Los Alamos
NATIONAL LABORATORY
EST. 1943
NWIS-TP
Mail Stop J594, Los Alamos, NM 87545

TRU-TD-0301

Determination of Plutonium Mass Fractions and Uncertainties from Waste Assay Measurements

Effective Date: 11/23/2005

Controlled Distribution Date:

Next Review Date:

Process Owner	Signature	Date
J. Veilleux		10/12/05

Team Leader		
P. Carson		10/17/05

Group Leader or Facility Manager		
D. Christensen		11/18/05

YOU ARE RESPONSIBLE FOR VERIFYING THAT YOU ARE WORKING TO THE MOST CURRENT REVISION OF THIS DOCUMENT.

LA-UR-05-8922
November 23, 2005

Determination of Plutonium Mass Fractions and Uncertainties from Waste Assay Measurements

John M. Veilleux
Los Alamos National Laboratory
Los Alamos, NM 87545, USA 505/667-7434
Email: veilleux@lanl.gov

Abstract

Gamma ray isotopic measurements of 3297 Los Alamos National Laboratory 55 gallon type nuclear waste drums from June 25, 1999 through April 26, 2005 have been evaluated to determine Department of Energy plutonium material type mass fractions and their uncertainties. These drums were primarily debris matrices. Previous estimates only included mass fractions while uncertainties were not available to propagate the total measurement uncertainty for non-destructive assay (NDA) equipment. The data was decay corrected from the measurement date to a common date and the number distributions evaluated at the 90th percentile. The mass fraction of each plutonium isotope and americium 241 are compared to original estimates. The data was found to be sufficient to quantify the plutonium isotopic mass fractions for a subset of weapons grade waste, Pu242 enriched plutonium waste, and heat source plutonium waste. For the other plutonium material types or wastes that are mixtures of material types, a composite material type is recommended.

1. Introduction

The nondestructive assay (NDA) characterization process for transuranic (TRU) waste at the Los Alamos National Laboratory (LANL) occasionally requires the use of acceptable knowledge (AK) isotope mass fractions. The isotopic mass fractions are needed with passive and active neutron systems and certain gamma systems to quantify the total plutonium content in terms of activity, mass, power, fissile gram equivalent (FGE), and plutonium equivalent activity (PECi).

The original memorandum[1] describing the AK isotopic mass fractions estimated the plutonium and Am241 mass fractions by material type (MT) but the data did not include any estimate of the uncertainty of the mass fractions. The material type is used to describe the isotopic composition of common blends of radioactive materials used within the DOE complex. The most common material types present in LANL TRU waste are weapons-grade plutonium (MT-51 and -52); fuel-grade plutonium (MT-53 and -54); reactor-grade plutonium (MT-55 through -57); enriched Pu-242 (MT-42); and heat-source plutonium (MT-83). Other material types include Pu with various enrichments of uranium.

Based on the MT identification of isotopic mass fractions described above, AK isotopic files[2] were generated for use with NDA instruments with an arbitrary 10% relative uncertainty for all isotopes. This uncertainty was combined with other measurement uncertainties in order to propagate the total measurement uncertainty for the assay. During the review process of CCP-TP-125[3], the Carlsbad Field Office (CBFO) enquired as to the source of the relative uncertainty. Since the 10% relative uncertainty was arbitrary, we decided to use FRAM[a] gamma isotopic measurements taken between

[a] FRAM (Fixed energy Response function Analysis with Multiple efficiencies) consists of a high purity germanium detector with associated spectrometer, acquisition software, and analysis software.

1999 and 2005 as a source of data to estimate those uncertainties. Although the FRAM software[4] quantifies isotopic ratios for several isotopes in addition to plutonium, only the plutonium and Am241 data were evaluated in this study.

2. Methodology

2.1. FRAM System

The FRAM system consists of a high efficiency germanium detector and spectrometer with associated acquisition and analysis software. After the spectrum is acquired, it is evaluated with the PC/FRAM analysis software which generates an ASCII results file. These data have been used in a WIPP-certified program and data from 1999 to the present are available. Each of nearly 4000 ASCII files were read into a summary table for subsequent evaluation. The data consists of the mass fractions and uncertainties for each of the following isotopes: Pu238 through Pu242, Am241, Am243, Np237, Ac227, Cs137, Ce144, Pa231, U233 through U235, U238, Th230, Th232, and Cm244. However, only the Pu isotopes and Am241 will be evaluated in this study. In all, there were 3952 assays in this group, including replicates.

2.2. Waste Characterization

The waste drums were from the Los Alamos National Laboratory and stored in 55-gallon drums that included 99.2% heterogeneous (debris) type waste. The remaining drums (21) were organic and inorganic sludge matrices and seven were from unknown matrices.

2.3. Data Processing

Subsequently, the replicates were removed since we did not wish to bias the results with multiple evaluations of the same drum. Also removed were replicate evaluations from MT42 where two assay results are normally included, one in which the Pu242 is determined by correlation and one where the Pu242 is determined based on an AK input. The Pu242 assay by correlation was retained. As a result of this filtering process, 3297 assays remained. This original FRAM data is included in appendix A.

The material type database by drum was provided from AK Documentation for the current Central Characterization Project (CCP) LANL Waste Isolation Pilot Plant (WIPP) assays and from the former LANL project 2010 concert database (via the CCP AK Documentation). If conflicting data was present, then the CCP AK MT documentation took precedence over the project 2010 data. There remained some unspecified material types for 65 drums and no attempt was made to further identify the material types for these drums.

Data integration was accomplished next. In this phase, the FRAM isotopic data was integrated with the material type database and the isotopic data was decay corrected to a common reference date, June 1, 2005. Decay correction for the plutonium isotopes and Am241 may be expressed as shown in the following equations[5] using the definitions listed in Table 1.

For the plutonium isotopes, with j = Pu238, Pu239, … Pu242, the mass fraction at the reference date is given by:

$$f_j = \frac{f_j^0 e^{-\lambda_j \Delta t}}{\left(m / m^0 \right)} \qquad (1)$$

For Am241, the mass fraction at the reference date is given by:

$$f_{Am241} = \frac{m_{Am241}}{m} = b\frac{M_{Am241}}{M_{Pu241}}\left(\frac{t_{Am241}}{t_{Pu241} - t_{Am241}}\right)\left(\frac{f^0_{Pu241}}{m/m^0}\right)\left[e^{-\lambda_{Pu241}\Delta t} - e^{-\lambda_{Am241}\Delta t}\right] + \left(\frac{f^0_{Am241}}{m/m^0}\right)e^{-\lambda_{Am241}\Delta t} \quad (2)$$

In the above equations, the ratio of the Pu mass on the reference date to the mass at the time of the assay is given by:

$\frac{m}{m^0} = \sum_j f_j^0 e^{-\lambda_j \Delta t}$ The sum is over the plutonium isotopes only.

The decay corrected FRAM data, including the material type assignments, are shown in appendix A.

Table 1. Definition of terms

Symbol	Description
f_j	Mass fraction of isotope j to total plutonium at the reference date.
0 superscript	Refers to the value of the specified quantity on the measurement date.
no superscript	Refers to the value of the specified quantity on the reference date.
$\lambda_j \equiv \frac{\ln(2)}{t_j}$	The decay constant of isotope j, which is equal to the natural logarithm of 2 divided by the half life of the isotope, t_j.
t_j	The half-life of isotope j
Δt	The elapsed time between the measurement date and the reference date.
m	The total plutonium mass on the reference date
M_{Am241}	The molecular weight of Am241, 241.056822 g/mole
M_{Pu241}	The molecular weight of Pu241, 241.056884 g/mole
b	The branching ratio of Pu241 to Am241 (0.999977)

2.4. Data Evaluation

Data evaluation consists of calculating the number of assays (count) and the mass fraction of the mean, the standard deviation, the relative standard deviation (%RSD), and the median. The data were evaluated by material type and for the mixtures combined. The median is useful in performing an alternate evaluation (see below) because it represents the mass fraction in the middle of a set of assays; that is, half the assays have mass fractions that are greater than the median, and half have values that are less. Next the frequency number distribution of the mass fraction for each isotope is evaluated with bin size of 0.01. The bin size was expanded to 0.0001 mass fractions for isotopes with mass fractions in that range.

Where enough assays are available for a given material type (e.g., more than 15), the mean and relative standard deviations were calculated from the distribution that included 90% of all assays centered at the median. This effectively removed large outlier measurements from the calculation which inappropriately skewed the number distribution. In practice, this means that when a drum was identified as being a member of a unique material type, there were occasions when it likely contained a mixture even though it was identified as unique. This approach reduced those occasions where the mean and standard deviations were significantly affected by this type of error. This approach was also influenced by the Student-t distribution[6] where the 90th percentile indicates that 5% of the assays should fall in the high and low tails of the distribution. In the data evaluation, whether the mean and

relative standard deviation is based on the 90th percentile or the entire distribution is indicated by 90% or 100% respectively.

The distribution parameters are shown in tables for each material type evaluated. The table includes a row depicting the raw decay corrected data from appendix A summarizing the mean, the relative standard deviation (%RSD), the median, the maximum value and the minimum value. Next, the original material type mass fractions are shown (where specified). The next row shows the evaluated data that includes the upper and lower mass fractions of the 90th percentile, the evaluated mass fraction of the mean (f) and the %RSD and whether it was based on all the data (100%) or the 90th percentile (90%). Finally, the last row of the data shows the normalized mass fraction (f) and the %RSD.

3. Results

3.1. Number of Assays by Material Type

A total of 3297 assays are included in the database spanning the range from June 25, 1999 through April 26, 2005, as summarized in Table 2. The number of unique assays is shown, e.g., the number of assays containing only MT12 is zero. However, there were 10 assays in which MT12 appeared with one or more other material type (e.g., a mixture). Finally, the description[7] of the MT is included.

Table 2. Material Type Description

MT	Unique Occurrences	Occurrences in a Mixture	Description (Source DOE 5633.5, 5/22/87)
12	0	10	Depleted Uranium
23	0	1	Enriched U235 1.15 to 1.60%
24	1	4	Enriched U235 1.60 to 2.00%
31	0	1	Enriched U235 4.1-5%
32	0	3	Enriched U235 5 to 10%
33	0	4	Enriched U235 10-20%
34	0	2	Enriched U235 20-35%
35	0	5	Enriched U235 35-45%
36	1	7	Enriched U235 45-80%
37	0	4	Enriched U235 80-92%
38	0	25	Enriched U235 92-94%
39	0	20	Enriched U235 94% up
41	0	1	Pu242 Enriched 20-60%
42	69	76	Pu242 Enriched above 60%
44	0	6	Am241
51	0	56	Enriched Pu240 < 4%
52	2076	2681	Enriched Pu240 4-7%
53	7	163	Enriched Pu240 7-10%
54	210	584	Enriched Pu240 10-13%
55	1	12	Enriched Pu240 13-16%
56	19	113	Enriched Pu240 16-19%
57	1	4	Enriched Pu240 > 19%
62	0	1	Enriched Li6 55-80%
63	1	1	Enriched Li6 >80%
81	0	1	Uranium (normal)
82	1	4	Np237 Enriched
83	204	256	Pu238 Enriched
88	0	3	Thorium enriched
Mixtures	705	NA	Mixed U, mixed Pu, or both
TOTALS	3297		

For determining the isotopic composition of the Pu types, only assays containing unique material types are included. From this data, all the Uranium MTs have inadequate number of assays to derive mass fraction data. Only MT42, 52, 53, 54, 56, and 83 provide sufficient data to determine the mass fractions. The mixtures combine various material types and have therefore been omitted from the mass fraction calculations. For Pu material types that cannot be determined (MT 51, 55, 57, and mixtures), an average for all MTs that excludes MT83 is proposed.

3.2. MT42

There were a total of 69 assays that were reportedly unique material type 42 and the assessment of the data are shown in Table 3. The "Raw Data" includes the mean, the relative standard deviation (%), the median, and the maximum and minimum values of the mass fraction distribution derived from all 69 assays. Next, the original material type mass fraction (see reference 1) is summarized. As already noted, there is no specified uncertainty associated with these values. The evaluated data includes the determination of the upper and lower bounds of the mass fraction which yield 45% of the assays above and below the median (i.e., in total, this is 90% of the assays). If the 90th percentile is to be used for the evaluated mass fraction, then the "Base On" term is given as 90%. Otherwise, the entire distribution is used and specified as "100%". The normalized mass fractions along with the %RSD are the recommended values.

Table 3. MT42 Mass Fractions and Uncertainties

Data	# Assays	Description	Pu238	Pu239	Pu240	Pu241	Pu242	Am241
Raw Data From Appendix B	69	Mean	2.66E-02	3.67E-01	2.72E-01	3.82E-02	2.95E-01	8.23E-02
		%RSD	60.9%	70.5%	58.0%	62.3%	114.1%	88.0%
		Median	2.82E-02	3.24E-01	2.96E-01	4.21E-02	1.39E-01	6.94E-02
		Max	6.86E-02	9.33E-01	6.59E-01	1.04E-01	9.74E-01	3.46E-01
		Min	6.35E-04	6.46E-03	1.06E-02	1.33E-03	6.27E-05	1.56E-03
Original MT Info	NA	Original f	7.30E-03	1.06E-02	6.40E-02	1.97E-02	8.98E-01	3.77E-02
		%RSD	Not Specified					
Evaluated Data	69	f_Upper	6.10E-02	8.21E-01	6.20E-01	9.25E-02	9.10E-01	2.00E-01
		f_Lower	1.70E-03	2.00E-02	2.85E-02	3.00E-03	1.00E-04	4.50E-03
		Base On	90%	90%	90%	90%	90%	90%
		f	2.63E-02	3.51E-01	2.61E-01	3.77E-02	2.63E-01	7.22E-02
		%RSD	50.2%	63.8%	48.0%	51.2%	115.9%	72.2%
Normalized	69	f	2.80E-02	3.74E-01	2.78E-01	4.01E-02	2.80E-01	7.69E-02
		%RSD	50.2%	63.8%	48.0%	51.2%	115.9%	72.2%

The mass fraction number distributions for each radioisotope for MT42 provides information on how these measurements were distributed as demonstrated in Figure 1. These data are useful in determining the upper and lower mass fraction for the 90th percentile. Applying the 90th percentile reduces to total relative error (%RSD) by approximately 10% for all cases except Pu242. Consequently, the 90th percentile values were used to determine the mean and %RSD for all isotopes.

Gamma systems cannot directly measure the Pu242 isotope but rely on correlations involving the Pu239, Pu240 and Pu241 which can be readily measured by their gamma ray signatures. The large variations in the Pu242 mass fractions (which span nearly 0 to 1) are due to similar variations in one or more of the isotopes used in the correlation.

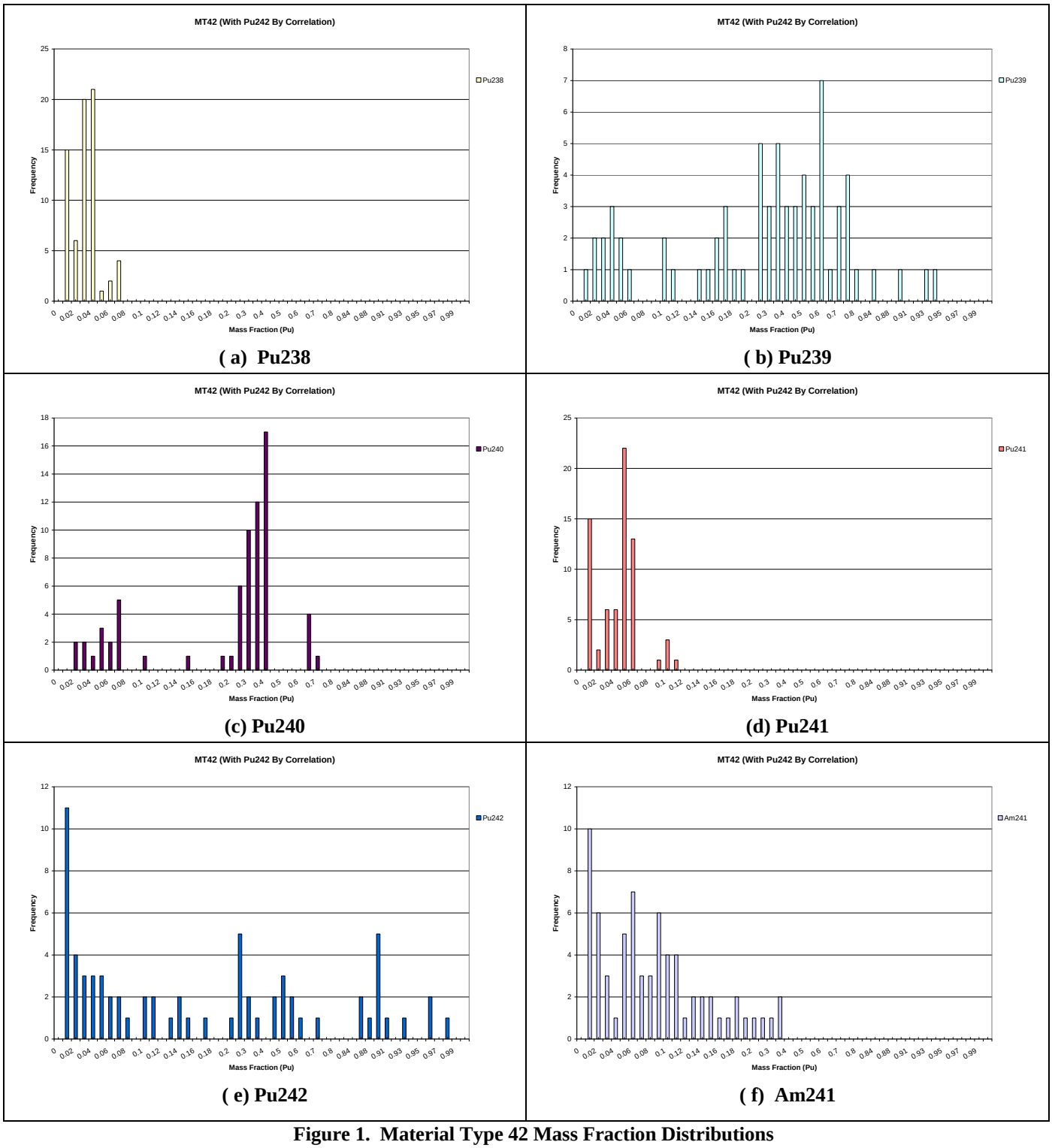

Figure 1. Material Type 42 Mass Fraction Distributions

NOTE: The scale on each figure is not necessarily linear—the distribution bin sizes were adjusted to best portray the distribution. Bin sizes range from 0.0001 in some cases to 0.1 in others.

Figure 2 compares the original and normalized mass fractions and their related uncertainties (1σ) for each of the MT42 isotopes. The mass fractions of all the isotopes except Pu242 are larger than originally reported with Pu239 showing the largest difference. The Pu242 mass fraction uncertainty spans the range from nearly zero to 60% (mass fraction expressed as a percent) reflecting the large variation indicated for Pu239 and Pu240 depicted in the frequency number distribution of Figure 1.

The large variation indicated for Pu242 suggests that most MT42 drums contain mixtures of waste and not just highly enriched Pu242.

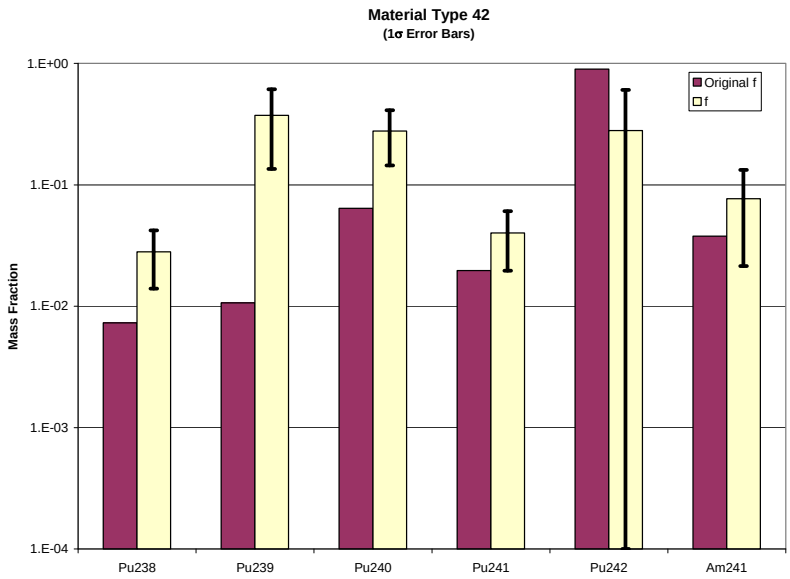

Figure 2. MT42 Comparison of Original and Evaluated Mass Fractions and Their Uncertainties

3.3. MT52

There were 2076 assays for MT52 that were reported as uniquely MT52. The statistical results for this material type are summarized in Table 4. The raw data gives the statistical information for all 2076 assays decay corrected to June 1, 2005. The normalized mass fractions along with the %RSD are the recommended values.

The frequency distribution (Figure 3) shows that the isotopic mass fractions of each isotope are relatively well grouped about the median, with some very large outliers. For example, the median mass fraction for Pu238 is 0.0148% with outliers as large as 97%. Pu241 and Am241 have similar large outliers and these outliers drive the %RSD to very large values (e.g., 1692% for Pu238). Applying the 90th percentile to these data dramatically reduces the %RSD for all isotopes, as shown in the evaluated data.

The outliers appear to be caused by mixtures in the waste. Although the known mixtures were eliminated, we surmise that reportedly pure MT52 often contain mixtures and these are correctly eliminated by using the 90th percentile.

Comparing the original mass fractions with the normalized values and their uncertainties is summarized in Figure 4. The comparison of the former and recommended mass fractions is in close agreement. Pu239 has an extremely small uncertainty (about 1%) while the largest variation is that for Am241 at 84.7%.

Table 4. MT52 Mass Fractions and Uncertainties

Data	# Assays	Description	Pu238	Pu239	Pu240	Pu241	Pu242	Am241
Raw Data From Appendix B	2076	Mean	1.19E-03	9.30E-01	6.72E-02	1.54E-03	1.86E-04	1.04E-02
		%RSD	1692.0%	3.4%	35.3%	85.3%	606.6%	285.2%
		Median	1.48E-04	9.36E-01	6.24E-02	1.34E-03	1.08E-04	4.33E-03
		Max	8.43E-01	9.97E-01	3.78E-01	3.11E-02	4.49E-02	5.38E-01
		Min	0.00E+00	1.45E-01	0.00E+00	2.72E-06	2.00E-07	2.73E-04
Original MT Info	NA	Original f	1.00E-04	9.38E-01	6.00E-02	2.00E-03	2.00E-04	1.59E-03
		%RSD	Not Specified					
Evaluated Data	2076	f_Upper	1.10E-03	9.55E-01	1.04E-01	2.56E-03	1.45E-04	3.60E-02
		f_Lower	5.50E-05	8.94E-01	4.50E-02	9.60E-04	0.00E+00	2.00E-03
		Base On	90%	90%	90%	90%	90%	90%
		f	2.05E-04	9.34E-01	6.46E-02	1.42E-03	9.93E-05	6.09E-03
		%RSD	78.9%	1.0%	14.9%	19.8%	22.5%	84.7%
Normalized	2076	f	2.05E-04	9.34E-01	6.46E-02	1.42E-03	9.93E-05	6.09E-03
		%RSD	78.9%	1.0%	14.9%	19.8%	22.5%	84.7%

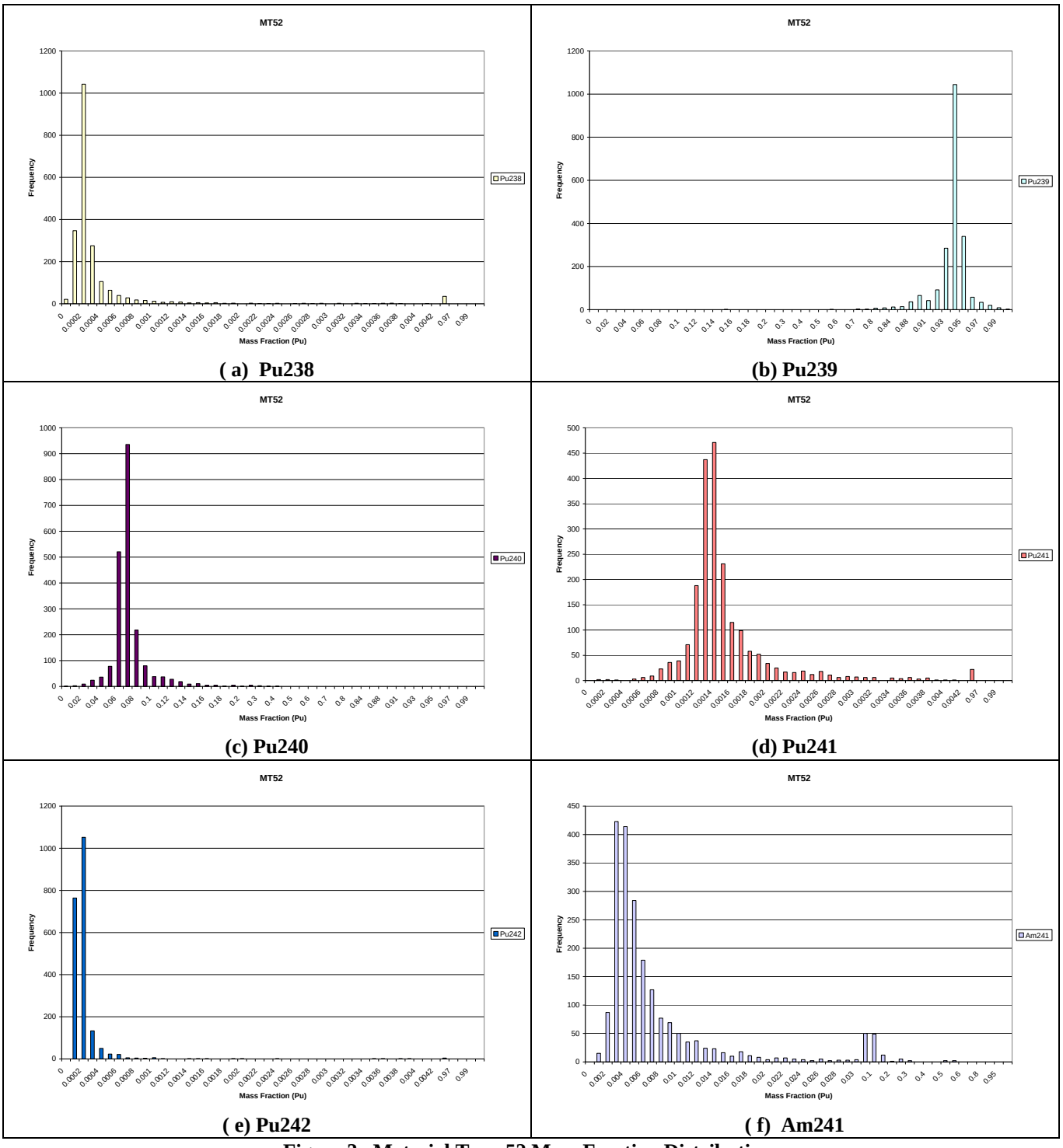

Figure 3. Material Type 52 Mass Fraction Distributions

NOTE: The scale on each figure is not necessarily linear—the distribution bin sizes were adjusted to best portray the distribution. Bin sizes range from 0.0001 in some cases to 0.1 in others.

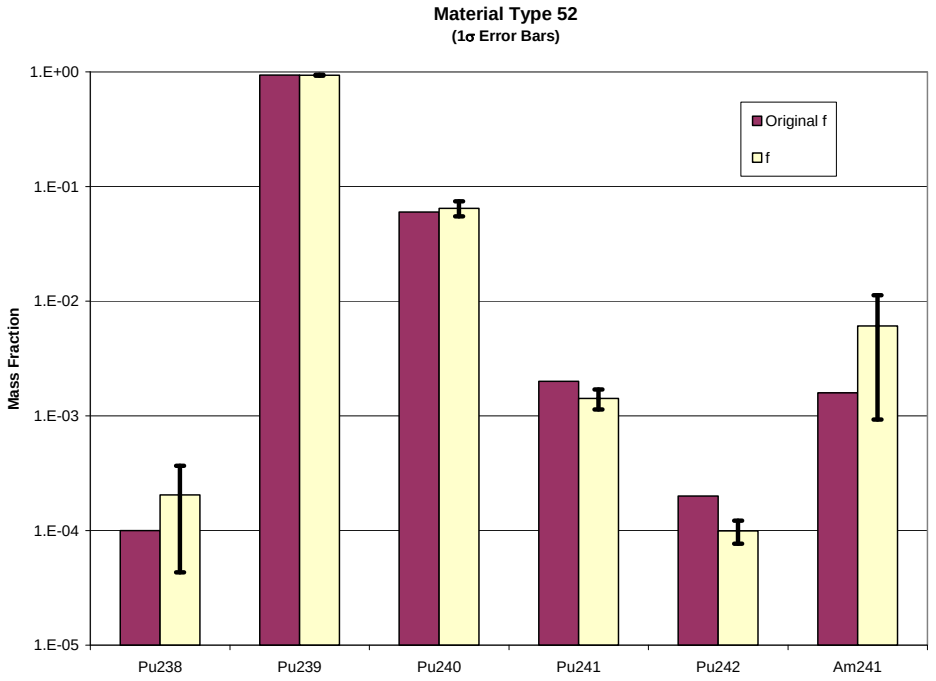

Figure 4. MT52 Comparison of Original and Evaluated Mass Fractions and Their Uncertainties

3.4. MT53

There were only 7 assays that were reported as containing uniquely MT53. Table 5 summarizes the statistics of the distribution and how they were evaluated. In contrast to the other material types, all the assays (100%) were used to generate the evaluated and normalized data. The normalized mass fractions along with the %RSD are the recommended values.

Table 5. MT53 Mass Fractions and Uncertainties

Data	# Assays	Description	Pu238	Pu239	Pu240	Pu241	Pu242	Am241
Raw Data From Appendix B	7	Mean	2.26E-04	9.10E-01	8.79E-02	2.06E-03	2.52E-04	7.78E-03
		%RSD	37.9%	2.5%	24.5%	41.5%	68.1%	52.7%
		Median	1.96E-04	9.17E-01	8.11E-02	1.57E-03	1.53E-04	6.68E-03
		Max	3.43E-04	9.42E-01	1.18E-01	3.32E-03	5.13E-04	1.37E-02
		Min	1.28E-04	8.78E-01	5.67E-02	1.21E-03	8.59E-05	3.00E-03
Original MT Info	NA	Original f	3.00E-04	9.11E-01	8.45E-02	3.66E-03	7.10E-04	2.92E-03
		%RSD	Not Specified					
Evaluated Data	7	f_Upper	NA	NA	NA	NA	NA	NA
		f_Lower	NA	NA	NA	NA	NA	NA
		Base On	100%	100%	100%	100%	100%	100%
		f	2.26E-04	9.10E-01	8.79E-02	2.06E-03	2.52E-04	7.78E-03
		%RSD	37.9%	2.5%	24.5%	41.5%	68.1%	52.7%
Normalized	7	f	2.26E-04	9.10E-01	8.79E-02	2.06E-03	2.52E-04	7.78E-03
		%RSD	37.9%	2.5%	24.5%	41.5%	68.1%	52.7%

The reported isotopic values were reasonably well distributed (Figure 5) in spite of the few assays in the distributions.

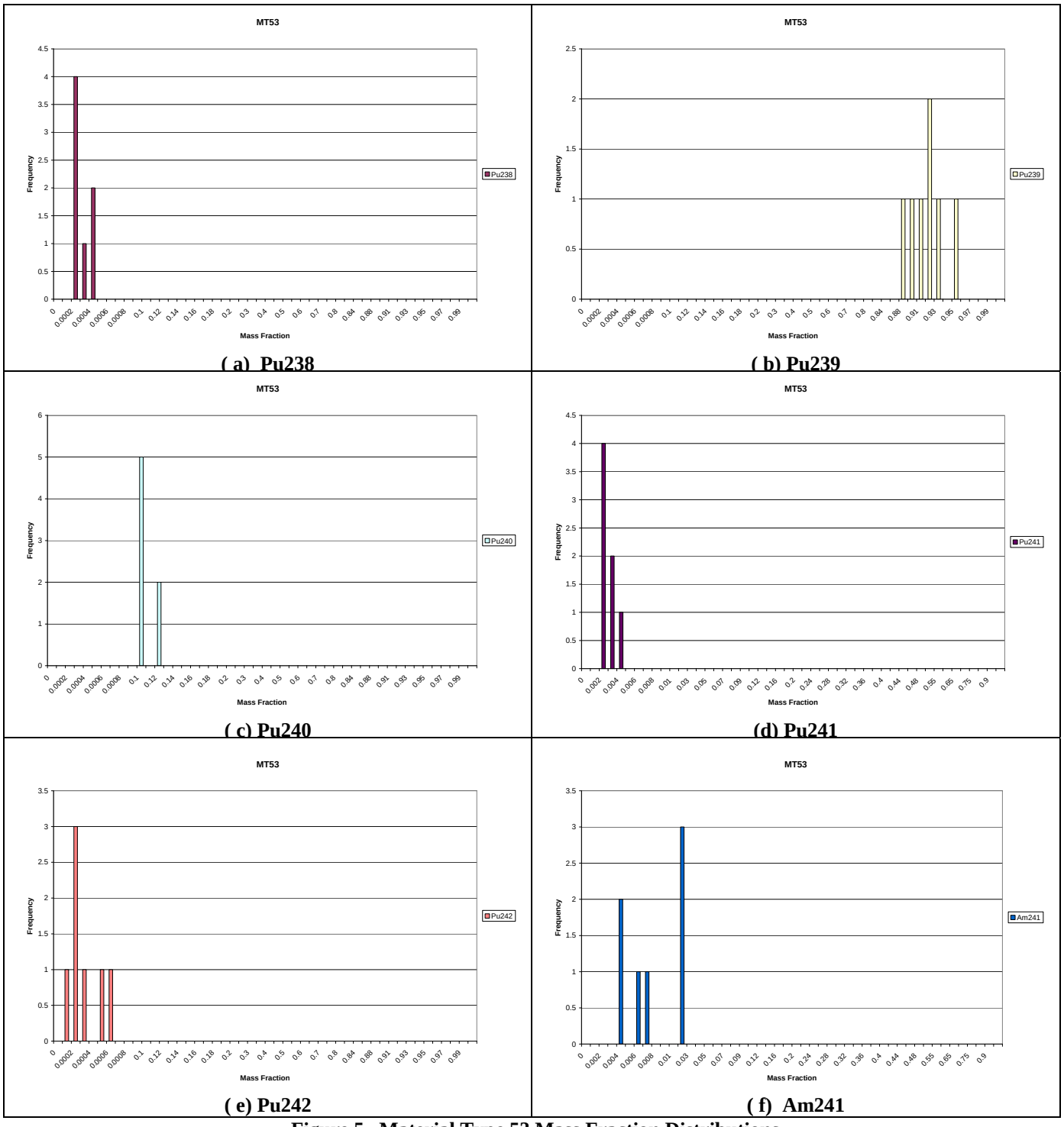

Figure 5. Material Type 53 Mass Fraction Distributions

NOTE: The scale on each figure is not necessarily linear—the distribution bin sizes were adjusted to best portray the distribution. Bin sizes range from 0.0001 in some cases to 0.1 in others.

Comparison of the original and revised mass fractions, along with their uncertainties, for MT53 is shown in Figure 6. There is close agreement with previously reported mass fractions and the measured values.

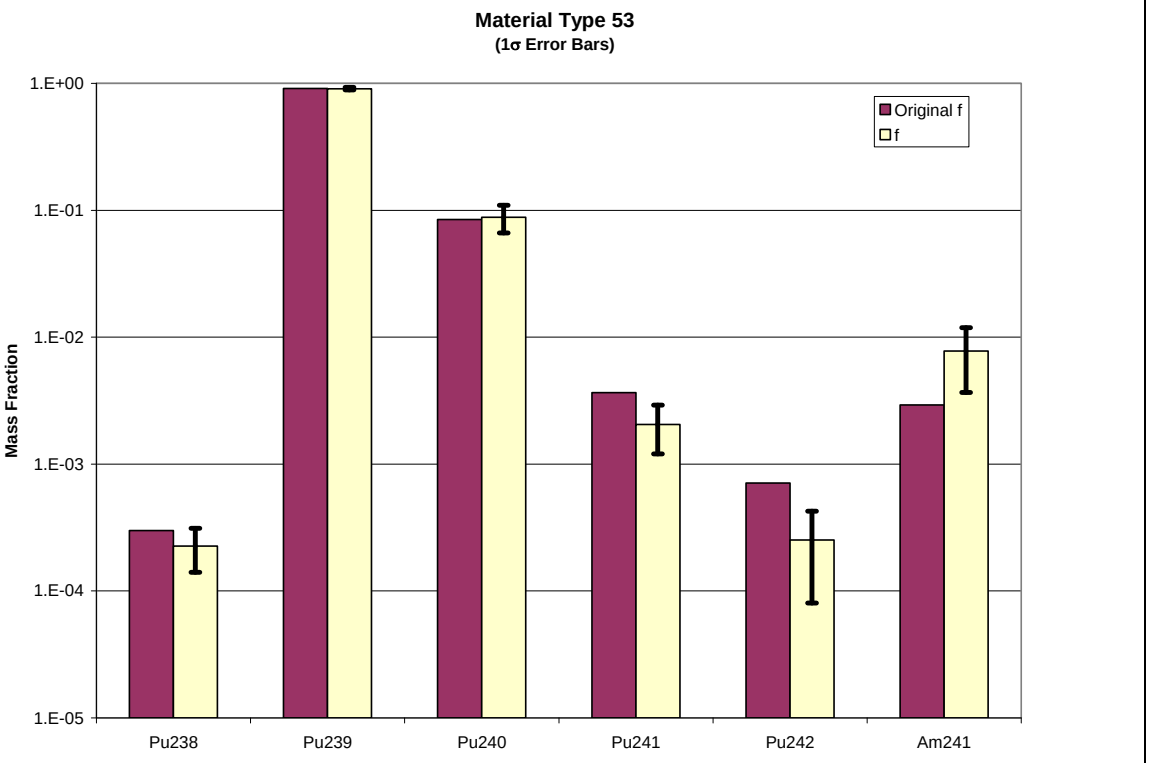

Figure 6. MT53 Comparison of Original and Evaluated Mass Fractions and Their Uncertainties

3.5. MT54

There were 210 assays in which MT54 was uniquely reported. The statistical results are summarized in Table 6. In all cases, the evaluated and normalized data are based on the 90th percentile and are the recommended values for mass fraction and relative uncertainties.

Table 6. MT54 Mass Fractions and Uncertainties

Data	# Assays	Description	Pu238	Pu239	Pu240	Pu241	Pu242	Am241
Raw Data From Appendix B	210	Mean	4.10E-03	9.07E-01	8.68E-02	2.23E-03	3.13E-04	2.45E-02
		%RSD	1232.4%	6.3%	39.8%	58.3%	84.5%	279.3%
		Median	2.99E-04	9.14E-01	8.21E-02	1.63E-03	1.80E-04	8.83E-03
		Max	7.31E-01	9.85E-01	2.54E-01	6.23E-03	1.40E-03	6.86E-01
		Min	0.00E+00	2.64E-01	1.52E-03	5.43E-06	5.00E-07	1.34E-03
Original MT Info	NA	Original f	4.60E-04	8.74E-01	1.15E-01	8.10E-03	2.20E-03	6.41E-03
		%RSD	Not Specified					
Evaluated Data	210	f_Upper	1.23E-03	9.74E-01	1.40E-01	3.90E-03	6.80E-04	9.96E-02
		f_Lower	1.50E-05	8.67E-01	4.05E-02	7.73E-04	4.30E-05	2.96E-01
		Base On	90%	90%	90%	90%	90%	90%
		f	3.27E-04	9.14E-01	8.59E-02	2.16E-03	2.96E-04	1.35E-02
		%RSD	68.9%	3.1%	31.2%	52.7%	77.7%	116.2%
Normalized	210	f	3.26E-04	9.12E-01	8.57E-02	2.15E-03	2.95E-04	1.35E-02
		%RSD	68.9%	3.1%	31.2%	52.7%	77.7%	116.2%

The distributions, Figure 7, are centrally distributed with some large outliers for all isotopes except Pu241. Application of the 90th percentile rule significantly reduces the relative standard deviation.

Comparison of the original and evaluated mass fractions, Figure 8, shows that both are generally similar. Pu239 error is small (about 3.1%) while the largest variation is for Am241 (116.2%).

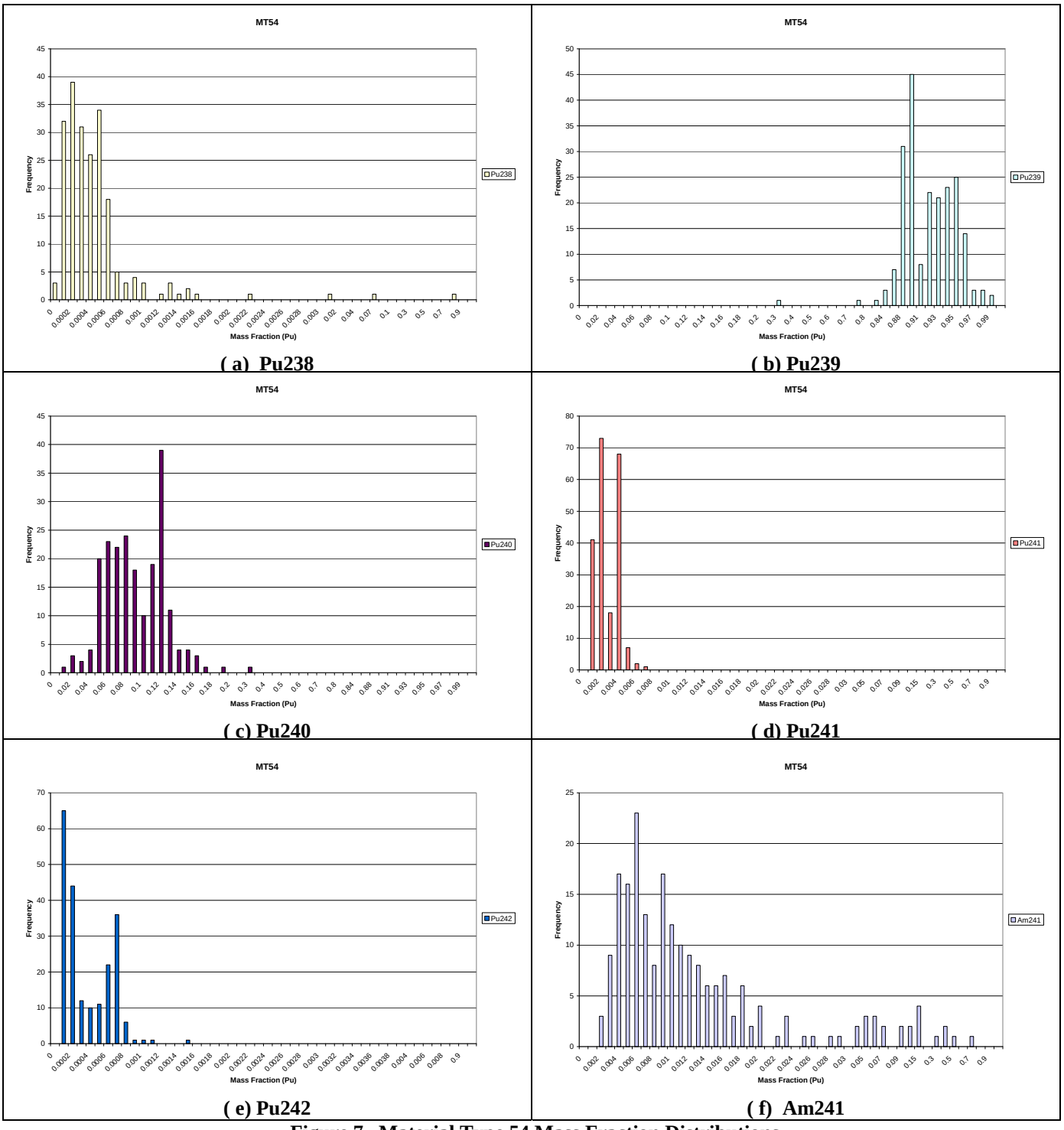

Figure 7. Material Type 54 Mass Fraction Distributions

NOTE: The scale on each figure is not necessarily linear—the distribution bin sizes were adjusted to best portray the distribution. Bin sizes range from 0.0001 in some cases to 0.1 in others.

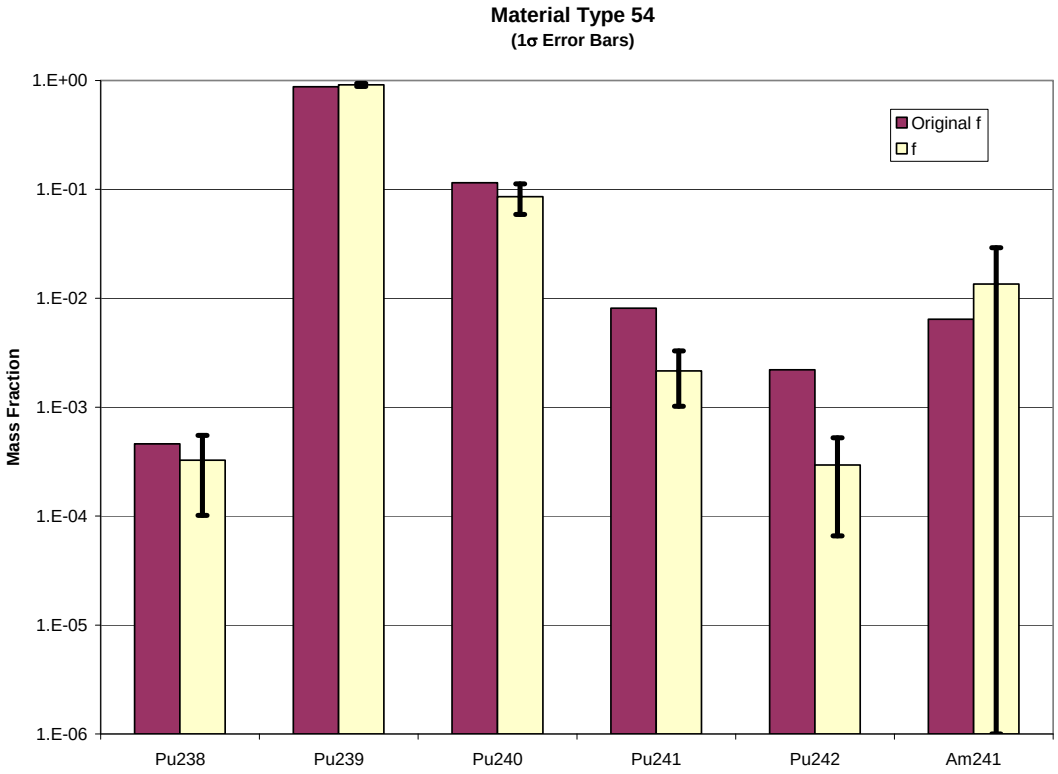

Figure 8. MT54 Comparison of Original and Evaluated Mass Fractions and Their Uncertainties

3.6. MT83

There were 204 assays that were reported as having uniquely MT83, heat source plutonium. The statistics of the mass fractions from the distributions are shown in Table 7.

Table 7. MT83 Mass Fractions and Uncertainties

Data	# Assays	Description	Pu238	Pu239	Pu240	Pu241	Pu242	Am241
Raw Data From Appendix B	204	Mean	7.46E-01	2.29E-01	2.21E-02	1.71E-03	1.03E-03	2.70E-03
		%RSD	21.4%	64.7%	84.0%	17.9%	92.4%	82.2%
		Median	8.05E-01	1.78E-01	1.68E-02	1.72E-03	7.98E-04	2.11E-03
		Max	8.58E-01	9.25E-01	9.72E-02	3.41E-03	5.65E-03	1.83E-02
		Min	7.55E-05	1.08E-01	0.00E+00	4.72E-04	1.01E-07	1.20E-04
Original MT Info	NA	Original f	8.39E-01	1.38E-01	1.90E-02	3.20E-03	9.00E-04	1.71E-03
		%RSD	Not Specified					
Evaluated Data	204	f_Upper	8.42E-01	4.80E-01	6.00E-02	2.67E-03	3.00E-03	6.15E-03
		f_Lower	4.72E-01	1.38E-01	2.00E-05	1.37E-03	9.50E-05	1.80E-04
		Base On	90%	90%	90%	90%	90%	90%
		f	7.79E-01	1.99E-01	2.04E-02	1.75E-03	9.21E-04	2.47E-03
		%RSD	8.5%	31.2%	67.6%	11.6%	67.4%	61.0%
Normalized	204	f	7.78E-01	1.99E-01	2.04E-02	1.74E-03	9.19E-04	2.47E-03
		%RSD	8.5%	31.2%	67.6%	11.6%	67.4%	61.0%

The mass fraction frequency distribution, Figure 9, appear normally distributed for Pu241 while the rest are either skewed or contain large outliers that erroneously affect the %RSD. Consequently, the 90^{th} percentile method was used for all isotopes and this had the effect of reducing the %RSD and is the recommended value for this material type.

Comparison of the original with the evaluated normalized mass fraction, Figure 10, shows that the two values are generally close.

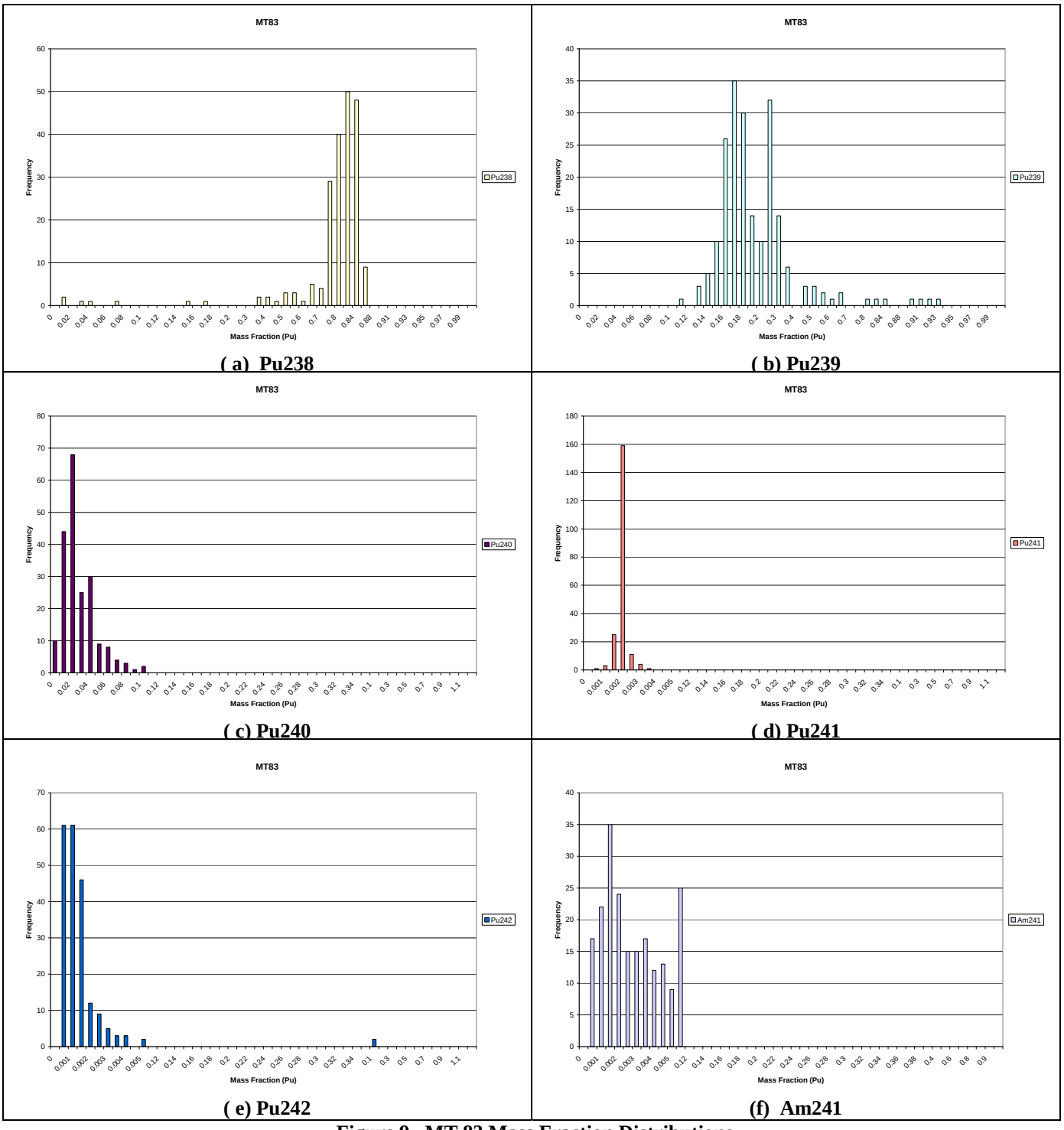

Figure 9. MT 83 Mass Fraction Distributions

NOTE: The scale on each figure is not necessarily linear—the distribution bin sizes were adjusted to best portray the distribution. Bin sizes range from 0.0001 in some cases to 0.1 in others.

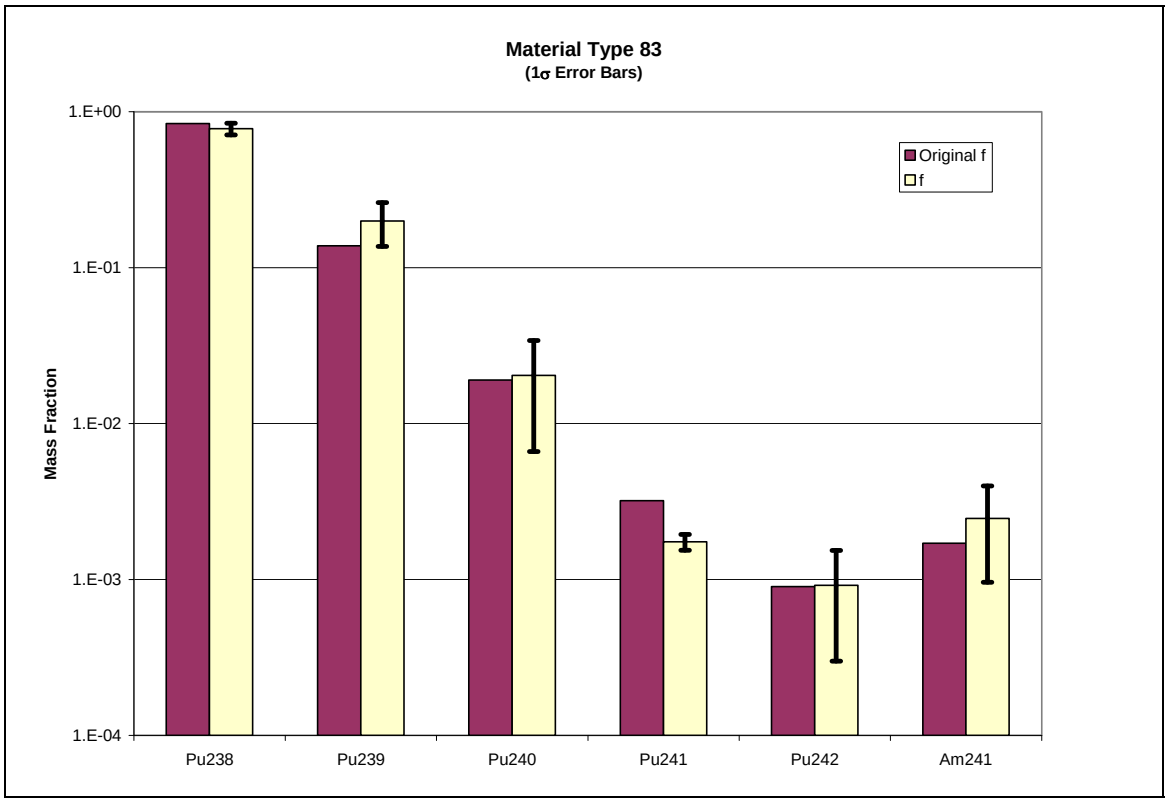

Figure 10. MT83 Comparison of Original and Evaluated Mass Fractions and Their Uncertainties

3.7. Average Isotopic Values

When a drum contains an unknown material type or a mixture with an unknown mix, an aggregate material type may be desired. Using assays from all material types except those that contain MT83 (either unique or in a mixture) appears to provide this type of aggregate. There are 3041 assays that meet the criteria and the statistics are tabulated in Table 8.

Table 8. Aggregate MT EXCEPT MT83

Data	# Assays	Description	Pu238	Pu239	Pu240	Pu241	Pu242	Am241
Raw Data From Appendix B	3041	Mean	7.81E-03	9.08E-01	7.50E-02	2.49E-03	7.21E-03	1.56E-02
		%RSD	904.6%	12.9%	62.7%	265.8%	955.4%	310.5%
		Median	1.62E-04	9.34E-01	6.38E-02	1.37E-03	1.13E-04	5.04E-03
		Max	8.61E-01	9.97E-01	6.59E-01	1.04E-01	9.74E-01	8.98E-01
		Min	0.00E+00	6.46E-03	0.00E+00	2.72E-06	2.00E-07	2.73E-04
Original MT Info	NA	Original f	NA	NA	NA	NA	NA	NA
		%RSD	Not Specified					
Evaluated Data	3041	f_Upper	3.00E-03	9.50E-01	1.27E-01	3.74E-03	7.10E-04	5.70E-02
		f_Lower	5.80E-05	8.50E-01	4.50E-02	9.30E-04	6.35E-05	2.08E-03
		Base On	90%	90%	90%	90%	90%	90%
		f	2.69E-04	9.28E-01	6.88E-02	1.57E-03	9.53E-05	8.09E-03
		%RSD	117.9%	1.9%	22.4%	34.1%	12.2%	107.7%
Normalized	3041	f	2.69E-04	9.29E-01	6.89E-02	1.57E-03	9.54E-05	8.10E-03
		%RSD	117.9%	1.9%	22.4%	34.1%	12.2%	107.7%

The frequency distributions for each isotope, Figure 11, appear to be normally distributed for Pu239 and Pu240 while the remaining isotopes are either skewed or contain large mass fraction outliers. Applying the 90^{th} percentile method significantly reduces the %RSD and provides a better representation of the uncertainties.

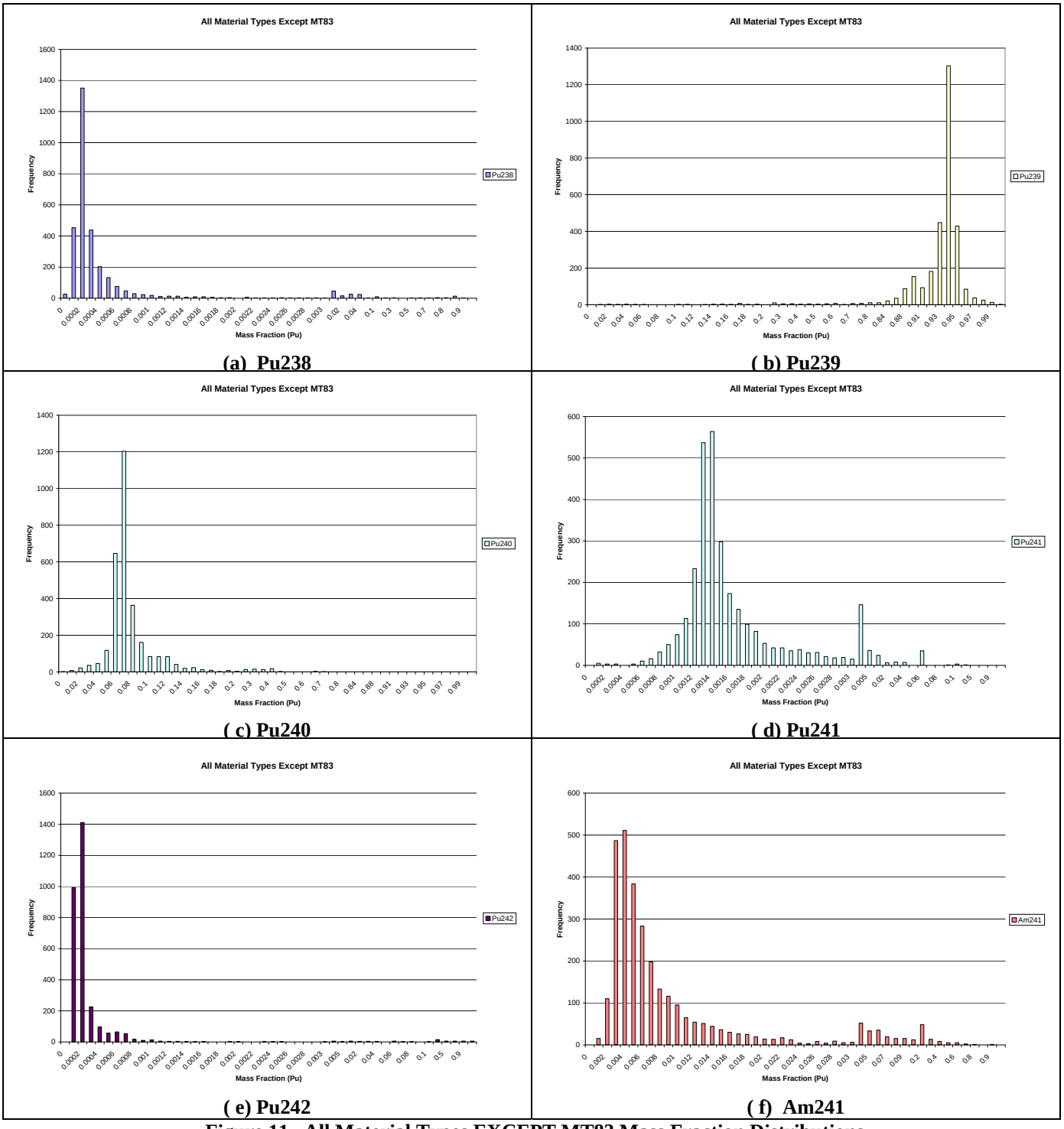

Figure 11. All Material Types EXCEPT MT83 Mass Fraction Distributions

NOTE: The scale on each figure is not necessarily linear—the distribution bin sizes were adjusted to best portray the distribution. Bin sizes range from 0.0001 in some cases to 0.1 in others.

The recommended mass fraction and the uncertainties are shown in Figure 12. The uncertainties for Pu238 and Am241 are large but the mass fractions are small, generally less than 1%, so the impact of using this aggregate will not overly impact the calculation of WIPP parameters (e.g., fissile gram equivalent, Pu239 equivalent curies, etc.). The largest variations are for Pu238 and Am241.

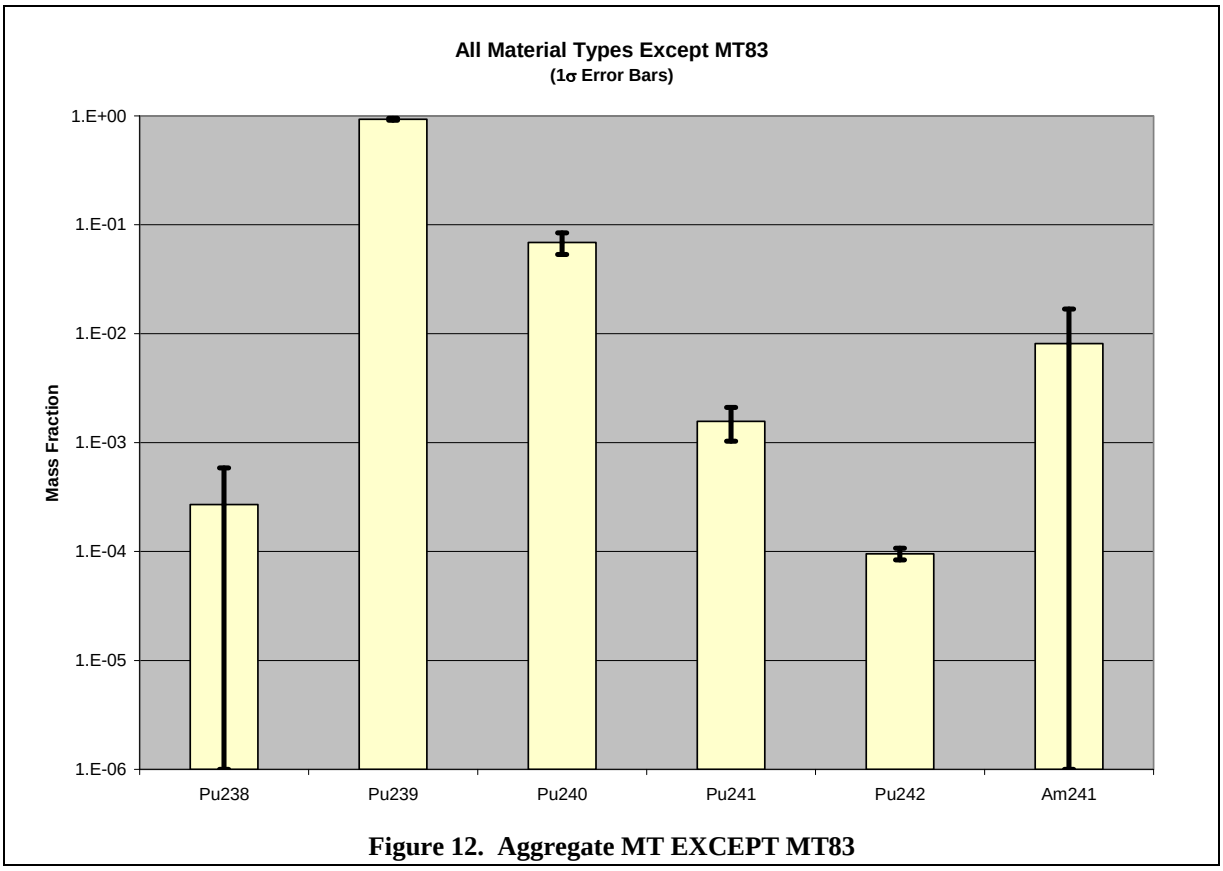

Figure 12. Aggregate MT EXCEPT MT83

4. Conclusions

Updated mass fractions for all the plutonium isotopes and Am241 have been calculated for material types 42, 52, 53, 54, and 83 and variations at the 68% confidence level (1-sigma) have been determined for each of those isotopes. In addition, an overall (or average) mass fraction and uncertainty by isotope has been recommended for other material types or when the material type contains a mixture excluding heat source (MT83). These results are summarized in Table 9.

Table 9. Mass Fractions and Uncertainties by Material Type

MT	Description	Pu238	Pu239	Pu240	Pu241	Pu242	Am241
42	Mass Fraction	2.8E-02	3.7E-01	2.8E-01	4.0E-02	2.8E-01	7.7E-02
	%RSD	50.25%	63.82%	48.04%	51.15%	115.92%	72.19%
52	Mass Fraction	2.05E-04	9.34E-01	6.46E-02	1.42E-03	9.93E-05	6.09E-03
	%RSD	78.9%	1.0%	14.9%	19.8%	22.5%	84.7%
53	Mass Fraction	2.26E-04	9.10E-01	8.79E-02	2.06E-03	2.52E-04	7.78E-03
	%RSD	37.91%	2.48%	24.49%	41.50%	68.13%	52.73%
54	Mass Fraction	3.26E-04	9.12E-01	8.57E-02	2.15E-03	2.95E-04	1.35E-02
	%RSD	68.90%	3.05%	31.22%	52.74%	77.65%	116.25%
83	Mass Fraction	7.78E-01	1.99E-01	2.04E-02	1.74E-03	9.19E-04	2.47E-03
	%RSD	8.54%	31.23%	67.56%	11.57%	67.40%	61.04%
Average (no MT83)	Mass Fraction	2.69E-04	9.29E-01	6.89E-02	1.57E-03	9.54E-05	8.10E-03
	%RSD	8.54%	31.23%	67.56%	11.57%	67.40%	61.04%

REFERENCES

[1] Benchmark Environmental Corporation Letter to Ms Dziewinska, *Material Type Isotopic Compositions*, Subcontract B45370007-8N, TWCP-0698, LA-UR-01-6170, May 2, 1997.
[2] Veilleux, J.M., *AK Isotopic Files For Input to NDA Radioassay Spreadsheets*, Los Alamos National Laboratory Memorandum, Sep 22, 2003.
[3] CCP-TP-125, *CCP Verification and Validation of FRAM and PTGS Nondestructive Assay Data Using a Manual Review Method*, Washington TRU Waste Solutions, Carlsbad, NM, Jun 5, 2005.
[4] PC/FRAM, *Plutonium and Uranium Isotopic Analysis Software*, Ortec, Oak Ridge, TN.
[5] Veilleux, J., *Test and Validation of Excel Spreadsheets: PuDecay and PuGamma*, Los Alamos National Laboratory Memorandum, TWCP-05390, Jan 26, 2001.
[6] Hamburg, M., *Statistical Analysis for Decision Making*, 2nd Edition, University of Pennsylvania, Harcourt Brace Jovanovich, Inc., NY, 1977
[7] DOE 5633.5, Figure VI-1, Nuclear Material Type Codes, May 22, 1987