

Radioactive Waste Inventory for the Clive DU PA

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1.0 Waste Inventory Parameters Summary

This section is a brief summary of parameters and distributions employed in the waste inventory component of the Clive Depleted Uranium Performance Assessment Model that is the subject of this white paper.

For distributions, the following notation is used:

- $N(\mu, \sigma, [min, max])$ represents a normal distribution with mean μ and standard deviation σ , and optional truncation at the specified *minimum* and *maximum*, and
- $Beta(\mu, \sigma, min, max)$ represents a generalized beta distribution with mean μ , standard deviation σ , minimum *min*, and maximum *max*.

A summary of values and distributions for waste inventory modeling inputs is provided in Table 1.

Table 1. Summary input parameter values and distributions

parameter	value or distribution	units	comments
Number of SRS DU drums	5,408	-	see Section 2.2.1
Mass of a 208-L (55-gal) drum	20	kg	see Section 2.2.1
Total mass of SRS DUO ₃ proposed for disposal	3,577	Mg	see Section 3.1.1
Number of DUF ₆ cylinders from Paducah GDP	36,191	-	see Section 3.5.1
Number of DUF ₆ cylinders from Portsmouth GDP	16,109	-	see Section 3.5.1
Number of DUF ₆ cylinders from K-25 GDP	4,822	-	see Section 3.5.1
Mass of DUF ₆ from Paducah GDP	436,400	Mg	see Section 3.5.1
Mass of DUF ₆ from Portsmouth GDP	195,800	Mg	see Section 3.5.1
Mass of DUF ₆ from K-25 GDP	54,300	Mg	see Section 3.5.1
Diameter of cylinders	4	ft	see Section 2.3.1
Length of cylinders	12	ft	see Section 2.3.1
Fraction of GDP DU that is contaminated	Beta(0.0392, 0.0025, 0, 1)	-	see Section 3.5.2.3

Mean and standard deviation values for uranium isotopes and other fission products in the uranium trioxide (UO₃) from the Savannah River Site (SRS) are developed in Section 3.0. These concentrations are summarized in Table 2. Note that the standard deviations are those used in the GoldSim PA model. They are intended to be estimates of the standard deviation of the mean concentration, hence addressing the spatio-temporal scale of the input distribution.

Table 2. Summary of mean and standard deviations for SRS DUO₃ concentrations, assuming a normal distribution

radionuclide	SRS DUO ₃ concentration	
	mean (pCi/g)	standard deviation (pCi/g)
⁹⁰ Sr	4.70E+1	1.28E+1
⁹⁹ Tc	2.38E+4	1.10E+4
¹²⁹ I	1.86E+1	1.59E+0
¹³⁷ Cs	1.21E+1	7.10E-1
²¹⁰ Pb	0	0
²²² Rn	0	0
²²⁶ Ra	3.17E+2	1.91E+1
²²⁸ Ra	0	0
²²⁷ Ac	0	0
²²⁸ Th	0	0
²²⁹ Th	0	0
²³⁰ Th	0	0
²³² Th	0	0
²³¹ Pa	0	0
²³² U	0	0
²³³ U	5.29E+3	4.78E+2
²³⁴ U	3.31E+4	2.17E+3
²³⁵ U	2.97E+3	7.50E+2
²³⁶ U	4.91E+3	1.17E+3
²³⁸ U	2.72E+5	6.64E+3
²³⁷ Np	5.68E+0	1.17E+0
²³⁸ Pu	2.10E-1	4.00E-2
²³⁹ Pu	1.28E+0	2.00E-1
²⁴⁰ Pu	3.40E-1	5.00E-2
²⁴¹ Pu	4.04E+0	7.40E-1
²⁴² Pu	0	0
²⁴¹ Am	1.42E+1	9.10E-1

The DU inventories from the gaseous diffusion plants (GDPs) are based upon estimates from the DOE (DOE 2004a and 2004b) for mass of DUF₆ and U₃O₈ produced. The inventories for the other actinides and fission products is highly uncertain, but is informed by studies performed by Oak Ridge National Laboratory (ORNL 2000a, 2000b, 2000c, 2000d). Until adequate information concerning DU inventory is received from the GDPs, which may not happen until the DU oxide product has been produced and sampled, the actinides and fission products are assumed to be in equal concentrations in the DUF₆ waste as has been found in the SRS DUO₃

waste, as shown in Table 2. This is only a rough approximation and will need to be revised as data from the GDP waste are provided.

2.0 Uranium Oxide Inventory

This document describes three categories of depleted uranium waste form at the Clive, Utah disposal facility:

1. Depleted uranium oxide (UO_3) waste from the Savannah River Site (SRS) proposed for disposal at the Clive facility,
2. DU from the GDPs at Portsmouth, Ohio and Paducah, Kentucky, which exists in two principal populations:
 - a) DU contaminated with fission and activation products from reactor returns introduced to the diffusion cascades, and
 - b) DU consisting of only “clean” uranium, with no such contamination.

The DU oxides that are to be produced at these sites’ “deconversion” plants will be primarily U_3O_8 . The remainder of this section provides background on the uranium cycle and origins and nature of DU waste in particular.

2.1 Depleted Uranium

In order to produce suitable fuel for nuclear reactors and/or weapons, uranium has to be enriched in the fissionable ^{235}U isotope. Uranium enrichment in the US began during the Manhattan Project in World War II. Enrichment for civilian and military uses continued after the war under the U.S. Atomic Energy Commission, and its successor agencies, including the DOE.

The uranium fuel cycle begins by extracting and milling natural uranium ore to produce “yellow cake,” a varying mixture of uranium oxides. Low-grade natural ores contain about 0.05 to 0.3% by weight of uranium oxide while high-grade natural ores can contain up to 70% by weight uranium oxide (NRC, 2010). Naturally occurring uranium contains three isotopes, ^{238}U , ^{235}U , and ^{234}U . Each isotope has the same chemical properties, but they differ in radiological properties. Naturally occurring U has an isotopic composition of about 99.2739±.0007% ^{238}U , 0.7204±.0007% ^{235}U , and 0.0057±.0002% ^{234}U (Rich et al., 1988).

The milled ore is refined to remove the decay products (^{226}Ra , ^{230}Th , etc.) that have built up in the material naturally to the degree of secular equilibrium, leaving more or less pure uranium oxide. This uranium, still at natural isotopic abundances, is enriched to obtain the ^{235}U , with vast quantities of ^{238}U as a by-product. Although a variety of technologies exist for enrichment, the most prevalent enrichment process at the time was by gaseous diffusion, which requires that the uranium be converted to a gaseous form: uranium hexafluoride (UF_6). This gas is introduced to a diffusion cascade, which separates the isotopes, generating enriched uranium as a product, and depleted uranium hexafluoride (DUF_6) as a waste stream. Depleted uranium isotopic ratio values from gaseous diffusion plants are roughly 99.75% ^{238}U , 0.25% ^{235}U , and 0.0005% ^{234}U (Rich, et al., 1988), but the ^{235}U assay found in the cylinders today varies with fluctuating enrichment goals, operational conditions, and where in the cascade process the DU was removed. Because

processing of uranium has been practiced for only about 60 years, there has not been sufficient time for appreciable in-growth of decay products in this by-product. Depleted uranium is therefore considerably less radioactive than natural uranium because it has less ^{234}U and other decay products per unit mass. The bulk of this material is still stored in the original cylinders in which it was first collected at the GDPs.

Uncontaminated (clean) depleted uranium consists principally of three isotopes of uranium (^{238}U , ^{235}U , and ^{234}U) and a small amount of progeny from radioactive decay of these isotopes. Trace amounts of other uranium isotopes (^{232}U , ^{233}U , and ^{236}U) may also exist. The bulk of the DU at the GDPs is clean uranium, but a significant amount of contaminated DU also exists, both at the GDPs and in all the DU waste from the SRS.

The contamination problem arises from the past practice of introducing irradiated nuclear materials (reactor returns) into the isotopic separations process. Irradiated nuclear fuel underwent a chemical separation process to remove the plutonium for use in nuclear weapons. Uranium, then thought to be a rare substance, was also separated out, but contained some residual contamination from activation and fission products. This uranium was again converted to UF_6 for re-enrichment, and was introduced to the gaseous diffusion cascades, contaminating them and the storage cylinders as well. Based on laboratory analysis of the contents of contaminated DU waste (including all radionuclides in the containers), the species in the disposed inventory include those in Table 3 (Beals, et al. 2002, *EnergySolutions* 2009b, and ORNL 2000c).

Table 3. Radionuclide constituents of contaminated depleted uranium

category	radionuclides
uranium isotopes	^{232}U , ^{233}U , ^{234}U , ^{235}U , ^{236}U , ^{238}U
decay products	^{226}Ra
activation products	^{241}Am , ^{237}Np , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu
fission products	^{90}Sr , ^{99}Tc , ^{129}I , ^{137}Cs

In order to clarify that the contaminated DU wastes contain more than just uranium or DU, they are termed “DU waste”. When this term is used, it refers to wastes that contain DU and a perhaps small but potentially significant amount of contamination from actinides and fission products.

2.2 Savannah River Site Depleted Uranium

Depleted uranium was generated at the SRS as a byproduct of the nuclear material production programs (Fussell and McWhorter, 2002). Depleted uranium billets were produced at the DOE Fernald, Ohio, site, fabricated into targets at SRS, then irradiated in one of the SRS production reactors. The irradiated targets were transported to F-Canyon where the targets were dissolved. After dissolution, the fission products were separated from the plutonium and uranium which were then separated from each other. After additional purification, the uranium stream was transferred to the FA-Line Facility where it was processed into uranium trioxide (UO_3) for storage in about 36,000 drums. Since the chemical separations process is imperfect, the DUO_3

contains trace quantities of fission products and transuranic elements (Beals et al, 2002, *EnergySolutions*, 2009b) as discussed above.

2.2.1 Mass of SRS Depleted Uranium Proposed for Disposal

The SRS DUO₃ is a solid powder at room temperature and pressures. This DU oxide is stored in 208-L (55-gal) steel drums, with plastic liners. Steel drums have a tare mass of about 20 kg each. The drums are approximately 2/3 full with an average mass of about 1500 lbm (750 kg) apiece (Fussell and McWhorter, 2002). This DUO₃ is considered to be relatively homogeneous, based on known process controls and operations. The condition of the drums varies from good to poor with a high percentage of the drums having some degree of outer surface corrosion.

In December 2009, SRS made a shipment of drums to the Clive, Utah facility. This shipment contained 52 rail-cars (referred to as gondolas in the manifests), each holding 104 drums, for a total of 5,408 drums. This shipment of DU waste is considered in this PA.

2.2.2 Composition of SRS Depleted Uranium

There are three main sources of data for establishing the concentration of uranium isotopes, fission products, and transuranics in the SRS DU. In 2002 SRS sampled and analyzed their DU oxide in preparation for shipment to Utah (Beals, et al., 2002). A total of 33 drums were sampled; this is approximately 1% of 3300 drums that were available for sampling. The samples were analyzed at the Savannah River Technology Center (SRTC) and by a Utah certified laboratory (BWXT Services, Inc) for uranium, fission, and transuranic radionuclides. The analytical results from SRTC are presented in Beals et al, 2002, and in an *EnergySolutions* Radioactive Waste Profile Record, referred to here as the 2002 Waste Profile Record (*EnergySolutions*, 2009b).

The 2002 Waste Profile Record (*EnergySolutions*, 2009b) provides activity concentration data for isotopes of uranium and for potential contaminants such as ⁹⁹Tc. The latter are used to characterize the contaminant radionuclides for the PA (see Section 3). The data for uranium isotopes are in the form of both activity concentration by alpha spectrometry, and atomic percent by mass spectrometry. ²³³U was not detected by mass spectrometry. The alpha spectrometry, also used to characterize the samples, cannot differentiate between ²³³U and ²³⁴U (or ²³⁵U and ²³⁶U) thereby requiring the mass spectrometry analysis. Note, the ²³⁵U and ²³⁶U results are also based on mass spectrometry analysis.

The 33 samples were characterized for uranium isotopes, fission products, transuranics, and some metals and organic compounds (pesticides, herbicides, semi-volatile and volatile organic compounds) as recorded in the Waste Profile Record (*EnergySolutions*, 2009b). No organic compounds were detected but low levels (mg/kg) of lead, arsenic, cadmium, chromium, selenium, silver, zinc and copper were found. These low levels of metal make up less than 5 ppm of the DU, and are not considered in this PA because they are not radioactive, and they are not in excess of minimum regulated concentrations for hazardous waste (i.e., the DU waste is not classified as “mixed waste”).

Data for other characteristics of the DU waste are also available from the 52 Waste Manifests (*EnergySolutions*, 2009d). The shipment consisted of 52 gondola railroad cars, each car

containing 104 drums. The 2009 Waste Manifests from that shipment provide the volume (total 1,133.2 m³) and weight (total of 7,886,738 pounds, corresponding to a mass of 3,577 Mg).

Based on the physical properties description in the Waste Profile Record (EnergySolutions, 2009b), the DU is stoichiometrically 83.22% uranium, indicating that the DU is essentially 100% UO₃. The isotopic mass percent of ²³⁸U is over 99%.

Since the arrival in Clive of the 52 gondolas of SRS DU waste, EnergySolutions has performed two separate sampling and analysis events. In January of 2010 EnergySolutions collected 15 samples that were analyzed for uranium isotopes (Table 14, in the Appendix). In April 2010 EnergySolutions collected 11 samples that were analyzed for uranium isotopes and ⁹⁹Tc (Table 15, in the Appendix). In August of 2010 the State of Utah analyzed 173 samples that EnergySolutions collected from the drums (Johnson, 2010). These samples were analyzed for ⁹⁹Tc only. The data are described in greater detail in Section 3, in which input distributions for the GoldSim PA model are developed.

2.3 Depleted Uranium Oxide from the Gaseous Diffusion Plants

Three large GDPs were constructed to produce enriched uranium. The first diffusion cascades were built in Oak Ridge, Tennessee, at what was the K-25 Site, but is now known as the East Tennessee Technology Park (ETTP). Two others of similar design were constructed in Paducah, Kentucky (PGDP), and Portsmouth, Ohio (PORTS) (DOE 2004a and 2004b). The cascades at the K-25 Site ceased operations in 1985, the Portsmouth plant ceased in 2001, the Paducah GDP continues to operate. The two more recent GDPs are host to a large inventory of stored DUF₆, including the ETTP material that was moved to Portsmouth.

The DOE is currently managing approximately 60,000 cylinders at both PGDP and PORTS (DOE 2004a, 2004b). For many years, interest has been expressed in converting the DUF₆ in these cylinders to an oxide form to support their long-term disposal. In May, 1995 an independent DOE oversight board recommended a study to determine a suitable chemical form for long-term storage of DU. Also, in 1994 the DOE began work on a *Programmatic Environmental Impact Statement for Alternative Strategies for the Long-Term Management and Use of Depleted Uranium Hexafluoride* (DOE 1999a). Later, DOE issued the *Final Plan for the Conversion of Depleted Uranium Hexafluoride as Required by Public Law 105-204* (DOE 1999b). As a result of these efforts the DOE developed a Conversion Plan that describes the steps that would allow DOE to convert the DUF₆ inventory to a more stable chemical form. Two Environmental Impact Statements (EIS) were prepared as part of the plan, one for Paducah, DOE/EIS-0359, (DOE 2004a) and one for Portsmouth, EIS-0360 (DOE 2004b). These EISs describe the background and alternatives for DUF₆ conversion. With the completions of the EISs, “deconversion” plants were built at both the PORTS and PGDP locations. In 2002, DOE awarded a contract to Uranium Disposition Service, LLC (UDS) to design, construct, and operate two DUF₆ deconversion facilities at these locations. As of this writing, both plants have been built by UDS and have begun test processing DUF₆ into oxide form.

The UDS dry conversion is a continuous process in which DUF₆ is vaporized and converted to a mixture of uranium oxides (primarily DU₃O₈ but with some UO₂) by reaction with steam and hydrogen in a fluidized-bed conversion unit. The hydrogen is generated using anhydrous ammonia (NH₃). Nitrogen is also used as an inert purging gas and is released to the atmosphere

through the building stack as part of the clean off-gas stream. The DU_3O_8 powder is collected and packaged in the former DUF_6 cylinders for disposition. The process equipment is arranged in parallel lines. Each line consists of two autoclaves, two conversion units, a HF recovery system, and process off-gas scrubbers (DOE 2004a).

2.3.1 Mass of GDP Depleted Uranium

According to the EISs the PGDP facility has been designed to convert approximately 18,000 Mg (one Mg is one metric tonne, or about 2,200 lbm) of DUF_6 per year, which will require approximately 25 years for full conversion of the PGDP inventory. At Portsmouth, 13,500 Mg of DUF_6 per year (approximately 1,000 cylinders per year) is expected to be converted.

Several different cylinder types are in use. Most cylinders are expected to range from 11 to 12 Mg full. The cylinders with a 12-Mg capacity are 12 ft (3.7 m) long by 4 ft (1.2 m) in diameter; most have a steel wall that is 5/16 in (0.79 cm) thick. Similar but slightly smaller cylinders with a capacity of 9 Mg are also in use. Most of the cylinders were manufactured in accordance with an American National Standards Institute standard (ANSI N14.1, Uranium Hexafluoride Packaging for Transport) as specified in 49 CFR 173.420, the Federal regulations governing transport of DUF_6 .

To develop an estimate for the mass of DU oxide from the two GDPs, the mass of DUF_6 was converted to mass of uranium and thence to mass of U_3O_8 . This simple stoichiometric conversion, based on moles of uranium, fluorine, and oxygen, is performed within the Clive DU PA Model. Details are provided in Section 3.5.1.

2.3.2 Composition of GDP Depleted Uranium

The depleted uranium oxides from Portsmouth and Paducah that are proposed for disposal have yet to be manufactured. Until their production is complete, with associated testing of composition, estimates of composition must be relied upon to construct distributions and make decisions. At the most coarse level, there are two distinct populations of GDP DU composition: 1) DU derived from "clean" (a.k.a. "green") uranium, which contains no contamination, and 2) contaminated DU, which contains varying amounts of fission and activation products, as well as transuranics, resulting from the introduction of reactor returns into the gaseous diffusion cascade.

The clean DU is characterized by its abundance of uranium isotopes, and includes those radionuclides as well as their decay products. Isotopic abundance analyses were focused on determining the amount of U-235 in the DU, since this isotope was the "product" of the entire enrichment enterprise, and little attention was given to the exact abundance of other uranium isotopes, all of which were considered waste products.

Little information is available at this time regarding the exact nature and extent of the contamination within the contaminated DU population. The uranium isotopic abundance estimates are the same as for the clean DU. Estimates of the contamination by reactor return radionuclides, however, must rely on the SRS DU as a proxy until better GDP-specific information becomes available. For the purposes of this PA, then, the contaminated fraction of the GDP DU is assumed to have the same contaminant composition as the SRS DU.

3.0 Input Parameter Distribution Development

The probabilistic Clive DU PA Model relies on stochastic parameters in order to evaluate uncertainty and sensitivity. The statistical development of input parameter distributions is provided here.

3.1 Parameters for Depleted Uranium from the Savannah River Site

Parameters of interest for the PA include the mass of DU waste, and the concentrations of each radio-isotope contained in the DU waste. The contents of the SRS drums were described in Section 2.1. The purpose of this section is to describe the characterization of the mass of DU, and the concentrations of the radioisotopes. The mass of DU is considered fixed for the purpose of this PA, and is presented without uncertainty. The concentrations are presented in terms of the best estimate of the mean concentration, and the uncertainty of the mean concentration for each radio-isotope.

3.1.1 Mass of SRS Depleted Uranium

The single source of information regarding the mass of total depleted uranium shipped from SRS to Clive are shipping manifests (EnergySolutions, 2009d). Key pieces of information on these forms include the following

- Total mass in kg and corresponding weight in US tons
- Total volume in cubic meters and in cubic feet
- Net waste volume in cubic meters and in cubic feet
- Net mass in kg and corresponding net weight in US tons

Reviewing these manifests suggest that each gondola rail car was weighed empty (tared) and fully loaded, and the tare weight was subtracted to arrive at the “Net Waste Weight” reported on the manifests. Since this is a measured amount, it will be considered a fixed value and a distribution will not be assigned. There is no reason to believe that the mass of the drums was deducted from this net weight. Such drums do not have a standardized tare weight, but for the purposes of calculation it is assumed that each drum has a mass of 20 kg. This is considered a representative weight for a 55-gallon drum.

The net weights from the manifests were summarized by W. Johns in a spreadsheet (“100105 9021-33 Iso With Calcs.xls”) sent to Neptune. These values have been summed to create a total mass data value for total mass of the depleted uranium shipped from SRS to Clive, Utah. Weights of DU plus drums for the individual 52 rail cars range from 50.37 Mg to 75.56 Mg. The total amount shipped is 3,577 Mg.

3.1.2 Composition of SRS Depleted Uranium

Three data sources are available for the development of probability distributions for the concentrations of radio-isotopes in the SRS DU waste: The SRS-2002 dataset consists of activity concentration data and uranium isotopic abundance as atomic percent from Beals, et al. (2002). The ES-2010 dataset has uranium activity concentration and total uranium mass concentrations from two EnergySolutions sampling and analysis events: GEL (2010a and 2010b), and GEL (2010c). Finally, the Utah-2010 analysis obtained activity concentrations of ⁹⁹Tc from

EnergySolutions sampling and State of Utah requested analysis (Johnson 2010). These datasets are briefly described in Table 4 and the individual values are presented in Appendix A.

Table 4: Summary of available uranium and technetium data for the SRS DU

Source	Date	Number	Constituents	Units
SRS-2002: Table 16 of Beals et al, (2002)	2002	6 (2 replicates per sample)	^{233}U , ^{234}U , ^{235}U , ^{236}U , ^{238}U	Isotopic abundances (atomic % U)
SRS-2002: Table 17 and Table 4 of Beals et al, (2002)	2002	33	$^{233+234}\text{U}$, $^{235+236}\text{U}$, ^{238}U , ^{99}Tc	Activity % U
ES-2010 (GEL, 2010 a,b)	January 2010	15	Total U, $^{233+234}\text{U}$, $^{235+236}\text{U}$, ^{238}U	$\mu\text{g/g}$ for Total U; pCi/g for others
ES-2010 (GEL 2010 c)	April 2010	11	Total U, ^{99}Tc , $^{233+234}\text{U}$, $^{235+236}\text{U}$, ^{238}U	$\mu\text{g/g}$ for Total U; pCi/g for others
Utah-2010 (Johnson, 2010)*	August 2010	173 (plus 30 duplicates)	^{99}Tc	pCi/g

* Note that splits of these samples were also submitted for analysis by EnergySolutions.

Note that the 33 samples included in the SRS-2002 data also include concentrations of the other contaminants presented in Table 3 (decay, activation and fission products), which are used to developed input probability distributions for the concentrations of these radionuclides.

The spatio-temporal scale of interest for the Clive DU PA Model includes a large volume of DU waste and fill material in the Class A South embankment, a 10 ky quantitative analysis followed by a 2.1 My qualitative analysis. This, and the dynamic nature of the PA modeling environment in which time steps of many years are used, affects the approach to characterizing probability distributions of the inventory. Conceptually, the PA model incorporates compartments or cells that are fully mixed at each time step. The physical samples used in this statistical analysis represent very small volumes of waste, but the mean concentrations are representative of the entire inventory. This approach is reasonable so long as there is not a strong non-linear effect due to spatial variation within the waste cell. For this model the waste is fully mixed within a waste layer. The appropriate spatio-temporal scaling suggests that characterization of the mean activity concentration of each radionuclide is needed. This is the basic approach that is taken in each case, however, because the data sources are different for some of the radionuclides, different approaches are needed for estimation of the probability distributions (Table 5 and Table 6):

- The probability distribution of mean activity concentration for uranium isotopes is estimated from the ES-2010 data. Because activity from combinations of isotopes $^{233+234}\text{U}$ and $^{235+236}\text{U}$ is reported in ES-2010, the atomic percent data from SRS-2002 is used to partition these isotopes.
- There are three sources of ^{99}Tc data: SRS-2002, ES-2010, and Utah-2010. These datasets are used to estimate mean ^{99}Tc activity concentrations. Note that the duplicate measurements in Utah-2010 were not used because there are many samples (173) without

the duplicates, and the duplicates were found to be dependent on their original samples (separating out those dependencies statistically is complicated and unnecessary given the large number of samples available).

- The SRS-2002 data provide the only data available for the other radionuclides (americium, cesium, radon, iodine and plutonium). Consequently, these data are used to estimate distributions of mean activity concentrations for these radionuclides. The parameter estimates for the probability distributions of the mean activity concentrations for these radionuclides are presented in Table 5.

Therefore, the approach for distribution development is to establish the uncertainty distribution of the mean activity concentration for each radionuclide. Each individual data set available is reasonably well-behaved statistically, not exhibiting large skew or multi-modality. There are also enough data that the Central Limit Theorem can be applied, implying a normal distribution for the distribution of the mean. The normal distributions are characterized with the mean concentration and the standard error (i.e., the standard deviation of the mean). However, different sampling events for ^{99}Tc and U indicate potentially different measurement types between sampling events. Consequently, for ^{99}Tc and uranium isotopes, bootstrap re-sampling of the samples and the sampling events is used to address possible differences between sampling events. For the remaining radionuclides, the SRS data are used directly to estimate the parameters. The final distributions are presented in Table 5 and Table 6. Details of the development of these distributions are in the following sections.

Table 5: Summary of probability distributions of mean activity concentrations (pCi/g) for uranium and technetium

Radioisotope	Mean	Standard Error	Source
^{99}Tc	23,800	11,000	SRS-2002, ES-2010 (Jan), Utah-2010
$^{233}\text{U}^*$	5,290	478	ES 2010 (Jan/Apr)
$^{234}\text{U}^*$	33,100	2,170	ES 2010 (Jan.Apr)
$^{235}\text{U}^*$	2,970	750	ES 2010 (Jan.Apr)
$^{236}\text{U}^*$	4,910	1,170	ES 2010 (Jan.Apr)
^{238}U	272,000	6,640	ES 2010 (Jan.Apr)

* Isotopes are partitioned using SRS 2002 atomic percentage data.

Table 6: Summary of probability distributions for mean activity concentrations (pCi/g) for other radioisotopes. (Source: SRS-2002.)

Radioisotope	N	Mean	Std. Error
^{241}Am	33	14.2	0.91
^{137}Cs	33	12.1	0.71
^{129}I	33	18.6	1.59
^{237}Np	33	5.68	1.17
^{238}Pu	31*	0.21	0.04

Radioisotope	N	Mean	Std. Error
²³⁹ Pu	31*	1.28	0.20
²⁴⁰ Pu	31*	0.34	0.05
²⁴¹ Pu	31*	4.04	0.74
²²⁶ Ra	33	316.8	19.1
⁹⁰ Sr	33	47.0	12.8

* - note that results for plutonium isotopes were not reported for 2 samples in the SRS-2002 data

3.2 Analysis of Uranium Composition in SRS Depleted Uranium

Direct comparison between uranium concentrations represented in the SRS-2002 data and in the ES-2010 data is complicated by several factors. The ES-2010 data represent activity concentrations for uranium, where the SRS-2002 data represent isotopic abundance as activity percent (%) of uranium, rather than activity concentration. These different expressions of uranium activity cannot be reconciled without recourse to the total proportion of uranium in each sample—information that is not available. Further, the pedigree of the SRS-2002 data is not clear. Information is available in Beals et al. (2002) about the analytical methods performed in the laboratory, but the actual laboratory reports for the SRS-2002 data are not available. In contrast, the pedigree of the ES-2010 data is well known, and the laboratory reports are available to support the reported uranium activity concentrations. Consequently, only the ES-2010 data are used to generate distributions of the mean uranium activity concentration for each uranium isotope. However, an exploratory comparison is made between the SRS-2002 and the ES-2010 activity data to understand the differences between the SRS and ES uranium data. Development of input probability distributions is presented after the exploratory comparison.

For the PA model, separation is also needed for the uranium isotopes in the pairs ²³³⁺²³⁴U and ²³⁵⁺²³⁶U. The ES-2010 laboratory analysis and subsequent uranium data do not distinguish between these pairs of isotopes, but report ²³³⁺²³⁴U and ²³⁵⁺²³⁶U activity concentrations combined. However, the SRS-2002 study also includes some uranium isotopic abundance data presented as atomic percent (%) for all uranium isotopes. These SRS-2002 atomic% data are used to partition the ²³³⁺²³⁴U and ²³⁵⁺²³⁶U activity concentration data obtained from ES-2010.

3.2.1 Exploratory Comparison of Uranium Data

In SRS-2002, activity% for all uranium isotopes was measured at SRS using alpha spectrometry. In ES-2010, activity concentrations (pCi/g) were measured for ²³³⁺²³⁴U, ²³⁵⁺²³⁶U, and ²³⁸U. As noted above, only the ES-2010 data will be used to develop input distributions for uranium concentrations for the PA model. However, a comparison of the ES-2010 and SRS-2002 data is presented to better understand the limitations of the SRS-2002 data, and to support the contention that the ES-2010 data are more appropriate for use in developing input distributions for uranium activity concentrations for the PA model.

A major consideration in the decision to focus on the ES-2010 for development of input distributions for the PA model is the lack of supporting documentation for the SRS-2002 data and the difficulty of converting from data presented in activity% to activity concentration. The ES-2010 and SRS-2002 data are compared by first translating one of the datasets to the units of the other dataset. The approach taken is to convert the ES-2010 data to activity%. This is a relatively simple step that facilitates comparison of the SRS-2002 and ES-2010 datasets.

Activity% can be calculated directly from activity concentrations (Equation 1).

$$A_i = \frac{c_i}{\sum_j c_j} \times 100 \quad (1)$$

where

A_i = activity% of uranium component i , and

c_i = activity concentration for uranium component i , which indexes $^{233+234}\text{U}$, $^{235+236}\text{U}$, ^{238}U

The results of this conversion are presented graphically in Figure 1. This figure shows pairs of scatter plots for the different uranium components. These plots show clear difference between the datasets. For example, there is a cluster of points from the SRS-2002 dataset (circles). As originally ordered and labeled in Beals et al. (2002), the first 21 samples form the close cluster of points while the last 12 points form the more dispersed cluster of points. Without any further information, this is suggestive of either sampling or laboratory differences or biases within the SRS-2002 data. Sample IDs could be surrogates for sample location, perhaps representing samples from barrels of similar wastes, which would be an example of a potential sampling bias if the entire waste stream is not relatively homogeneous. Alternatively, the samples could have been analyzed in separate batches on different days – with different ambient background concentrations being subtracted from each batch, which would be an example of laboratory bias. No information has been found to explain these differences, but this provides further evidence for why these data are not included in the development of the probability distributions for uranium isotopic inventory for the Clive DU PA.

Data from the two 2010 ES sampling events form clusters that are different but with some overlap. The data from the ES-2010-January sampling event have greater standard deviation than those from the April sampling event. The $^{235+236}\text{U}$ data tend to be slightly greater for the January sampling event, whereas the ^{238}U data tend to be slightly greater for the April sampling event.

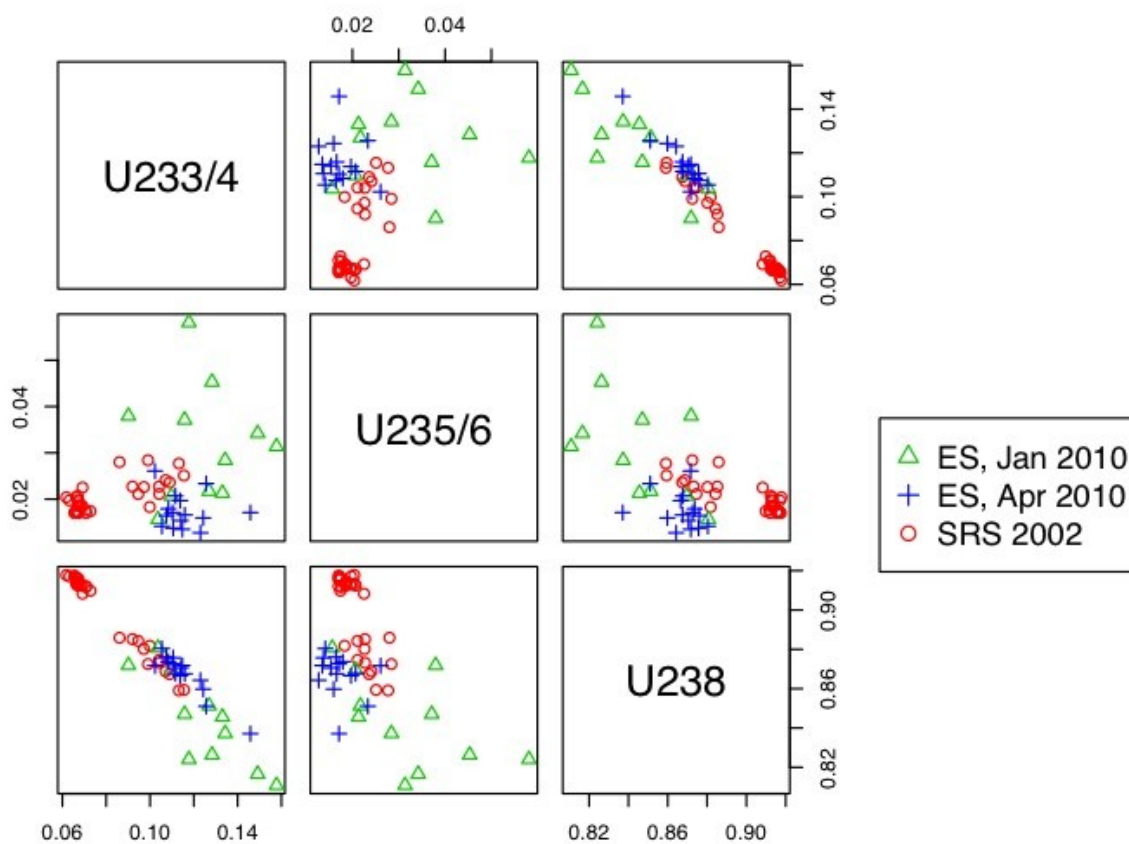
The greatest overall difference is between the first cluster (21 samples) from SRS-2002 and the rest of the data. This cluster has markedly lower $^{233+234}\text{U}$ activity% values than the remainder of the data, and, consequently, markedly greater ^{238}U activity% values.

The summary statistics for each dataset in Figure 1 are presented in Table 7. They further demonstrate the differences between the datasets. The questionable pedigree and difference between the two clusters in the SRS-2002 data are sufficient to justify not using these data for distribution development for the PA. The differences, particular in standard deviation, between the two ES datasets suggest that these two datasets should not be combined when estimating input probability distributions for the uranium activity concentrations for the PA model.

The next stage in this exploratory analysis of the SRS-2002 and ES-2010 Uranium data is to convert the SRS-2002 data from activity% to activity concentrations. This is done to see if the same basic results are obtained, considering different inputs are needed for this conversion.

Table 7: Summary statistics for the Uranium activity% data

Radioisotope	SRS-2002 (33 samples)		ES-2010-January (11)		ES-2010-April (15)	
	Mean	Std. Dev.	Mean	Std. Dev.	Mean	Std. Dev.
$^{233+234}\text{U}$	8.0%	1.8%	11.6%	1.1%	12.4%	2.0%
$^{235+236}\text{U}$	2.0%	0.3%	1.7%	0.4%	3.2%	1.2%
^{238}U	90.0%	2.0%	86.7%	1.1%	84.4%	2.3%

**Figure 1. Comparison of activity percent for the SRS DU uranium isotopes**

3.2.2 Partitioning $^{233+234}\text{U}$ and $^{235+236}\text{U}$

The Clive DU PA model requires probability distributions of activity concentration for each uranium isotope. Because of the methods used to measure radioactivity, most samples collected in 2002 and in 2010 do not distinguish between ^{233}U and ^{234}U or between ^{235}U and ^{236}U , but rather report combined quantities. To separate the isotopes some data on the relative contributions of each isotope in each pair is needed.

From the SRS-2002 data, 6 samples were analyzed using mass spectrometry. These 6 samples are from the original 33 samples that were analyzed for activity% of uranium. The mass spectrometry method identified all uranium isotopic abundances and the results are expressed as atomic% (see Table 12). The dataset provides two values for each sample. These values are treated as duplicates and the values are averaged for use in subsequent analyses.

All abundance values for ^{233}U are reported as 0.0000%, because it was not identified in any sample. However, to allow for the possibility of a trace quantity of ^{233}U in the SRS DU, for both SRS-2002 and ES-2010 datasets, ^{233}U atomic percentage values are assumed to be 0.00005%, a value that was chosen because any value smaller than that would be recorded as 0.0000% to four decimal places.

To partition activity% and activity concentrations for $^{235+236}\text{U}$ and $^{233+234}\text{U}$, uranium abundances expressed as atomic% are multiplied by their respective specific activities, and renormalized to calculate activity%. Ratios are presented in Table 8. The atomic% data do not sum to exactly 100%, hence the renormalization causes small differences in the ^{233}U activity% values.

Table 8: Partitioning Ratios for Uranium Isotopes

Sample	Radionuclide				Ratios	
	^{233}U	^{234}U	^{235}U	^{236}U	$^{234}\text{U}/^{233}\text{U}$	$^{236}\text{U}/^{235}\text{U}$
3	1.29%	7.03%	0.73%	1.12%	5.45	1.54
9	1.29%	6.73%	0.73%	1.11%	5.20	1.53
17	1.29%	7.13%	0.73%	1.14%	5.54	1.56
20	1.28%	7.33%	0.74%	1.16%	5.70	1.58
25	1.22%	11.50%	0.83%	1.56%	9.43	1.89
30	1.25%	9.80%	0.78%	1.46%	7.87	1.86

Both sets of ratios show similar patterns, clearly demonstrating that the last two samples are different than the first four samples. This also matches the differences observed in the activity% data reported in the 33 samples, for which the first 21 samples are clearly different than the last 12 samples (see Figure 1). However, all six samples are used to separate these isotopes for the PA model, the effect of which is to increase the variance of the ratios, which introduces more uncertainty in the PA model. In general, the differences this causes in uranium activity concentrations are fairly small relative to the likely effect on the PA model results, however, this will be tested in the model evaluation and sensitivity analysis. If the uranium isotopic distributions prove to be sensitive in the PA model, then it might be necessary to collect data that are aimed more specifically at the needs of the PA.

3.2.3 SRS Depleted Uranium Activity Concentration

As illustrated in Figure 1, there are differences between concentrations measured by ES in the January and April, 2010, data. (Note, as described in Section 3.2.1, the SRS-2002 uranium data are not included in the development of input distributions for uranium activity concentrations for the PA model.) The focus is on the ES-2010 datasets. The data from these two ES-2010 dataset are not considered independent or exchangeable, in which case they cannot be directly combined.

Consequently, in order to estimate the population mean and the standard deviation of the mean, a bootstrap method is used giving equal weight to both ES-2010 sampling events.

To simulate the two sampling events, all combinations of the ES-2010 January and April sampling events were used. The samples are bootstrapped within each sampling event, the mean value is calculated for each study, and the study means are averaged to obtain an overall mean value. The bootstrap method is applied as follows:

1. The two sampling events are selected with replacement. Since there are only 4 possible combinations of sampling events (select the January event twice, select the April event twice, select the January event followed by the April event, and select the April event followed by the January event – this is analogous to the results that could be obtained by tossing a coin twice), all combinations are used and weighted equally.
2. For each sampling event selected, the data are sampled with replacement and a mean mean calculated. An overall mean is calculated as an average of the two means.
3. This simulation is repeated 10,000 times for each of the 4 sampling event combinations, to construct a distribution of means. The simulations were selected at random. This large number of simulations provided adequate convergence of the distribution of the mean.

The effect of this approach is that the effective sample size is related more to the two sampling events than to the 26 samples. This leads to a comparatively wide distribution. If instead, all 26 samples had been treated as independent, then the standard deviation would be considerably smaller. The conceptual difference between the two possible approaches is that treating the data as independent assigns the information content, or uncertainty, to each sample, whereas, the approach used assigns the information content to the sampling events. That is, the sampling events themselves are considered more important for characterizing the distribution of the uranium isotopes than the individual sample results.

10,000 bootstrap samples are used, to create the distributions of mean values for each uranium component shown in Figure 2. The distributions for the uranium components are presented to show how the distributions relate to the two ES-2010 datasets. The red lines on the plots show how the April data exhibit greater activity concentrations for all three uranium components. The plots also show how the distributions bound the means of the two datasets for all three uranium components. If an approach had been taken that treated all 26 data points as independent, then the distributions of the means would probably have fallen between the two means.

The distributions of the uranium components $^{233+234}\text{U}$ and $^{235+236}\text{U}$ are partitioned using a randomly assigned ratio from one of the 6 ratios presented in Table 8. That is, each of the 10,000 simulated means is partitioned, so that there are 10,000 realizations of the distributions of the individual uranium isotopes. The resulting distributions of the mean uranium isotope activity concentrations were fit using a normal distribution. The resulting distributions are presented in Table 5.

The activity concentrations of uranium are dominated by ^{238}U at an average of 272,000 pCi/g. This is to be expected, although the mean activity concentration of ^{234}U is also large compared to the other isotopes. These distributions could be narrowed (i.e., reduced uncertainty) by collecting new data under an experimental design that is aimed at the needs of the PA. This includes activity concentrations over a wide range of drums, locations in drums, and laboratory

analysis that provides activity concentrations for every uranium isotope. The ES-2010 datasets provide reasonable data, but the two datasets present different mean uranium activity concentrations, in which case there would be benefit from a more complete study of uranium in the SRS DU waste. If, given these relatively broad distributions, the uranium isotopes are not sensitive to any PA model endpoint, then the need to refine these distributions will be less.

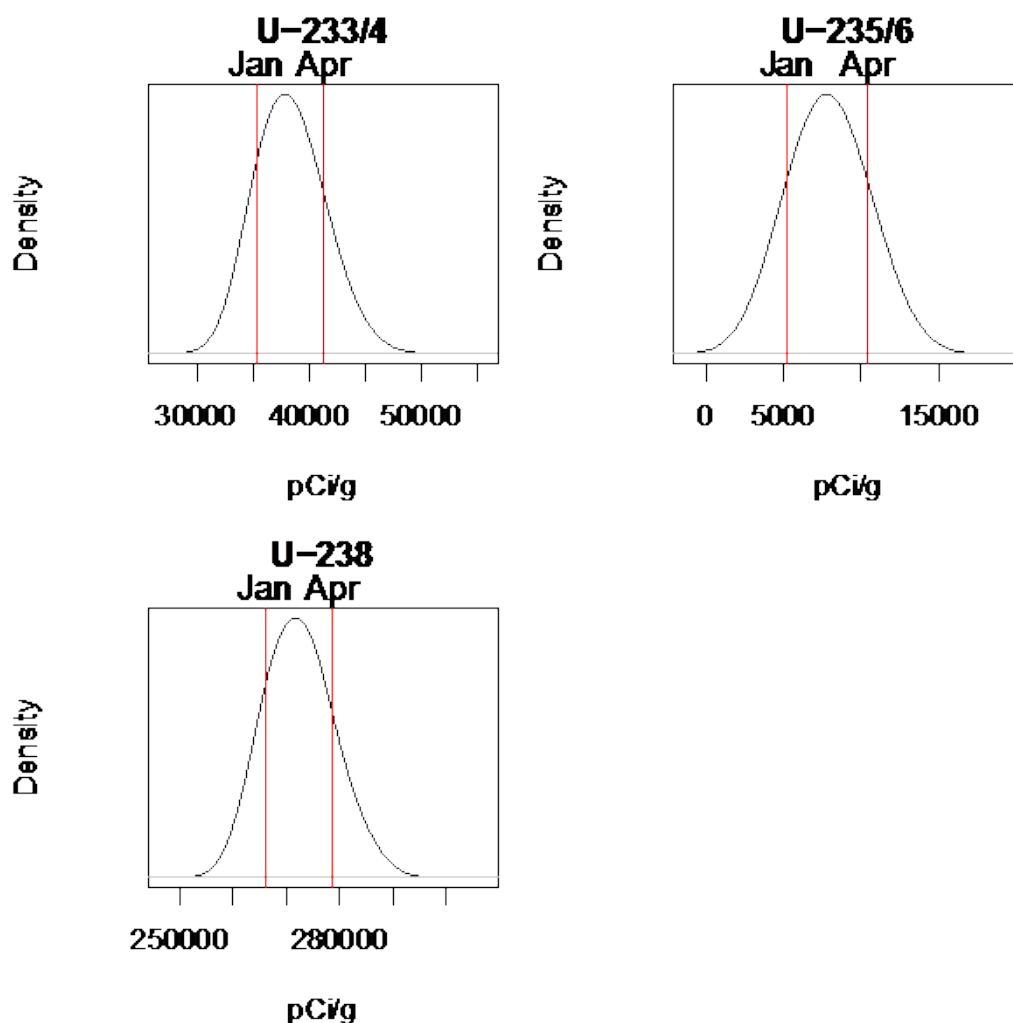


Figure 2. Distribution of mean activity concentration values from bootstrap resampling.

Mean concentration from each input data set are denoted by vertical red lines. To compare with original ES data, mean concentrations of $^{233+234}\text{U}$, $^{235+236}\text{U}$ and ^{238}U components are shown (red lines) for both the ES-2010 January and April datasets.

3.3 Analysis of Technetium Concentrations in SRS DU

Technetium-99 is the most important of the contaminants contained in the SRS DU waste, because of its potential for relatively fast transport to groundwater. Other mobile radionuclides were reported as not detected in the SRS-2002 samples.

Three sources of data exist for ⁹⁹Tc from the following sampling events: SRS-2002 (33 samples), ES-2010 (11 samples), and Utah-2010 (173 samples – without duplicates). Figure 3 shows that the samples from these three sampling events have different mean concentrations and different standard deviations. The original SRS-2002 data show the greatest concentrations. EnergySolutions attempted to verify these concentrations in January 2011. However, the ES-2010 ⁹⁹Tc showed lower concentrations. Given the uncertainty and importance of understanding the ⁹⁹Tc concentrations, the State of Utah commissioned a study involving sampling and analysis of ⁹⁹Tc for 173 samples (Johnson, 2010). However, these exhibited lower concentrations again.

Table 9: Summary statistics for Technetium data (concentration in pCi/g)

Statistic	Data Source		
	SRS-2002	ES-2010 (January)	Utah-2010
Number of Samples	33	11	173
Mean	49,370	17,800	4,340
Standard Deviation	29,260	5,910	3,550

The pattern of ⁹⁹Tc concentrations in the SRS-2002 data is similar to the pattern seen in the uranium data. That is, the concentrations are considerably greater in the last 12 samples (particularly in the last 9 samples) than in the first 21 samples, by sample ID (see Table 13). This could be reason to exclude the SRS-2002 ⁹⁹Tc data from the distribution development. The data do not seem to come from one population, possibly because of sampling or laboratory differences or biases, and the pedigree of the data is lacking because there are no laboratory reports available for the data. However, these data have been included because they show greater concentrations than the two datasets from 2010, which causes the developed distribution of ⁹⁹Tc concentrations to extend out to cover the SRS-2002 data. The effect of the inclusion of these data has been tested during model evaluation and is reported as part of the sensitivity analysis. If, as might be expected, the ⁹⁹Tc concentrations are a sensitive part of the model, then it might warrant reconsideration of the available data.

Of further concern is the difference between the ES-2010 data and the Utah-2010 data. These data were collected less than a year apart, and several of the samples from the Utah-2010 data were from the same drums used for the ES-2010 samples. The only clear difference between the two datasets is that different analytical laboratories were used in each case. The ES-2010 samples were analyzed by GEL Laboratories. The Utah-2010 samples were analyzed at a different laboratory. It is possible that the differences are analytical.

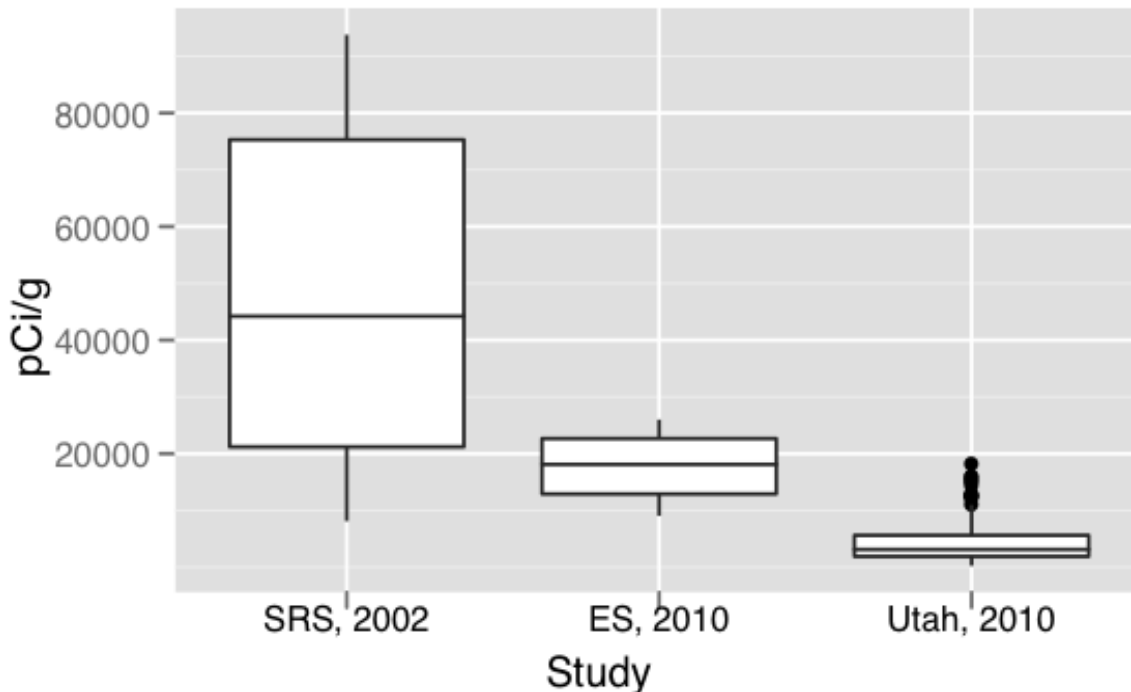


Figure 3. Tc-99 Activity Concentration. Sample sizes: SRS-2002 = 33; ES-2010 = 11; Utah-2010 = 173.

As a consequence of the differences in ^{99}Tc concentrations between the different sampling events, the approach taken to development of an input distribution of mean ^{99}Tc concentrations is similar to the one used for uranium. That is, it is considered more important to model the information content in the sampling events rather than each individual sample. This approach reduces the effect of the Utah-2010 data, which would otherwise dominate estimation of the input distribution.

A simple approach to distribution development is to treat each measurement across all three sampling events as independent and identically distributed and calculate the mean and standard error using all the data. However, this approach weights the data based on the number of samples, giving the Utah-2010 data the most influence. Further, to the extent that the data within each study are not independent, the standard error would be artificially small. The individual data points might not be independent because analyses were often performed on samples from the same drum. To address these issues, a bootstrap method was developed and used to estimate the distribution of the mean ^{99}Tc value that treats the three datasets as independent, rather than each data point across sampling events.

Note that the Utah-2010 dataset contains 18 laboratory and 12 field duplicate measurements. These data were examined and found to be correlated with the associated primary samples.

Since these measurements cannot be considered independent and a relatively large number of samples (173) were analyzed, the duplicates are not included in this distribution analysis.

The three datasets are treated independently in the bootstrap approach, which leads to a wide distribution that covers the range of all three datasets combined. The more simple approach of treating each data point as independent across the three sampling events would result in a very narrow distribution, because of the large number of data points, and the center of the distribution would be lower because the Utah-2010 dataset would dominate given the large sample size.

The bootstrap method is applied as follows:

1. The three studies are selected with replacement from the three available sources of ^{99}Tc data (SRS-2002, ES-2010 and Utah-2010). Since there are only 27 possible combinations of sampling events, all combinations were used and weighted equally.
2. For each study, the data are sampled with replacement and a study mean calculated. An overall mean is calculated as an average of the three study means.
3. This simulation is repeated 10,000 times for each of the 27 study combinations, to construct a distribution of the estimated mean concentrations for ^{99}Tc .

The density plot describes the distribution of the overall mean (Figure 4). Because of smoothing in the plotting algorithm, the distribution appears to include negative values, however, the smallest value from the simulations is 3,800 pCi/g. This distribution is reasonably described by a normal distribution, which is used in the PA model (see Table 5). The mean of the distribution is 23,800 pCi/g, and the standard deviation is 11,000 pCi/g. In the PA model, the distribution is truncated at zero, so that negative mean concentrations are not possible. Since this is a distribution of the mean concentration, this distribution indicates that the mean concentration of ^{99}Tc could be as low as zero, or greater than 60,000 pCi/g (see Figure 4). This is a large range, and reflects the uncertainty in the three data sources because of their differences. Different decisions regarding combination of the available data would almost certainly lead to a narrower distribution of the mean concentration, given the large number of data points available. For example, if the Utah-2010 data were used alone, then the 173 data points would lead to a mean of about 4,340 pCi/g and a standard error of about 270 pCi/g, which is the distribution that would then be used in the PA. That is, most of the distribution of the mean concentration would fall between 3,800 pCi/g and 4,880 pCi/g. This is very different than the distribution that is currently proposed for use in the PA.

Note in Figure 4 that the mean concentrations for the three data sources are also presented. These show clearly that the distribution of the mean ^{99}Tc concentration spans the means of the available datasets. As noted above, if the mean ^{99}Tc concentrations proves to be sensitive for any given endpoint of the PA model (dose, groundwater concentrations, or deep time concentrations), then the development of this input distribution should be revisited, including a re-examination of how the three data sources have been combined. Given the mobility of ^{99}Tc and the width of the input distribution defined above, it is reasonable to expect that concentration of ^{99}Tc will be a sensitive parameter.

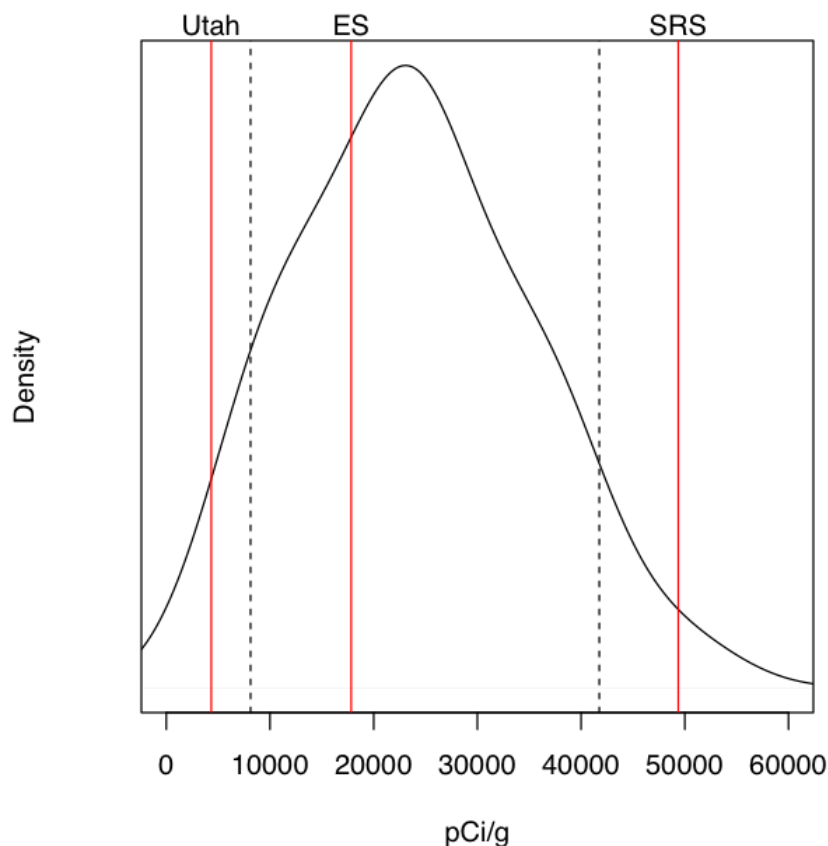


Figure 4. Distribution of Tc-99 mean values. Red lines indicate mean values of Utah-2010, ES-2010, and SRS-2002.

3.4 Concentrations of Other Radionuclides in the SRS Depleted Uranium

As noted in Section 2.1, there are other potential contaminants in the SRS DU, including decay, activation and fission products (see Table 3). Given the only source of data for these radionuclides in SRS-2002, the concentrations are very low, and are unlikely to significantly contribute to the PA, however, input distributions for the mean concentrations of each of these radionuclides are developed and included in the PA to confirm that this is the case.

The measurement of other radionuclides is reported only in the SRS-2002 dataset. These include ^{241}Am , ^{226}Ra , ^{137}Cs , ^{90}Sr , ^{237}Np , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu and ^{129}I . Distributions of these values are shown in Figure 5. With the exception of the plutonium isotopes, all measurements were below the detection limit. Non-detects were set to their detection limits for this analysis. This is a conservative approach, which over-estimates the activity concentrations of these radionuclides. However, the impact of these radionuclides on the PA is likely to be very small, in which case use of the detection limits probably has insignificant effect on the concentrations and doses output by the PA model.

The final distributions are presented in Table 6. The distributions are assumed to be normal, and they are truncated at zero in the PA model.

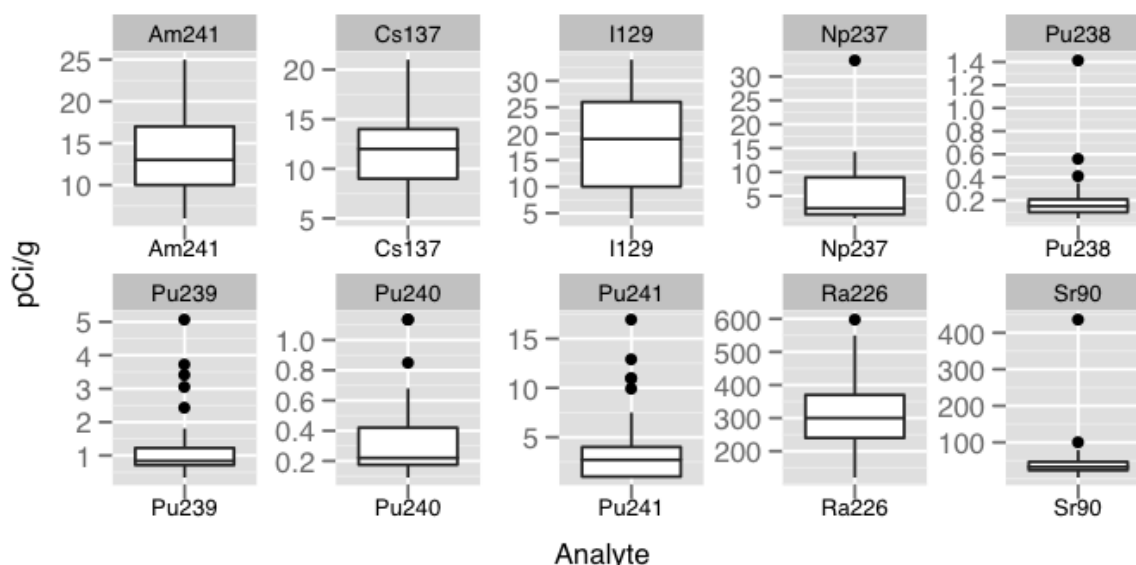


Figure 5. Additional radionuclide data (SRS-2002). Sample size = 33.

3.5 Parameters for Depleted Uranium Oxide from the GDPs

The exact nature of the DU oxides that will be generated by the deconversion plants at Portsmouth and Paducah will not be known until their production, so this PA relies on the best information available to develop estimates. What is known is that the oxides will be primarily U_3O_8 , and that they will be shipped and disposed in used DUF_6 cylinders, some of which will contain residual contamination from reactor returns.

3.5.1 Mass of GDP DU

The total mass of anticipated GDP DU oxide is estimated from the reported mass of DUF_6 currently residing in the cylinder yards and a mass conversion from DUF_6 to DU_3O_8 .

Although the exact number of cylinders at each facility varies from day to day, the Depleted Uranium Management Information Network reports the numbers as 36,191 at Paducah, 16,109 from the Portsmouth GDP, and 4,822 from the K-25 GDP, now moved to Portsmouth (DOE, 2010). However, there are discrepancies in the available information regarding the numbers of cylinders. Consequently, these numbers are used only for rough estimates of the volume needed for disposal.

Estimates of the total mass of DUF_6 from each of the GDPs is also provided at the Depleted UF_6 Management Information Network web site (DOE, 2010). These estimates are 436,400 Mg for Paducah, 195,800 Mg for Portsmouth, and 54,300 Mg for the K-25 GDP, now stored at Portsmouth. These estimates are used in the PA model. No uncertainty is assigned to them. They are a condition of the PA model until more information is made available. Uncertainty is,

instead, included in the concentration estimates, which serves as a reasonable measure in this PA model for inventory uncertainty

3.5.2 Composition of GDP DU

As of this writing, only a single cylinder of oxide has been produced from the deconversion plants, and only one sample from that cylinder has been analyzed. The DUF_6 processed for this sample was of low ^{235}U assay, and contained no TRU or fission product contaminants, and is therefore not representative of the entire populations of GDP DU oxides.

The GDP DU is considered to have two distinct compositions: Clean DU is pure uranium, derived from natural sources, and Contaminated DU includes at least some TRU and fission products from reactor returns. Each of these is discussed below, and the fraction of the total that is contaminated is estimated for use in the PA model.

3.5.2.1 Clean GDP DU

The constituents comprising the clean DU are naturally-occurring isotopes of uranium, significantly depleted in everything but ^{238}U , and whatever decay products may have developed in the short time since their purification and separation. No quantitative information is available about the relative abundance of the uranium isotopes that characterizes the entire waste stream. Given the lack of definitive information about the relative abundances of the uranium isotopes, it is assumed that Clean DU from the GDPs shares the same uranium composition as the DU from SRS. The same isotopic abundances and contaminant concentrations developed for the SRS DU in Section 3.2.3 are therefore applied to the uranium fraction of GDP DU cylinders.

3.5.2.2 Contaminated GDP DU

No quantitative information is available about the contamination of the GDP DU Cylinders, other than limited research determining that some are contaminated and some are not. Given the lack of definitive information about the degree of contamination, it is assumed that contaminated DU from the GDPs shares the same composition as the DU from SRS. The same isotopic abundances and contaminant concentrations developed for the SRS DU in Section 3.2.3 are therefore applied to the contaminated fraction of GDP DU cylinders. There are no other data that are available at this time. The processes under which the DU waste is generated is similar in both case, with material being processed in a diffusion cascade. In both cases the cascades were contaminated, and this is the source of the contaminants in the DU. Without further information on the contamination concentration levels, use of the SRS DU contaminant concentrations is the only information available, even though it is surrogate information.

3.5.2.3 Fraction of Contaminated GDP DU

Assuming that each GDP cylinder is either “clean” or “contaminated”, an estimate is needed for the number of each type, so that the total amount of contaminant radionuclides in the GDP inventory can be estimated. At the time of this writing, the best available information about this comes from a study by Henson (2006): DUF6-G-G-STU-003 (Draft for UDS review). This document reviews information about the Paducah population of cylinders as recorded on cylinder history cards, which were used until 1988, and all contaminated cylinders are represented in this

population. Table 1 (reproduced here as Table 10) in Henson (2006) categorizes the cylinders as follows:

- "Category 1 – 13,240 cylinders: Cleared" cylinders, which are not contaminated,
- "Category 2 – 1,335 cylinders: TRU and/or Tc" cylinders, which are confirmed to have some degree of contamination,
- "Category 3 – 971 cylinders: >1% U235" cylinders, which do not contain DU and so are not considered in this PA, and
- "Category 4 – 22,382 cylinders: To Be Determined" cylinders which have unknown status regarding contamination. 9,407 of these cylinders have history cards and 12,975 do not.

Note that these values are in numbers of cylinders, rather than mass of DU, so an assumption is made for the purposes of estimating the fraction of waste that is contaminated that each cylinder contains the same mass of DU. Note also that the total number of cylinders here is not the same as the number of cylinders suggested in Section 3.5.1. This reflects both uncertainty in the total number of cylinders, and the change in number through time as cylinders are reprocessed or transferred.

The Paducah data can be summarized as follows for the purposes of building a distribution for the fraction of cylinders that are contaminated:

- 13,240 are known to not be contaminated
- 1,335 are known to be contaminated
- Of the unknowns 9,407 have history cards, and, hence, can be considered part of the same population of reconciled cylinders. These are assumed to be pre-1988 cylinders.
- Of the unknowns, 12,975 do not have history cards. These are post-1988 cylinders.

The cylinder history card system at Paducah was discontinued May 31, 1988 (Henson, 2006). Paducah cylinders post-1988 are considered much more likely to be clean of contaminants. Consequently, unknown cylinders are modeled differently for pre-1988 and post-1988.

The cylinders at Portsmouth also need to be considered. The Depleted Uranium Management Information Network reports the numbers as 16,109 from the Portsmouth GDP, and 4,822 from the K-25 GDP, now moved to Portsmouth (DOE, 2010). These cylinders are also considered unlikely to be contaminated (personal communication, Tammy Stapleton, April 2011).

This completes the summary of the population of cylinders that are considered for disposal at the Clive facility. The available information is used to construct an estimate of the total fraction of the cylinders that are contaminated. In effect the proportion contaminated at Paducah for the cylinders that have known status is used as an estimate of the fraction of all cylinders with history cards that are contaminated. These are presumed to be all of the pre-1988 cylinders. For the post-1988 cylinders at Paducah, which have no history cards, and the Portsmouth cylinders, a much smaller fraction of the cylinders is assumed to be contaminated.

Consequently, the fraction of Pre-1988 cylinders at Paducah that is assumed to be contaminated is about 9% [$1,335 / (1,335 + 13,240)$]. The Portsmouth cylinders might also have a small fraction that are contaminated. Using expert opinion, this is estimated at less than 1%, with a best guess at no more than 10 cylinders contaminated (personal communication, Tammy Stapleton, April 2011). These values were interpreted as expert judgment of the 95th and 50th percentiles of the distribution, respectively. A beta distribution was fit to these values, following the procedures outlined in the Fitting Probability Distributions white paper.

The total number of contaminated cylinders was then simulated by adding the number of confirmed contaminated cylinders with simulated numbers for the unknown cylinders. Table 11 shows the inputs that were used for the simulations. A distribution was constructed based on the simulation output for the overall proportion of cylinders that are contaminated. This Beta(0.0392, 0.0025) probability density function is shown in Figure 6.

In terms of the number of contaminated cylinders, this distribution has 1st, 50th, and 99th percentiles of 1,946, 2,266, and 2,619, respectively. This is a fairly narrow distribution given the lack of information available. It is narrow because nearly 15,000 of the Paducah cylinders have been characterized, an assumption is made that all other pre-1988 cylinders will show a similar ratio, and the remaining cylinders are expected to be clean of contamination. As more information is gathered when the depleted uranium is prepared for disposal, then input distributions used to characterize the GDP waste should be revisited. Information that will be needed will include total amount of DU, chemical speciation of DU, and activity concentrations of the DU and contaminants.

Table 10: Categorization of Paducah Cylinders Using Cylinder History Cards (reproduced from Table 1 in Henson, 2006)

Category 1: Cleared	Category 2: TRU and/or Tc	Category 3: >1% ²³⁵ U	Category 4: To Be Determined
Filled once with natural normal or depleted material. (9,728)	Never filled with 1% or greater assay, but have a history of containing recycled feed material. These cylinders may have "hidden heels" containing both transuranics (TRU) and Tc. (1,334)	Filled at some time with material >1% assay, and also used to contain recycled material. These cylinders may have "hidden heels" containing both transuranics (TRU) and Tc. (584)	No Paducah history card. (12,975)
Filled more than once, but only with natural normal or depleted material. (2,681)	No history of recycled feed service, but used to hold Paducah product (at <1% enrichment). These cylinders may also have "hidden heels" which could contain Tc. (1)	No history of recycled feed service, but used to hold Paducah product (at >1% enrichment). These cylinders may also have "hidden heels" which could contain Tc. (387)	History card does not provide enough information. (9,407)
Washed and subsequently filled with only natural normal or depleted material. (832)		Filled at some time with >1% assay, but have never contained recycled uranium or Paducah product. (n/a for Phase II)	
TOTAL = 13,240	TOTAL = 1,335	TOTAL = 971	TOTAL = 22,382

Table 11: Inputs for the Simulation of the Fraction of Contaminated GDP Cylinders

Cylinder Type	Paducah Category 2	Paducah Category 1	Paducah Category 4 Pre-1988	Paducah Category 4 Post-1988	Portsmouth (not from Oak Ridge)	Portsmouth (from Oak Ridge)
Number	1,335	13,240	9,407	12,975	16,109	4,822
Simulated Binomial Proportion	NA (confirmed value)	NA (confirmed value)	Beta(0.092, 0.0024)	Beta(0.0020, 0.0042)		

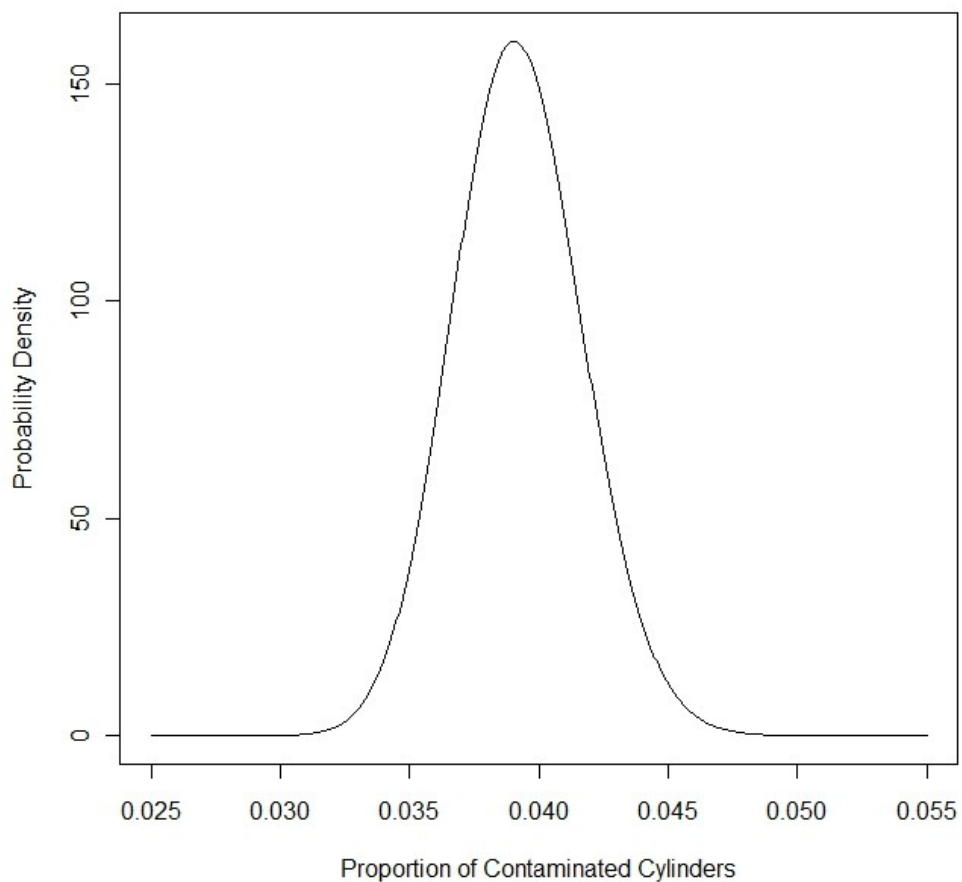


Figure 6. Probability density function for the proportion of contaminated cylinders.

4.0 References

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Appendix

Table 12. Uranium isotopic abundances by mass spectrometry, atomic percent, including replicates (data summarized in Table 16, Beals, et al. 2002)

Sample	Replicate	²³⁴ U	²³⁵ U	²³⁶ U	²³⁸ U
3	a	0.0004%	0.1270%	0.0065%	99.87%
3	b	0.0004%	0.1260%	0.0065%	99.87%
9	a	0.0004%	0.1260%	0.0064%	99.87%
9	b	0.0004%	0.1250%	0.0064%	99.87%
17	a	0.0004%	0.1260%	0.0066%	99.87%
17	b	0.0004%	0.1260%	0.0066%	99.87%
20	a	0.0005%	0.1270%	0.0068%	99.87%
20	b	0.0004%	0.1290%	0.0067%	99.86%
25	a	0.0008%	0.1510%	0.0096%	99.84%
25	b	0.0007%	0.1510%	0.0095%	99.84%
30	a	0.0006%	0.1410%	0.0088%	99.85%
30	b	0.0006%	0.1400%	0.0086%	99.85%

Table 13. Uranium isotopic abundances by alpha spectrometry (as percent of total uranium activity) (Table 17, Beals, et al. 2002) and Technetium concentrations in the SRS-2002 data (Beals, et al. 2002)

Sample	²³⁸ U	²³⁵ + ²³⁶ U	²³⁴ U	⁹⁹ Tc (nCi/g)
1	91.7	1.72	6.57	44.2
2	91.0	1.74	7.28	57.5
3	91.3	2.04	6.63	21.2
4	91.3	1.86	6.82	33.3
5	91.6	1.73	6.67	15.7
6	91.2	1.76	7.07	19.1
7	91.2	1.85	6.91	18.5
8	91.6	1.71	6.67	24.5
9	91.3	1.98	6.72	90.2
10	91.8	1.7	6.55	79.7
11	91.6	1.7	6.75	89.8
12	91.8	2.04	6.18	79.7
13	91.3	1.95	6.74	37.5
14	91.2	1.7	7.09	75.3
15	91.6	1.74	6.63	34.2
16	91.4	1.86	6.7	74.2
17	91.2	2.07	6.7	41.4
18	91.4	1.86	6.71	64.7
19	91.7	1.97	6.32	16.1
20	90.8	2.25	6.92	14.9
21	91.6	1.73	6.69	27.2
22	87.5	2.11	10.42	8.1
23	88.4	2.11	9.46	15.7
24	85.9	2.51	11.55	9
25	86.9	2.41	10.71	93.8
26	86.7	2.36	10.9	92.7
27	87.3	2.27	10.41	32.5
28	88.0	2.26	9.72	55.3
29	87.3	2.84	9.91	53.8
30	88.5	2.27	9.2	88.5
31	85.9	2.77	11.32	93.7
32	88.6	2.8	8.61	54.3
33	88.2	1.83	9.99	73
Mean	90	2.05	7.99	49.37
Std.Dev	2.03	0.34	1.77	29.26

Table 14. January 2010 EnergySolutions Data Analyzed by GEL (GEL 2010a and 2010b)

Sample ID	bulk density (g/cm ³)	⁹⁹ Tc (pCi/g)	total uranium (µg/g)	²³³⁺²³⁴ U (pCi/g)	²³⁵⁺²³⁶ U (pCi/g)	²³⁸ U (pCi/g)
243721001	3.31	2.28E+4	7.93E+5	4.84E+4	1.11E+4	2.65E+5
243721002	3.45	9.78E+3	8.54E+5	4.50E+4	7.21E+3	2.86E+5
243721003	2.84	1.78E+4	8.06E+5	3.83E+4	1.89E+4	2.68E+5
243721004	3.15	9.04E+3	8.27E+5	3.26E+4	4.92E+3*	2.77E+5
243721005	2.50	1.44E+4	8.48E+5	4.25E+4	7.27E+3*	2.85E+5
243721006	3.21	2.08E+4	8.80E+5	3.04E+4	1.28E+4	2.94E+5
243721007	4.00	2.25E+4	9.90E+5	6.44E+4	1.28E+4	3.31E+5
243721008	2.36	1.14E+4	6.50E+5	3.37E+4	1.19E+4	2.17E+5
244495001	3.46	2.60E+4	8.44E+5	3.57E+4	6.72E+3	2.83E+5
244495002	3.66	2.35E+4	8.00E+5	3.65E+4	1.17E+4	2.67E+5
244495003	4.00	1.81E+4	8.76E+5	4.70E+4	9.94E+3	2.93E+5
Mean	3.27	1.78E+4	8.33E+5	4.13E+4	1.15E+4	2.79E+5
Std.Dev	0.54	5.91E+3	8.15E+3	9.73E+3	3.57E+3	2.74E+3

* - reported as non-detects – detection limits used for statistical analysis.

Table 15. April 2010 EnergySolutions Data Analyzed by GEL (GEL 2010c)

Sample ID	bulk density (g/cm ³)	⁹⁹ Tc (pCi/g)	total uranium (µg/g)	²³³⁺²³⁴ U (pCi/g)	²³⁵⁺²³⁶ U (pCi/g)	²³⁸ U (pCi/g)
249710001	-	-	7.95E+5	3.42E+4	6.34E+3	2.66E+5
249710002	-	-	8.31E+5	3.65E+4	6.31E+3	2.78E+5
249710003	-	-	8.15E+5	3.35E+4	5.12E+3	2.73E+5
249710004	-	-	8.74E+5	3.84E+4	5.17E+3	2.93E+5
249710005	-	-	8.28E+5	3.66E+4	4.30E+3	2.78E+5
249710006	-	-	8.74E+5	4.17E+4	4.31E+3	2.93E+5
249710007	-	-	7.07E+5	2.94E+4	4.86E+3	2.37E+5
249710008	-	-	6.46E+5	3.78E+4	4.43E+3	2.17E+5
249710009	-	-	7.42E+5	3.66E+4	6.80E+3	2.48E+5
249710010	-	-	7.97E+5	3.86E+4	4.95E+3	2.67E+5
249710011	-	-	8.29E+5	3.51E+4	4.36E+3	2.78E+5
249710012	-	-	7.58E+5	2.98E+4	7.60E+3	2.54E+5
249710013	-	-	7.45E+5	3.16E+4	4.89E+3	2.50E+5
249710014	-	-	7.71E+5	3.09E+4	4.14E+3	2.58E+5
249710015	-	-	8.97E+5	4.02E+4	5.75E+3	3.01E+5
Mean			7.88E+5	3.54E+4	5.34E+3	2.64E+5
Std.Dev.			6.61E+4	3.60E+3	1.07E+3	2.21E+4

Table 16. Technetium-99 concentrations collected by State of Utah, (Johnson, 2010)

Sample ID	pCi/g	Sample ID	pCi/g	Sample ID	pCi/g
1337	6.30E+3	3800	1.24E+4	0249	3.28E+3
1348	1.27E+4	3824	5.59E+3	0370	4.77E+3
1423	2.13E+3	3849	4.13E+3	0434	2.80E+3
1428	3.45E+3	3857	2.56E+3	0461	4.09E+3
1429	7.05E+3	3870	1.55E+4	0488	3.09E+3
1467	2.66E+3	3951	1.79E+3	0499	8.22E+2
1584	3.50E+3	4052	2.07E+3	0555	1.12E+3
1622	7.99E+3	4104	2.44E+3	0562	2.08E+3
1697	3.09E+3	4138	4.23E+3	0565	7.19E+3
1712	5.21E+3	4162	3.51E+3	0571	4.11E+3
1739	5.62E+3	4172	6.85E+3	0626	1.78E+3
1794	2.74E+3	4185	2.64E+3	0629	4.41E+3
1808	2.54E+3	4207	2.01E+3	0662	2.74E+2
1834	1.53E+4	4244	1.56E+3	0670	1.95E+3
1835	7.12E+3	4275	1.22E+3	0697	1.63E+3
1853	2.49E+3	4303	8.86E+2	0739	2.37E+3
1876	1.47E+3	4322	1.01E+3	0756	3.56E+3
1918	2.90E+3	4362	3.06E+3	0800	1.57E+3
1946	2.08E+3	4376	6.66E+3	0809	5.73E+2
2061	1.84E+4	4384	2.32E+3	0813	2.22E+3
2077	1.83E+3	4385	9.72E+3	0852	4.45E+3
2098	1.10E+4	4393	3.58E+3	0853	2.31E+3
2102	7.65E+2	4414	3.78E+3	0854	2.83E+3
2140	7.86E+3	4415	8.86E+3	0879	4.52E+3
2250	6.71E+3	4425	5.87E+3	0884	4.76E+3
2256	7.19E+3	4431	1.29E+4	0893	2.02E+3
2343	1.30E+3	4486	5.83E+3	0910	2.24E+2
2424	6.27E+2	4487	2.63E+3	0911	8.23E+2
2449	4.86E+3	4504	8.48E+3	0927	6.38E+2
2481	1.32E+3	4535	5.25E+3	0928	7.42E+2
2497	1.62E+4	4606	1.72E+3	1000	5.85E+3
2517	8.06E+2	4611	3.47E+3	1021	1.24E+3
2528	1.66E+3	4687	1.51E+3	1030	1.63E+3
2550	3.02E+3	4760	3.04E+3	1117	6.56E+3
2614	1.49E+3	4790	2.28E+3	1140	1.76E+3
2674	1.89E+3	4817	2.25E+3	1147	1.29E+3
2675	2.92E+3	4822	2.62E+3	1216	1.44E+3
2823	4.89E+3	4851	1.32E+4	1505	2.26E+3

Sample ID	pCi/g	Sample ID	pCi/g	Sample ID	pCi/g
2827	1.61E+4	4866	1.45E+4	1511	3.96E+3
2878	2.86E+3	4940	4.41E+3	1646	6.19E+3
3035	7.59E+3	4955	3.68E+3	1678	1.00E+4
3059	5.01E+3	4962	5.89E+3	2393	4.08E+3
3067	1.77E+3	5023	1.89E+3	2657	5.52E+3
3080	4.36E+3	5054	2.36E+3	2693	1.97E+3
3085	1.53E+3	5061	1.68E+3	3127	3.31E+3
3089	2.37E+3	5084	6.22E+3	3160	6.34E+3
3197	2.28E+3	5191	1.13E+4	3288	7.08E+3
3234	4.62E+3	5224	5.75E+3	3336	5.12E+3
3303	5.61E+3	5277	1.58E+3	3337	5.37E+3
3347	5.53E+3	5322	7.33E+2	3446	3.25E+3
3543	1.67E+3	0023	3.89E+3	3471	2.86E+3
3668	3.12E+3	0057	1.15E+3	3546	4.73E+3
3685	3.03E+3	0157	1.28E+3	4016	1.09E+4
3695	7.46E+3	0162	6.53E+3	4098	8.93E+3
3717	4.56E+3	0168	3.42E+3	4200	1.68E+3
3726	4.94E+3	0180	2.80E+3	4514	3.28E+3
3728	1.28E+4	0210	3.72E+3	4581	1.81E+3
3760	2.38E+3	0214	2.24E+3		

Number of samples = 173

Average ⁹⁹Tc concentration = 4,340 pCi/g

Standard Deviation = 3,550 pCi/g

Table 17. Concentration data for other radioisotopes, SRS 2002. (Beals, et al. 2002)

Sample	²⁴¹ Am *(pCi/g	²²⁶ Ra *(pCi/g	¹³⁷ Cs *(pCi/g	⁹⁰ Sr *(pCi/g	²³⁷ Np pCi/g	²³⁸ Pu pCi/g	²³⁹ Pu pCi/g	²⁴⁰ Pu pCi/g	²⁴¹ Pu pCi/g	¹²⁹ I *(pCi/g
1	6	120	6	8.6	0.44	0.114	0.53	0.14	2.80	13
2	24	500	19	5.9	2.34	0.099	0.69	0.15	nd	7
3	21	450	17	3.4	0.33	0.065	0.48	0.12	1.00	7
4	17	330	14	6.7	4.61	0.129	0.84	0.17	nd	4
5	25	600	20	7.2	12.8	0.086	0.95	0.23	2.50	12
6	20	390	15	14	8.89	0.163	0.40	0.10	nd	10
7	16	314	13	8	14.3	0.090	0.34	0.10	1.60	9
8	16	310	12	7.7	3.85	1.420	0.91	0.48	10.00	4
9	10	240	9	50.7	6.52	0.350	3.43	1.14	11.00	8
10	21	470	19	32.7	2.43	0.244	0.48	0.18	3.80	6
11	16	370	14	23.4	13.6	0.240	3.10	0.68	13.00	20
12	11	250	10	29.3	11.9	0.090	1.15	0.29	2.70	14
13	11	260	10	46.6	8.55	0.230	5.09	1.14	17.00	18
14	13	340	12	31.2	1.3	0.123	2.46	0.55	7.50	20
15	17	360	13	40	6.38	0.127	0.36	0.09	0.90	16
16	12	300	11	68.2	33.5	0.099	0.66	nd	nd	16
17	11	230	10	28.4	6.08	0.125	1.63	0.50	4.00	17
18	11	230	8	38.3	2.86	0.081	0.75	0.20	nd	19
19	10	210	7	51	10.2	0.043	3.74	0.86	11.00	26
20	6	170	5	45.6	11.3	0.088	1.07	0.27	nd	32
21	14	300	13	27.1	1.92	0.094	0.50	0.12	1.10	33
22	9	250	8	28.6	0.77	0.149	0.81	0.22	3.40	27
23	18	380	15	45.7	1.67	0.186	1.81	0.52	5.30	24
24	16	340	13	26.9	0.69	0.242	1.30	0.36	nd	27
25	13	280	11	45.7	1.18	0.178	0.88	0.24	2.60	26
26	9	250	9	100.5	0.65	0.560	0.79	0.22	2.70	7
27	10	280	10	59.1	0.94	0.181	0.79	0.22	2.80	30
28	25	550	21	28	1.61	0.154	0.74	0.21	3.40	34
29	16	410	14	57.9	11.1	0.420	0.79	0.18	nd	24
30	10	190	10	32.9	0.87	0.123	0.85	0.22	4.00	27
31	16	350	15	78.9	1.04	0.250	1.02	nd	nd	26
32	9	190	7	438.2	1.32	0.155	1.09	0.32	2.50	22
33	9	240	9	35.8	1.58	0.153	0.82	0.24	1.70	28

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