of P measurements that is provided by the largest value in the sample, 130 µg P/g Pu.

P (µg/g Pu)				ln(P)			

Normal(57.4138,38.3373)				Normal(3.84652,0.64185)			

Quantiles (P)

100.0%	maximum	130
99.5%		130
97.5%		130
90.0%		130
75.0%	quartile	89
50.0%	median	44
25.0%	quartile	29
10.0%		22
2.5%		18
0.5%		18
0.0%	minimum	18

Quantiles (ln(P))

100.0%	maximum	4.86753
99.5%		4.86753
97.5%		4.86753
90.0%		4.86753
75.0%	quartile	4.48706
50.0%	median	3.78419
25.0%	quartile	3.36491
10.0%		3.09104
2.5%		2.89037
0.5%		2.89037
0.0%	minimum	2.89037

Moments (P)

Mean	57.413793
Std Dev	38.337335
Std Err Mean	7.1190643
Upper 95% Mean	71.996535
Lower 95% Mean	42.831051
N	29

Moments (ln(P))

Mean	3.8465234
Std Dev	0.6418476
Std Err Mean	0.1191881
Upper 95% Mean	4.0906691
Lower 95% Mean	3.6023776
N	29

Fitted Normal
Parameter Estimates

Type	Parameter	Estimate	Lower 95%	Upper 95%
Location	µ	57.413793	42.831051	71.996535
Dispersion	σ	38.337335	30.423721	51.849394

-2log(Likelihood) = 292.791039546614

Fitted Normal
Parameter Estimates

Type	Parameter	Estimate	Lower 95%	Upper 95%
Location	µ	3.8465234	3.6023776	4.0906691
Dispersion	σ	0.6418476	0.509357	0.8680678

-2log(Likelihood) = 55.5809762922963

Goodness-of-Fit Test
Shapiro-Wilk W Test

W	Prob<W
0.824394	0.0002*

Note: Ho = The data is from the Normal distribution. Small p-values reject Ho.

Goodness-of-Fit Test
Shapiro-Wilk W Test

W	Prob<W
0.913826	0.0213*

Note: Ho = The log data is from the Normal distribution. or Ho = The data is from the Lognormal distribution Small p-values reject Ho.
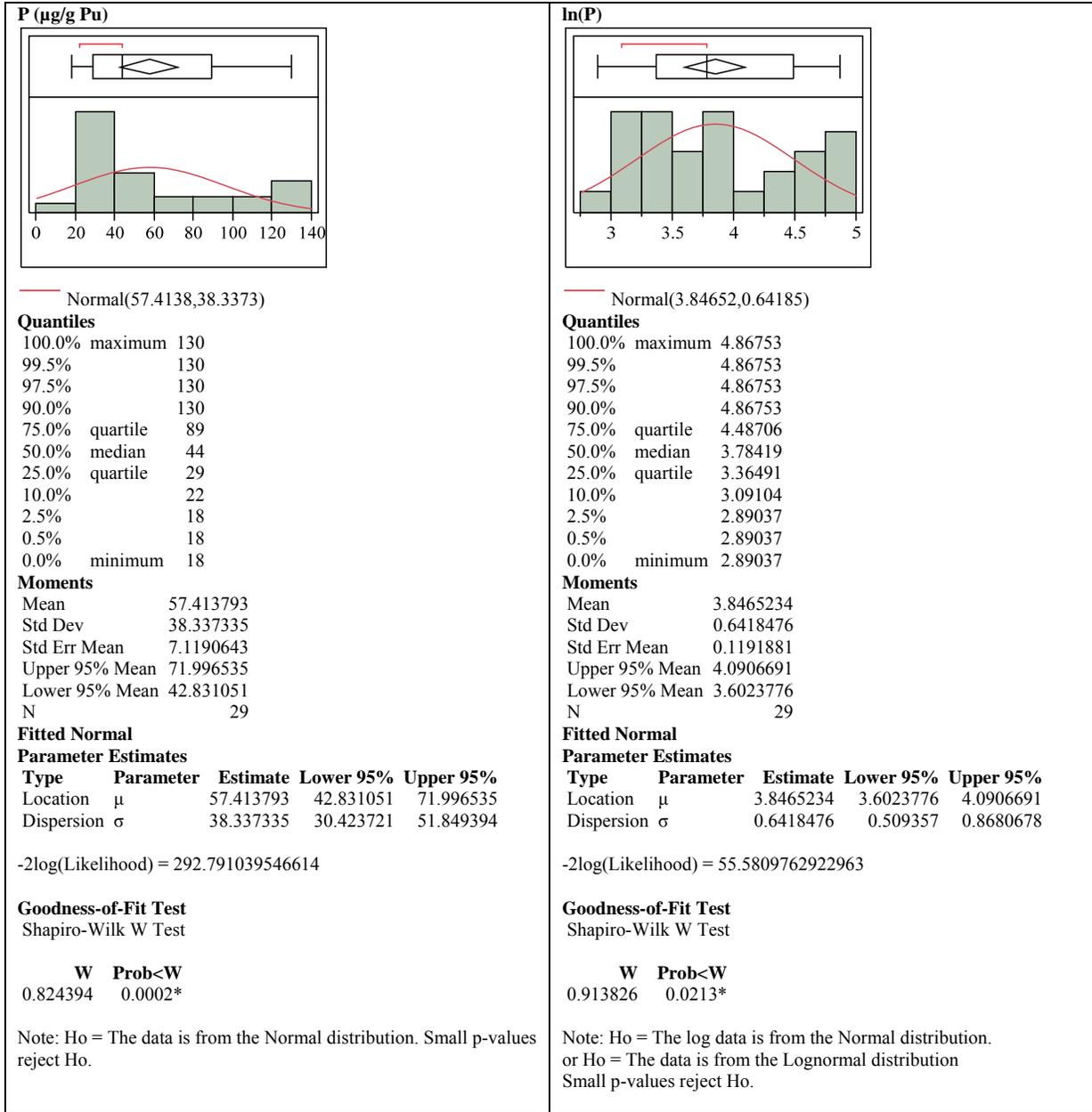
Exhibit 1. JMP Results for Candidate Probability Distributions for P Concentrations

Next consider similar results from JMP for the C data given in Exhibit 2. The left-side of this exhibit considered the use of a normal probability to represent this population while the right-side explored the use of the lognormal distribution (i.e., by fitting a normal probability distribution to the natural logarithm of the measured C values). The p-value of the Shapiro-Wilk test for the measurements was 0.0080 which leads to the rejection of the hypothesis that these measurements follow a normal probability distribution. The p-value for the natural logarithm of the measurements was 0.9981, which is greater than 0.05, and thus, the lognormal distribution was considered as a candidate distribution for the C measurements.
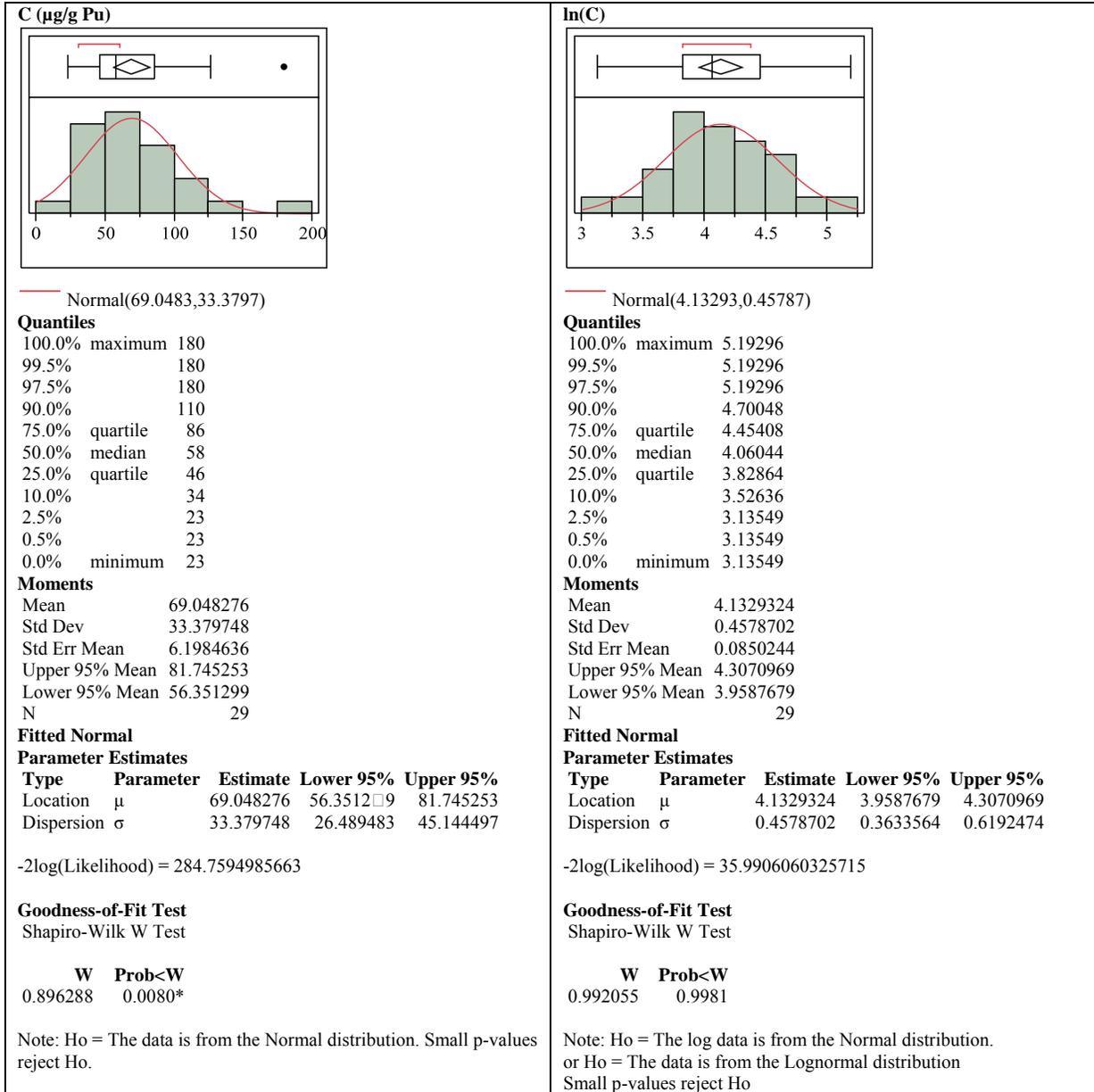
C (µg/g Pu)

Normal(69.0483,33.3797)

Quantiles

100.0%	maximum	180
99.5%		180
97.5%		180
90.0%		110
75.0%	quartile	86
50.0%	median	58
25.0%	quartile	46
10.0%		34
2.5%		23
0.5%		23
0.0%	minimum	23

Moments

Mean	69.048276
Std Dev	33.379748
Std Err Mean	6.1984636
Upper 95% Mean	81.745253
Lower 95% Mean	56.351299
N	29

Fitted Normal

Parameter Estimates

Type	Parameter	Estimate	Lower 95%	Upper 95%
Location	µ	69.048276	56.3512 9	81.745253
Dispersion	σ	33.379748	26.489483	45.144497

-2log(Likelihood) = 284.7594985663

Goodness-of-Fit Test

Shapiro-Wilk W Test

W	Prob<W
0.896288	0.0080*

Note: Ho = The data is from the Normal distribution. Small p-values reject Ho.

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ln(C)

Normal(4.13293,0.45787)

Quantiles

100.0%	maximum	5.19296
99.5%		5.19296
97.5%		5.19296
90.0%		4.70048
75.0%	quartile	4.45408
50.0%	median	4.06044
25.0%	quartile	3.82864
10.0%		3.52636
2.5%		3.13549
0.5%		3.13549
0.0%	minimum	3.13549

Moments

Mean	4.1329324
Std Dev	0.4578702
Std Err Mean	0.0850244
Upper 95% Mean	4.3070969
Lower 95% Mean	3.9587679
N	29

Fitted Normal

Parameter Estimates

Type	Parameter	Estimate	Lower 95%	Upper 95%
Location	µ	4.1329324	3.9587679	4.3070969
Dispersion	σ	0.4578702	0.3633564	0.6192474

-2log(Likelihood) = 35.9906060325715

Goodness-of-Fit Test

Shapiro-Wilk W Test

W	Prob<W
0.992055	0.9981

Note: Ho = The log data is from the Normal distribution.
or Ho = The data is from the Lognormal distribution
Small p-values reject Ho

Exhibit 2. JMP Results for Candidate Probability Distributions for C Concentrations

Once a candidate probability distribution was identified, it was used to bound quantiles of the distribution with the desired confidence. Specifically, from [3], if a population is normally distributed with unknown mean and variance and if a random sample of n observations from this distribution is taken, then the sample mean, $\bar{x}$, and the sample variance, s^2, may be computed and used to determine an upper $100(1-\alpha)$% tolerance bound for the distribution of values with $100\ (1-\gamma)$% confidence as follows:

$$\bar{x} + k \cdot s$$

where the equation for k involves the noncentral Student t distribution and is given by:

$$k = \frac{t\left(\gamma, n-1, \sqrt{n} \cdot z_{1-\alpha}\right)}{\sqrt{n}}$$

where $t(\cdot)$ represents the upper-γ tail of the noncentral Student t distribution with n-1 degrees of freedom and noncentrality parameter given by $\sqrt{n} \cdot z_{1-\alpha}$, where $z_{1-\alpha}$ is the (1-α)100%-tile of the standard normal distribution. The noncentral Student t values were computed using JMP and were used to prepare Table 4, which provides an Upper Tolerance Limit (UTL) for 98% of the each of the populations considered in this section with 95% confidence. For the natural logarithm, ln, values of for the C measurements, the sample mean, sample standard, and UTL were computed for the log values, and then the UTL results were converted back to the original units of µg/g Pu by exponentiation. The last column of Table 4 provides the maximum limit for C. Thus, the results for C indicated no issue in the LANL material meeting the maximum limit for this element assuming that the measurements for this element follow a lognormal distribution.

**Table 4. Upper Tolerance Limits for the 98th Percentiles
for Specified Probability Distributions**

Impurity	n	average	standard deviation	confidence	coverage	k	UTL (µg/g Pu)	Maximum Limit (µg/g Pu)
ln(C)	29	4.13293	0.45787	0.95	0.98	2.73947	219	500

The 99.9[th] quantile is the exceptional limit; it is a value in the very upper-tail of the population; and as such, it is very sensitive to the assumption of the probability distribution used to represent the concentration values. With that caution understood, Table 5 provides an estimate expressed in µg/g P for the 99.9[th] quantile of the C distribution. The last column of Table 5 provides the exceptional limit for C. The results from this table indicate no issue regarding the LANL material satisfying the exceptional limits for C if the C measurements follow a lognormal.

**Table 5. Upper Tolerance Limits for the 99.9[th] Quantiles
for Specified Probability Distributions**

Impurity	n	average	standard deviation	confidence	coverage	k	UTL (µg/g Pu)	Exceptional Limit (µg/g Pu)
ln(C)	29	4.13293	0.45787	0.95	0.999	4.04318	397	1500

3.2.2 *Elements with Measurements above and below their Detection Limits*

As noted above, the sample results for N indicated no issue in the LANL material meeting the maximum and exceptional limits for MOX. It was also noted that for the nonparametic approach applied to the N measurements there was too little data available to lead to confident statements about the quantiles of interest for N. However, since there are 12 below detect values for N recorded in the measurements of Table 1, the JMP software could not be used to support a parametric analysis. One of the features of the ProUCL software package is that it handles data sets containing below detection results. Exhibit 3 provides the results from fitting a normal and a lognormal distribution to the N measurement data (which contains 12 below detection values) using ProUCL. The Shapiro-Wilk was

used to test for normality and for lognormality, and the p-values for both of these tests indicated that the hypotheses of normality and of lognormality are both rejected at the 5% significance level. Thus, neither of these proposed distributions is suitable for modeling the population of N concentration measurements. So, as in the case for P above, no discernible distribution was discovered for the N measurements, which leads to a reliance on the nonparametric results from Tables 2 and 3 for an interpretation on the bound for the population of N measurements that is provided by the largest value in the sample, 390 µg N/g Pu.

N

General Statistics

Number of Valid Data	29	Number of Detected Data	17
Number of Distinct Detected Data	15	Number of Non-Detect Data	12
Tolerance Factor	2.232	Percent Non-Detects	41.38%

Raw Statistics		Log-transformed Statistics	
Minimum Detected	14.9	Minimum Detected	2.701
Maximum Detected	390	Maximum Detected	5.966
Mean of Detected	76.23	Mean of Detected	3.646
SD of Detected	113.4	SD of Detected	1.06
Minimum Non-Detect	11	Minimum Non-Detect	2.398
Maximum Non-Detect	35	Maximum Non-Detect	3.555

Data with Multiple Detection Limits		Single Detection Limit Scenario	
Note: Data have multiple DLs - Use of KM Method is recommended		Number treated as Non-Detect with Single DL	22
For all methods (except KM, DL/2, and ROS Methods),		Number treated as Detected with Single DL	7
Observations < Largest ND are treated as NDs		Single DL Non-Detect Percentage	75.86%

Background Statistics

Normal Distribution Test with Detected Values Only		Lognormal Distribution Test with Detected Values Only	
Shapiro Wilk Test Statistic	0.583	Shapiro Wilk Test Statistic	0.791
5% Shapiro Wilk Critical Value	0.892	5% Shapiro Wilk Critical Value	0.892
Data not Normal at 5% Significance Level		**Data not Lognormal at 5% Significance Level**	

Assuming Normal Distribution		Assuming Lognormal Distribution	
DL/2 Substitution Method		DL/2 Substitution Method	
Mean	48.94	Mean (Log Scale)	3.048
SD	91.96	SD (Log Scale)	1.13
95% UTL 95% Coverage	254.2	95% UTL 95% Coverage	262.5
95% UPL (t)	208.1	95% UPL (t)	148.9
90% Percentile (z)	166.8	90% Percentile (z)	89.67
95% Percentile (z)	200.2	95% Percentile (z)	135.2
99% Percentile (z)	262.9	99% Percentile (z)	292
Maximum Likelihood Estimate(MLE) Method		Log ROS Method	
Mean	-134	Mean in Original Scale	47.51
SD	231.6	SD in Original Scale	92.59
95% UTL with 95% Coverage	382.8	95% UTL with 95% Coverage	334.5
		95% BCA UTL with 95% Coverage	390
		95% Bootstrap (%) UTL with 95% Coverage	390
95% UPL (t)	266.6	95% UPL (t)	171.6
90% Percentile (z)	162.7	90% Percentile (z)	94.42
95% Percentile (z)	246.9	95% Percentile (z)	153.1
99% Percentile (z)	404.7	99% Percentile (z)	379.3

Exhibit 3. ProUCL Results for N

4.0 SUMMARY AND CONCLUSIONS

The measurements evaluated as part of this study included LANL blend lots 1 through 29 and cover C, Cl, F, N, P, and S. The LANL material is being considered for processing through MOX at SRS. There are specification limits for impurities in the feed to the MOX facility: a maximum limit and an exceptional limit. All of the sample measurements were below these limits, so there was no immediate concern regarding the LANL material being suitable for processing through MOX. A nonparametric approach was used to investigate the population of impurity levels of the LANL material. While the results from this approach did not indicate any problems for any of the impurities, there was too little data available to lead to useful confidence statements about satisfying the maximum and exceptional limits. Impurity data from additional random samples of the LANL material would be needed to increase the confidence level associated with the results from a nonparametric approach for investigating the population quantiles of interest.

For S, F, and Cl, only a nonparametric approach was used. A second approach, a parametric approach, was attempted for C, P, and N. However, the results for the P and N measurements indicated that each of their respective populations was not well modeled by a normal or by a lognormal distribution. Thus, the conclusions for the P and N populations were those provided by the nonparametric approach. From the parametric approach, the results for C indicated no issue in the LANL material meeting the maximum limit for this element assuming that the measurements for this element follow a lognormal distribution.

5.0 REFERENCES

[1]   JMP Version 9.0.0, SAS Institute, Inc., Cary NC, 2010.

[2]   Singh, A, R Maichle, and N Armbya, "ProUCL Version 4.1 User Guide (Draft): Statistical Software for Data Sets with and without Nondetect Observations," EPA/600/R-07/041, US EPA, Office of Research and Development, Washington DC, May 2010.

[3]   Krishnamoorthy, K and T Mathew, **Statistical Tolerance Regions: Theory, Applications, and Computation**, John Wiley & Sons, Inc., Hoboken, New Jersey, 2009.

[4]   Satterthwaite, FE, "Binomial and Poisson Confidence Limits," Industrial Quality Control, Volume 13, Number 11, pp 56-59, May, 1957.

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Distribution:

Name:	Location:
J.S. Allender	703-H
T.B. Edwards	999-W
P.L. Lee	703-41A
E. N. Moore	707-24B
M.G. Schwenker	706-8F
E.P. Shine	703-41A