Pacific Northwest National Laboratory

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Methodology for Uncertainty Estimation of Hanford Tank Chemical and Radionuclide Inventories and Concentrations

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Summary

The exact physical and chemical nature of 55 million gallons of toxic waste held in 177 underground waste tanks at the Hanford Site is not known with sufficient detail to support the safety, retrieval, and immobilization missions presented to Hanford. The Hanford "Best Basis" team has made point estimates of the inventories in each tank. The purpose of this study is to estimate probability distributions for each of the analytes and tanks that the Hanford "Best Basis" team has made point estimates for. This will enable uncertainty intervals to be calculated for the "Best Basis" inventories and should facilitate the safety, retrieval, and immobilization missions.

The methodology presented in this paper is based on scientific principles, sound technical knowledge of the realities associated with the Hanford waste tanks, the Hanford "Best Basis" research, chemical analysis of samples from the tanks, and historical data. The method builds on research conducted by PNNL over the last few years. As a result of the processing histories, waste storage practices and historical records, some of the waste can be partitioned into more homogeneous subsets that can be identified to tanks and locations within tanks. Therefore, by using this we can maximize the information extracted from the relatively few samples we have for each tank, combining sample data information from similar tanks to generate a sample based estimate of the chemical and radionuclide concentrations of each of the many waste subsets. Then by multiplying the concentrations for a specific constituent (micrograms of the constituent per gram of waste, or microCuries of the constituent per gram of waste) by the density (grams per liter) by the volume in a tank (liters), we can get an estimate of the mass of a particular constituent (or radionuclide contribution) in each tank. This methodology does that but does it in a probabilistic framework; thus using and getting probability distributions instead of single numbers.

The approach partitions the waste into the following three groups:

- **Sludges** are residual solids that settled out of the waste slurries, often remaining in the same tank to which they were first transferred. Drainable interstitial liquids are considered to be a part of sludges. Four saltcakes are also included in this group: B, BY, T1, and R saltcakes due to the treatment used by the LANL HDW model. (Separation of these saltcakes from the sludges can be accomplished after the methodology has provided concentration and inventory probability distributions.
- **Supernatants** are liquid layers in the tanks.
- **Saltcakes** are solids formed as precipitates from supernatant, often as a result of cooling after the waste completes the evaporator process.

A probability distribution is generated for each of these waste phases based on "multiplying" the probability distributions for: waste concentrations, density, and volume of waste. The distribution of total inventory of constituent A in tank T is estimated by:

$$f_{I, A, T} = f_{I\text{Sludge}, A, T} \oplus f_{I\text{Saltcake}, A, T} \oplus f_{I\text{Supernatant}, A, T} \quad (\text{S.1})$$

where

- $f_{I\text{Sludge}, A, T}$ = sludge inventory probability distribution of constituent A in Tank T
- $f_{I\text{Saltcake}, A, T}$ = saltcake inventory probability distribution of constituent A in Tank T
- $f_{I\text{Supernatant}, A, T}$ = supernatant inventory probability distribution of constituent A in Tank T
- $\oplus$ = a summation of two probability distributions.

Each term on the right-hand-side of equation S.1 is estimated as the probability distributions of the product of three probability distributions: concentration, density and volume.

Concentrations. The probability distributions of concentrations for analytes that are measured frequently, such as Al, Bi, and Cr, are estimated in two ways depending on the waste group. For sludge wastes, we estimate the concentrations of waste types that are derived from sample data clustering and relate closely to the waste types used by the "Best Basis" and the Los Alamos National Laboratory (LANL) Hanford defense waste (HDW) model teams. All chemical assay data from all the aliquots that can be associated with each waste type form an empirical probability distribution for that waste type. The estimated concentration distributions by waste type can be applied to both sampled and unsampled tanks. For saltcake and supernatant wastes, we cluster data from the HDW supernatant mixing model (SMM) concentrations to group tanks together and then use an analysis of variance (ANOVA) to combine data appropriately and form probability distributions for concentrations of each analyte for the saltcake and supernatant wastes in each tank.

For those analytes with little or no measured data, we estimate the probability distributions of the concentrations to be centered at the "Best Basis" estimates and that reflect the variability indicated by "analog" analytes that have adequate data to form a probability distribution.

Density. Probability distributions for the density of the waste (g/L) are estimated in the same manner as the chemical analytes. This is possible since density is often measured as part of the chemical assay process.

Volume. The probability distributions of volume for each tank are needed for sludge volume by waste type, total supernatant volume, and total saltcake volume. The saltcake volume distribution is derived using total waste volume.

The approach to estimate the probability distributions of total waste volumes partitions the waste in each tank into volume elements (voxels), estimates the volume probability distributions for these voxels and then "sums" the probability distributions. The variability associated with top location, bottom location, and cross-area of each voxel is considered in the estimation. The total waste-volume estimates will exclude retained gas volumes in the tanks. PNNL and the Hanford community have conducted various studies estimating the volume of gas trapped in the waste.

LANL personnel (Steve Agnew's team) are preparing estimates of the probability distribution of volume for each waste type in each tank. These will be used to estimate the probability distribution of the inventory and concentrations associated with the tank layer model (TLM) solids waste. Estimates of the amount of supernatant in the tank can be based on the information in the "Best Basis" documents, TCR/TCO, Hanlon reports, and current collected data. We envision an approach similar to the approach proposed for the total-volume estimation, but which allows for measuring parameters that separate the sludge and supernatant. Our current approach for estimating the probability distributions for saltcake volumes is by "subtraction" of the probability distributions of total waste, sludge, and supernatant.

We are continuing to develop and implement the methodology. Nuances discovered in the available data may require adjustments to the methodology. Simpler or more effective algorithms may be developed that warrant modifying the methodology. And of course a major purpose of this report is to provide a structure for discussing the methodology for additional refinements. As such, the methodology implemented and used to generate the final uncertainty numbers will incorporate refinements and improvements.

List of Acronyms

ANOVA	analysis of variance
cdf	cumulative distribution function
DST	double-shell tank
FIC	Food Instrument Company
HCA	hierarchical cluster analysis
HDW	Hanford defense waste
HEABB	Hanford Engineering Analysis Best Basis
LANL	Los Alamos National Laboratory
LMHC	Lockheed Martin Hanford Company
LOCID	location identification
pdf	probability density function
REML	restricted maximum likelihood
SMM	supernatant mixing model
SOW	statement of work
SST	single-shell tank
TCD	tank characterization database
TCR	tank characterization report
TLM	tank layer model
TWINS/TCD	Tank Waste Information System/Tank Characterization Database
WSTRS	waste status and transaction records summary

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1.0 Introduction

1.1 Objective

The purpose of this effort by Pacific Northwest National Laboratory¹ is to estimate uncertainties associated with estimates of concentrations, waste volumes and densities, and total inventories of 71 analytes in the 177 nuclear waste tanks at Hanford. The estimated uncertainties are associated with the Best Basis estimates of the inventories. These estimates were determined by the Hanford Engineering Analysis Best Basis (HEABB) (Kupfer, et. al., 1997) task group from the available sampling data process flow sheet data, waste-type characterizations, historical records of tank transfers (Agnew 1997), and other available information. HEABB estimates are contained in the Tank Characterization Reports (TCRs), published by Westinghouse Hanford Company and Lockheed Martin Hanford Company (LMHC).

During fiscal year (FY) 1997, the generation and publication of the HEABB estimates of tank contents was completed. End-users of the "Best Basis" data have expressed the need for uncertainty estimates to provide bounds on safety analyses, risk assessments, performance assessments, equipment designs, and other projects requiring consideration of tank contents and consequences on facility operations, human health and the environment.

Also during FY 1997, PNNL investigated 4 different approaches to estimating tank contents and their uncertainties for sampled and unsampled analytes. Two of the approaches are of specific interest to this effort. One approach, denoted the "Grouping/ANOVA" approach uses multivariate statistical/mathematical techniques to group the tanks based on estimated chemical concentrations based on the LANL HDW model (Agnew 1997), and then uses sample data where available in an analysis of variance model to estimate tank concentrations and uncertainties. This method was used to estimate the concentrations of the tanks and resulted in prediction intervals for the four T-200 tanks. Sample results from 4 cores (1 from each of the 4 tanks) showed strong agreement to the prediction intervals and thus supported the approach. A second approach investigated in FY 1997, used segment and subsegment sample data to provide a sample based estimate of the Hanford defined wastes used in the LANL HDW model, with the intent to upgrade the inventory and concentration estimates provided by the model. This investigation showed considerable promise. These two FY 1997 investigations inspired the approach pursued this year.

The uncertainties associated with the HEABB estimates can be established for all 177 tanks. The measure of uncertainty depends directly on the amount and quality of information available for a given tank. Of the 177 tanks at the Hanford site, 108 of them have sample data being used in this report. This sampling information, however, varies for a given tank from very limited (e.g., safety screening data, which have few measured analytes) to reasonably comprehensive (many measured analytes). These sampling data are contained in the Tank Waste Information System/Tank Characterization Database (TWINS/TCD) and are accessible online at <http://pctwins.pnl.gov/tcr.nsf>.

1.2 Overview of the Document

This document reports the methodology developed to assess the uncertainties associated with tank-inventory estimates. The methodology described in this document is still under investigation and subject to change. Further development of the methodology will be reported in the year end report for this task.

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Our goal is to complete a matrix of 71 analytes and 177 tanks with estimates for mean, standard deviation, and percentiles for concentrations and inventories on a tank-by-tank basis, based on sample data. To outline the precision level of the uncertainties associated with estimates of concentrations, waste volumes and densities, and total inventories of 71 analytes in the 177 Hanford tanks, we defined a quadrant approach. These quadrants divide the task into four sections (see Table 1.1):

1. Q11 = analytes that are *frequently* measured and present in tanks with *significant* sample data
2. Q21 = analytes that are *frequently* measured and present in tanks with *minimal* sample data
3. Q12 = analytes that are *rarely* measured and present in tanks with *significant* sample data
4. Q22 = analytes that are *rarely* measured and present in tanks with *minimal* sample data.

Table 1.1. Quadrants by Analyte and Tank

	Tank	Frequently Measured Analytes				Rarely Measured Analytes			
		A1	A2	...	...	...	...	...	A71
Tanks with significant sample data	T1								
	T2								
	...			Q11				Q12	
	...	---	---	---	---	---	---	---	---
Tanks with minimal or zero sample data	...								
	...								
	...			Q21				Q22	
	T177								

The entries in the left quadrants, Q11 and Q21, use analysis results of waste samples as the primary source of information. To estimate concentration and density distributions for the tanks in both quadrants, some mechanism is required to extend the samples results from the tanks in Q11 to the tanks in Q21. A waste-type approach is applied to sludge wastes while a grouping-ANOVA approach is applied to supernatant and saltcake wastes to achieve such an extension.

The right half of the matrix, quadrants Q12 and Q22, are filled by identifying relationships between the measured and unmeasured constituents.

Section 2 of this document describes the overall approach used to estimate tank inventory uncertainties. Three major components are considered in this approach: chemical concentration, density, and waste volume. Section 2 also describes the two different methods used to evaluate the tank wastes in terms of sludges and in terms of supernatant or saltcakes. Sections 3 and 4 describe in detail the methodology to assess the probability distributions for each of the three components, as well as the data sources for implementation. The conclusions are given in Section 5.

2.0 Overview of Approach

2.1 Overall Concept

The inventory of constituent A in tank T ($Inv_{A,T}$) can be determined by the following equation

$$Inv_{A,T} = C_{A,T} \times D_T \times V_T \quad (2.1)$$

where

$C_{A,T}$ = mean mass based concentration of constituent A in tank T ($\mu\text{g/g}$)

D_T = mean density of the wastes in tank T (g/L)

V_T = total volume of wastes in tank T (L).

All three quantities for the 177 tanks at the Hanford site can only be estimated through very limited means. Equation 2.1 indicates the three major sources of variability that contribute to the uncertainty of the inventory estimate. Therefore, we need to assess the uncertainty (in terms of probability distributions) associated with the estimate of each component to obtain the uncertainty (in terms of probability distributions) of the tank-inventory estimate.

The tanks contain several waste phases, including sludge, supernatant, saltcake, etc. Wastes in different phases need to be handled separately since the concentrations and densities vary widely from phase to phase. Section 2.2 discusses issues on waste-phase separation. Sections 2.3 and 2.4 outline methodologies to assess the variability of the estimates for the three components in each waste phase. Section 2.5 describes the technique to obtain the uncertainty of total inventory estimate in a tank by combining all component uncertainty following the relationship in Equation 2.1.

2.2 Waste Phase Separation

Three waste phases are considered in this study and defined as following.

1. **Sludges** are residual solids that settled out of the waste slurries, often remaining in the same tank to which they were first transferred. Drainable interstitial liquids are considered to be a part of sludges.

This waste phase corresponds to the sludge wastes tracked in the Tank Layer Model (TLM), which is part of the Hanford Defense Waste (HDW) model (Agnew 1977). To be consistent with the TLM model, four types of saltcakes are also included in the sludge phase: B, BY, T1, and R saltcakes. This consistency is desired, but not critical, for developing sludge-volume distributions by waste type based on the estimates provided in the TLM.

2. **Saltcakes** are solids formed as precipitates from supernatant, often as a result of cooling after the waste completes the evaporator process.
3. **Supernatants** are liquids that stand on top of sludge or saltcake layers in the tanks.

Both saltcake and supernatant wastes are tracked in the Supernatant Mixing Model (SMM), which is another component of the HDW model. In the SMM, however, the two phases are not distinguished. This phase distinction is necessary in this study to match waste sample state, which is typically labeled as solid or liquid in the Tank Characterization Database (TCD).

With the three waste-phase separation approach, the distribution of total inventory of constituent A in tank T is estimated by:

$$f_{i,A,T} = f_{iSludge,A,T} \oplus f_{iSiltcake,A,T} \oplus f_{iSupernate,A,T} \quad (2.2)$$

where f denotes a probability density distribution, and the symbol $\oplus$ indicates a summation of two probability distributions. Each component inventory distribution is estimated following Equation 2.1. The probability distributions of concentration, density, and volume within each waste phase need to be assessed to achieve this goal.

2.3 Approach for Sludges

A waste-type approach is used for estimating sludge waste inventories. The approach assumes that sludges from different plant processes accumulated in the tanks in distinguishable layers. The waste types here are referring to the Hanford Defined Waste types defined in the HDW model. Specifically, the sludge inventory distribution of constituent A in tank T is estimated by:

$$f_{iSludge,A,T} = \sum_i f_{ASludge,W_i} \otimes f_{Di,Sludge,W_i} \otimes f_{Vi,Sludge,T,W_i} \quad (2.3)$$

where

- $f_{iSludge,A,T}$ = sludge inventory distribution of constituent A in Tank T
- $f_{ASludge,W_i}$ = sludge concentration distribution of constituent A in waste type W_i
- $f_{DiSludge,W_i}$ = sludge density distribution of waste type i
- $f_{ViSludge,T,W_i}$ = sludge volume distribution of waste type i in tank T
- $\otimes$ = product operation of two probability distributions.

This approach provides a mechanism to extend sample results to unsampled tanks because waste type composition can be estimated in tanks without sample data from tank-waste transaction records. This approach also produces chemical composition profiles for the HDW waste types.

The variability associated with constituent concentrations by waste type is estimated based on sample data. The waste samples were grouped through a cluster analysis based on set of key constituent concentrations. Using the TLM and other historical process information, the waste types that these sample groups represent were identified. The concentration distributions for each waste type are then developed from the corresponding groups of measurements using a non-parametric distribution estimation technique. Section 3.1 contains a more detailed discussion on how samples were mapped or grouped into waste types and how empirical distributions were developed for concentrations.

Some waste types have not been sampled. To obtain concentration distributions for these waste types, a site-wide variability is applied to each constituent. The site-wide variability is scaled based on the historical estimate of the mean concentration provided in the HDW model.

Among the 71 target constituents, 44 (principally radionuclides) are rarely measured. An analog list (see Section 3.1.2) is developed to extrapolate distributions developed for the analog constituents to the corresponding unmeasured constituents.

For all three situations, the detailed procedures for developing concentration distributions for sludges are described in Section 3.1.

The variability's associated with densities in each waste type are estimated similarly to that of concentration variability. The density distributions for each waste type are developed based on the density measurements within each cluster described above.

The Los Alamos National Laboratory (LANL) team will develop the variability associated with sludge volumes by waste type in a tank, based on tank transaction history. More discussion on volume distribution can be found in Section 4.

2.4 Approach for Supernatant and Saltcakes

Because of the nature of the supernatant and saltcakes, it is impractical to further partition supernatant or saltcake samples into waste type groups. For these two phases of wastes, inventories are estimated using the following two equations:

$$f_{\text{Saltcake}, A, T} = f_{A_{\text{Saltcake}}, T} \otimes f_{D_{\text{Saltcake}}, T} \otimes f_{V_{\text{Saltcake}}, T} \quad (2.4)$$

and

$$f_{\text{Supernate}, A, T} = f_{A_{\text{Supernate}}, T} \otimes f_{D_{\text{Supernate}}, T} \otimes f_{V_{\text{Supernate}}, T} \quad (2.5)$$

where f denotes a probability distribution and $\otimes$ indicates a multiplication operation of two probability distributions.

The concentration and density distributions are assessed at a tank level using an approach combining tank grouping with the analysis of variance (ANOVA) technique. First, 177 tanks are grouped based on the historical chemical composition estimates provided in the SMM model (Agnew et al. 1997). Based on the processing history, the tanks in the same groups are likely to have similar chemical compositions. This step is necessary so that concentrations for tanks that have not been sampled could be estimated using the information of sampled tanks that are in sample groups. An ANOVA model is then applied to supernatant and saltcake data, separately, using the same grouping results. The concentration and density distributions for both supernatant and saltcake wastes are the products of the analysis under a normal distribution assumption.

For the constituents without direct measurements, a similar mapping procedure is applied as described in Section 2.3. Detailed methods to develop concentration and density distributions for supernatant and saltcake are provided in Section 3.2.

The HEABB estimates for supernatant volumes in each tank are considered to be the point estimates of the supernatant volumes. Potential data sources for assessing the uncertainty associated with the supernatant volumes include gamma and/or neutron log data, waste temperature measurements, and tank photos.

Measurements for saltcake volumes are seldom available, and the HDW model does not provide historical estimates for the volumes. The saltcake volumes, however, can be derived using the total tank waste volumes, which have a fair amount of measurements available using various instrumentation. Specifically, the saltcake volume in a tank can be determined by subtracting the volume of sludges and

supernatant from the total waste volume in the tank. The methodology used to develop the distributions for these volumes is discussed in Section 4.

2.5 Monte Carlo Technique for Total Inventory

As discussed previously, the total inventory for a given constituent in a tank is determined by inventories of the three waste phases. The supernatant and saltcake inventories are each in turn determined by three components: concentration, density, and volume. The sludge inventory even contains several sets of such triplets for different waste types. It will be very difficult to derive the total inventory distribution analytically from the combination of such a large number of component distributions. An efficient alternative is using a Monte Carlo simulation technique.

A large number, say 1000, of realizations are generated from each component distribution (e.g. Al concentration distribution for a sludge of 224 wastes, supernatant volume distribution for Tank AY-101, etc.) to form 1000 realization sets. Because of the huge number of component distributions, a sampling strategy, called Latin Hypercube sampling, is used to ensure a reasonable coverage for this high-dimensional component space. Instead of generating realizations randomly, each component distribution is divided into 1000 equal area bins. A value is taken from the center of each of the 1000 bins as the realization of the distribution. The 1000 realizations from each component distribution are then randomly ordered, and realizations from all distributions are combined to form the 1000 realization sets. This last step is to arrange different bin combinations of the component distributions.

The calculation is conducted using Equations 2.3-2.5 for each realization set yielding the same number of realizations of the constituent inventory in the tank for the sludges, saltcakes, and supernatants, respectively. These results can be useful for waste retrieval operations. The realizations of the total inventory for the constituent are obtained using Equation 2.2. Empirical distributions for the total inventories can be estimated based on the 1000 realizations in terms of desired quantiles.

3.0 Methodology to Estimate the Concentration and Density Distributions

3.1 Chemical Composition Distributions by Waste Type for Sludge Waste

For sludge wastes, the entries in the upper left quadrant of Table 1.1, Q11, use analysis results of waste samples as the primary source of information to refine estimates of waste-type concentrations and quantify the uncertainties associated with the estimates of waste-type concentrations. These distributions of waste-type concentrations will be combined with information of the distributions of waste-type volumes in each tank for the analytes in the left half of the matrix (quadrants Q11 and Q21) and density distributions to estimate the distributions of volume and concentration for all 177 tanks for those analytes. The right half of the matrix, quadrants Q12 and Q22, will be filled by identifying relationships between the measured and unmeasured constituents.

The methodology is applied similarly for tanks in quadrants Q11 and Q21 (see Section 3.1.1). The constituent-concentration sample results from quadrant Q11 are mapped into sludge and saltcake waste types by means of a cluster-analysis exercise. Waste-type distributions are then fit using the mapped data. These waste-type distributions in conjunction with the density and volume distributions are used in the same manner to estimate the inventories for tanks in quadrants Q11 and Q21. Note the difference between the "waste-type" approach, which is used in the present methodology, versus the "tank" approach. For example, a tank in Q11 (sampled tank) will use its own sampling data along with data from other similar tanks for the estimate of its inventory.

Quadrants Q12 and Q22, the right half of the matrix, will be filled by identifying relationships between the measured and unmeasured constituents. This methodology is discussed in Section 3.1.2.

This is the approach outlined in the Statement of Work (SOW) and being refined by additional research and analysis in close coordination with Mr. Brett Simpson, the LMHC Technical Contact.

3.1.1 Measured Constituents

A large volume of tank sampling results was retrieved from the TCD in November 1997 via the TCD Website. A discussion of the data processing steps and data summaries is given in LoPresti et al. (1997). The data were retrieved from the database independently for each constituent. The constituents that had good coverage (i.e., analytical results for a majority of the tank samples) over the samples that have been taken from the Hanford tanks were classified as measured. These constituents are found in quadrants Q11 and Q21 of the matrix described in the last section.

The data were checked for quality. Only data approved under the Tri-Party Agreement (TPA) were retained in the extracted data. These are tank sample analyses conducted after 1989. The Washington State Department of Ecology has recognized these data as being adequate to generate TPA documents. Sample records for preferred analytical methods were the only records retained in the database. Sample records that were potential suspects were checked for potential exclusion from the data set. Other checks and conversions were also made, such as result-unit conversion and addressing less-than-detection records. Analyses of matrix spikes, blanks, and laboratory standards were not included in the quality assurance (QA) checks that were made. A more complete description of the QA issues addressed is found in LoPresti et al. (1997).

Once the data sets were organized for each constituent, a multivariate data set was organized that included all constituents deemed important for discriminating between waste types for data clustering purposes. These constituents are found in the list below. The technical staff at LMHC approved this list of constituents. To create this data set, it was necessary to match records from each of the analyte (univariate) data sets. To accomplish this, a unique identifier (location identification [LOCID]) was constructed for each tank-sampling location. A tank-sampling location could be an individual grab sample, quarter segment, half segment, whole segment, core composite, tank composite, or auger sample. The sample phase (liquid or solid) was also used in the LOCID.

Table 3.1. Constituents Used in Cluster Analysis

1. Aluminum	7. Fluoride	13. Nitrite
2. Bismuth	8. Water	14. Phosphorus
3. Calcium	9. Lanthanum	15. Lead
4. Chromium	10. Manganese	16. Uranium
5. Cesium-137	11. Sodium	17. Zirconium
6. Iron	12. Nickel	

All of the records for a given constituent and LOCID were simply averaged together. Typically, the records that were averaged were single primary and duplicate analysis results. The records for all of the constituents were matched using the LOCID to create rows of matrix of multivariate data. The columns of the matrix are the 18 constituents, and the rows are the LOCIDs. There were 1137 unique LOCIDs for the solids data and 376 LOCIDs for the liquid data used in the clustering analysis, which is discussed in a later section.

Both solids (sludges and saltcakes) and liquid samples were included in data-extraction efforts. The data for liquid and solid waste phases were separated for sample grouping work because of their different properties.

A technique called Hierarchical Cluster Analysis (HCA) using complete linkage (see Everitt 1993) was used as the primary method to group samples into classes with similar waste properties. This method was supplemented by other techniques, some of which will be mentioned later in this section.

The present cluster analysis, which focuses on mapping samples (LOCIDs) into waste types, involves 1137 solid sample LOCIDs. The tree-cutting rule has also been modified to better model the characteristics of the waste-type distributions that we expect exist.

The modification to the cutting rule will now be described. First, each LOCID is tagged with potential waste types that the sample could have come from based on the TLM (Agnew 1997). These potential waste types are the primary, secondary, and tertiary TLM waste types for the tank that the sample came from. The samples are then clustered using HCA methods. The resulting tree or dendrogram is then cut starting at 2 clusters and working down the tree to some specified stopping point (350 cuts for this application). At each cut, the resulting clusters are examined to see if there is a 75% majority for a single TLM waste type. If there is, the cluster is given the name of the waste type found in majority, and the cluster is removed from the data set. This continues from cut to cut down the tree until the specified stopping point is reached.

Table 3.2 shows a cluster with a 75% majority for one of the waste types. It was found at the 27th cut through the tree. It is comprised of 20 LOCIDs of which 16 have CWZr waste as one of their waste types (80% majority). Because this cluster met the majority criteria, the cluster was named "CWZr" and set aside, and the analysis continued. It is very interesting that the samples that make up the 20% minority

have CWP and CWR (other cladding wastes) as potential waste types. This finding lends some evidence that the different cladding wastes (CWP, CWR, and CWZr) have similar properties.

Table 3.3 contains a summary of the results of the clustering exercise. Eight super-groups of data were identified in the analysis. The table shows the number of samples that were mapped into each super-group. It also shows HDW/TLM waste types that were mapped into each super-group. For example, super-group 2 has 6 HDW/TLM waste types that have been mapped into it. The 59 samples that are in this super-group will be used to estimate distributions for these 6 waste types. Note that the distributions will be the same for these 6 waste types. Out of the 53 HDW waste types, waste-type concentration distributions can be obtained for 25 of them. These 25 waste types represent approximately 90% of the total waste volume.

Using the majority cutting criteria, 313 out of the 1137 solid samples did not cluster. We suspect samples that fall on waste-type boundaries (i.e., they are a mix of two or more waste types) are included in this number. The distributions for these waste types will be determined using site-wide sampling information or possibly these 313 miscellaneous samples.

Two key assumptions are used in this analysis. First, we assumed that the waste-composition differences between waste campaigns are marginal. Second, we assumed that the following waste types from the Bismuth Phosphate process also have negligible differences: 1C1, 1C2, 2C1, 2C2, UR, BSltCk. The data showed evidence supporting these assumptions.

Table 3.2. Majority Cutting Rule Example

Group Name	LOCID	Cut Number	Potential TLM Waste Types		
			Primary	Secondary	Tertiary
CWZr	SC19407LAW-103	27	CWZr		
CWZr	SC19309LAW-103	27	CWZr		
CWZr	SC19409UAW-103	27	CWZr		
CWZr	SC19408LAW-103	27	CWZr		
CWZr	SC19408UAW-103	27	CWZr		
CWZr	SC19308LAW-103	27	CWZr		
CWZr	SC19308UAW-103	27	CWZr		
CWZr	SC19309UAW-103	27	CWZr		
CWZr	SC19508LAW-105	27	CWZr	SMMA	PL
CWZr	SC19507LAW-105	27	CWZr	SMMA	PL
CWZr	SC19307UAW-103	27	CWZr		
CWZr	SC19509UAW-105	27	CWZr	SMMA	PL
CWZr	SC19508UAW-105	27	CWZr	SMMA	PL
CWZr	SC19608UAW-105	27	CWZr	SMMA	PL
CWZr	SC11003LS-107	27	CWR	R	SMMS
CWZr	SC11103LS-107	27	CWR	R	SMMS
CWZr	SC16201LC-104	27	CWP	OWW	CWZr
CWZr	SC10503LS-107	27	CWR	R	SMMS
CWZr	SC11003US-107	27	CWR	R	SMMS
CWZr	SC19608LAW-105	27	CWZr	SMMA	PL

Table 3.3. Mapping of Waste Types into Super Groups

Super-Group	# Samples mapped into Super-Group	Waste Types Mapped into Super-Groups
1	120	224
2	59	1C1, 1C2, 2C1, 2C2, UR, BSltCk
3	74	BYSltCk
4	51	R1, R2, CWR1, CWR2, CWP1, CWP2
5	53	CWZr1, CWZr2
6	37	PFeCN1, PFeCN2, TFeCN, 1CfeCN
7	217	SMMA1, SMMA2, SMMT2
8	213	SMMS1, SMMS2
Misc*	313	MW1, MW2, P1, P2, PL1, OWW1, OWW2, OWW3, Z, HS, TH1, TH2, AR, B, BL, SRR, CSR, DE, CEM, NIT, Salt.Slurry, DW, N, T1SltCk, RSltCk, P3, PL2, PASF

*Misc is the set of LOCIDs that were not mapped into the 8 super-groups.

Distribution Estimation: For each constituent and each super waste type, when there are enough sample concentration data, we estimate the concentration distribution based on data. When sufficient data do not exist, we use methods described in the Unmeasured Constituents section (Section 3.1.2) to produce an uncertainty distribution.

When both below detection-limit data and detected-concentration data exist for a constituent for a given super waste type, we convert all the below detection-limit data to half of the detection limits. When all the data for a constituent for a given super waste type are below detection limits, we use a uniform distribution between 0 and the largest detection limit as its distribution.

Because Hanford records do not provide sufficient information on the forms of the concentration distributions for constituents in different waste types to construct the distribution parametrically, we estimate the probability density functions (pdf) non-parametrically. We use the kernel smoothing method with a triangle kernel to estimate the probability density function. The bandwidth of the kernel is determined by visual inspection via several trial-and-error experiments to balance between the smoothness of the pdf and the fidelity of the estimated cumulative distribution function (cdf) to the empirical cdf. It turns out that the bandwidth that gives the appropriate amount of smoothing for most of the major constituents for the 8 super waste types and site-wise data is 1/5 of the distance between the second and 98th percentiles of the data. The estimated pdf is truncated at 0 and 1,000,000 and then normalized so that the area under it is always 1. It turns out that for most cases, the area under the extended curve on the lower end, i.e. the probability based on the extrapolated density curve on the lower end, lies around 2% while the probability based on the extrapolated density curve on the upper end lies around 1.5%.

Given the estimated pdf for any constituent and any one of the 8 super waste groups, we can generate 1000 samples with equal probabilities from the estimated distribution. Those 1,000 samples will be used in the Monte Carlo simulations to estimate the uncertainties.

Figures 3.1 through 3.3 show some examples of the procedure. Figure 3.1 shows the histograms of data for 4 selected constituents and super waste group 224. Also shown in Figure 3.1 are estimated pdf curves and Agnew's concentration values (the vertical lines) for the selected 4 constituents and the waste group. Figure 3.2 shows the estimated cdf's (based on estimated pdf's) and the empirical cdf's for the selected 4 constituents and the waste group. Figure 3.3 shows the histograms based on 1,000 equal

probability Monte Carlo samples for the 4 constituents and the waste group together with Agnew's values (the vertical lines).

Figure 3.1. Histograms and Estimated pdf Curves Based on Sample Data for Four Selected Constituents and Super Group 224. Vertical lines show Agnew's concentration values.

Bi, super waste group 224

Al, super waste group 224

K, super waste group 224

Cr, super waste group 224

Figure 3.2. Estimated cdf Curves (Solid Lines) Based on Estimated pdf's from Data and Empirical cdf's (Broken Lines) for the Four Selected Constituents and Super Group 224

Figure 3.3. Histograms Based on 1,000 Equal Probability Monte Carlo Samples from the Estimated pdf's for the Four Selected Constituents and Super Group 224. Vertical lines show Agnew's values.

3.1.2 Unmeasured Constituents

A large number of analytes (principally radionuclides) estimated by the best basis are not typically included in the analysis of tank farm samples, and for many analytes, no direct measurements are available. For these analytes, a relationship must be identified between a measured analyte and an unmeasured analyte that correlates to the uncertainty associated with the estimate. The relationships that are believed to be indicative of the variability of an analyte are as follows:

Elemental: Uncertainties developed that are based on measuring the total quantity of an element are assumed to apply to all isotopes of the element (e.g., Ni^{59} is assumed to have the same uncertainty as Ni).

Compound: Analytes found primarily in a compound that is typically measured are assumed to have the same uncertainty as that compound (e.g., C^{14} will have an uncertainty similar to CO_2 since almost all C^{14} will be found in CO_2).

Daughter Products: Short-lived daughter products are assumed to have the same variability of the parent radionuclide (e.g., Y^{90} is assumed to have the same uncertainty as Sr^{90}).

Periodic Table: Analytes that occur in similar locations on the periodic table typically possess similar chemical and physical behavior. They are assumed to be similarly distributed within a tank and therefore will have similar uncertainties (e.g., the uncertainty associated with the Tc^{99} inventory will be based on the uncertainty of Mn).

Activity: Many transuranic analytes can be determined from the total alpha measurement. The uncertainty of these analytes will be based on the uncertainty associated with total alpha (e.g., the inventory of Np^{237} can be estimated from total alpha; therefore the uncertainty distribution should be similar to that of total alpha).

Table 3.4 lists the analytes for which little or no sample measurements are available and the measured analyte (analog) being used to extrapolate the uncertainty. This table also lists the relationship used to establish the mapping.

Table 3.4. Unmeasured Analytes and the Corresponding Measured Analog

Unmeasured Analyte	Measured Analog	Relationship
^3H	Water	Compound
^{14}C	CO_2 (carbonate)	Compound
^{59}Ni	Ni	Element
^{60}Co	Ni	Periodic Table
^{63}Ni	Ni	Element
^{79}Se	SO_4	Periodic Table
^{90}Y	^{90}Sr	Daughter Product
$^{93\text{m}}\text{Nb}$	Zr	Periodic Table
^{93}Zr	Zr	Element
^{99}Tc	Mn	Periodic Table
^{106}Ru	Fe	Periodic Table
$^{113\text{m}}\text{Cd}$	Pb	Periodic Table
^{125}Sb	Bi	Periodic Table
^{126}Sn	Pb	Periodic Table
^{129}I	Cl	Periodic Table
^{134}Cs	^{137}Cs	Element

Unmeasured Analyte	Measured Analog	Relationship
^{137m} Ba	¹³⁷ Cs	Daughter Product
¹⁵¹ Sm	La	Periodic Table
¹⁵² Eu	La	Periodic Table
¹⁵⁴ Eu	La	Periodic Table
¹⁵⁵ Eu	La	Periodic Table
²²⁶ Ra	Sr	Periodic Table
²²⁷ Ac	La	Periodic Table
²²⁸ Ra	Sr	Periodic Table
²²⁹ Th	U	Periodic Table
²³¹ Pa	U	Periodic Table
²³² Th	U	Periodic Table
²³² U	U	Element
²³³ U	U	Element
²³⁴ U	U	Element
²³⁵ U	U	Element
²³⁶ U	U	Element
²³⁷ Np	total alpha	Activity
²³⁸ Pu	total alpha	Activity
^{239/240} Pu	total alpha	Activity
²³⁹ Pu	often measured/total alpha	Activity
²⁴⁰ Pu	total alpha	Activity
²⁴¹ Am	often measured/total alpha	Activity
²⁴¹ Pu	total alpha	Activity
²⁴² Cm	total alpha	Activity
²⁴² Pu	total alpha	Activity
²⁴³ Am	total alpha	Activity
²⁴³ Cm	total alpha	Activity
²⁴⁴ Cm	total alpha	Activity

The distributions developed for the measured analytes are expected to be scaled to account for the relative amount of the unmeasured analog and applied directly without an increase in uncertainty. It is recognized, however, that applying distributions for measured analytes to unmeasured analytes will introduce additional uncertainty. The additional uncertainty arises both from differences in the distributions between the measured analyte and its unmeasured analog, particularly those based on the periodic table, and from applying distributions developed for sample-based means to model-predicted values. Comparing the sample-based means to the model predictions for the measured analytes should provide an estimate of the uncertainty introduced by centering sample-based distributions around the model predictions for the unmeasured analytes. Also, analog relationships could be developed for two measured analytes using the same methodology used for the unmeasured analytes. The distributions that would result from these relationships could then be compared to the actual distributions based on the sample data to provide an estimate of the additional uncertainty.

3.1.3 Unsampld Waste Type

This subsection will discuss a site-wide variability approach that will be applied to unsampled waste types. A waste type is classified as an unsampled waste type under one of the following two conditions: 1) the waste type has not yet been sampled; or 2) although the waste type may have been

sampled, the samples were so contaminated by other waste types that no group of representative samples can be identified through the cluster analysis. For these waste types, a site-wide variability approach will be applied, which is discussed in this subsection.

This site-wide variability approach has two major steps. The first step is to develop an empirical site-wide distribution for the target constituent using all available measurements of this constituent. The method used to develop the empirical distribution is the same procedure used to develop the concentration distributions for each identified waste type (see Section 3.1.1 for details). The purpose of this step is to capture the range and shape of the site-wide distribution for the constituent. The second step is to re-scale the site-wide distribution for the waste type of interest by multiplying the estimated cdf with a constant so that the median of the re-scaled distribution is located at the mean concentration value of the waste type estimated from historical information, specifically, the mean concentration from the HEABB estimates or HDW model. If the upper tail of the re-scaled distribution is above 1,000,000 ppm, our tentative plan is to use two different constants to re-scale the cdf. For values below the median, we use one constant multiplier so that the new median is at the desired location; for values above the original median, we use another constant multiplier so that the new maximum value is at 1,000,000.

Because of the lack of information, the distributions developed using this approach may not represent the true variability of the concentrations in unsampled waste types. However, this approach can provide a conservative variability estimation provided the samples that have already been taken captured the site-wide extreme values.

Figures 3.4 through 3.7 give some examples of the procedure. Figure 3.4 shows the histograms of the concentrations for 4 selected constituents based on site-wide unclustered sample data. Also shown in Figure 3.5 are the estimated pdf curves. Figure 3.5 shows the estimated cdf's based on the estimated pdf's and the empirical cdf's for the selected 4 constituents based on site-wide unclustered sample data. Figure 3.6 shows the histograms based on 1000 samples with equal probabilities from the estimated site-wide distributions for the 4 constituents. Figure 3.7 shows the re-scaled (from the site-wide distribution) distributions based on 1000 simulated samples for waste-type T1.SaltCake.

Figure 3.4. Histograms and Estimated pdf Curves Based on Site-Wide Unclustered Sample Data for Four Selected Constituents. Vertical lines show Agnew's values.

Figure 3.5. Estimated cdf Curves (Solid Lines) Based on the Estimated pdf's from Site-Wide Unclustered Sample Data and the Empirical cdf Curves (Broken Lines) Based on Site-Wide Unclustered Sample Data for the Four Selected Constituents

Figure 3.6. Histograms Based on 1,000 Equal Probability Monte Carlo Samples from the Estimated pdf's for the Four Selected Constituents and Site-Wide Unclustered Sample Data

Al, T1.SltCk, Monte Carlo sample

Bi, T1.SltCk, Monte Carlo sample

Cr, T1.SltCk, Monte Carlo sample

K, T1.SltCk, Monte Carlo sample

Figure 3.7. Histograms Based on 1,000 Equal Probability Monte Carlo Samples from the Estimated Distributions for the Four Selected Constituents and Waste-type T1.SltCk. Vertical lines show the Agnew's values.

3.2 Concentration Distributions for Saltcake and Supernatant

As discussed in Section 2.2, a Grouping-ANOVA approach is used for saltcake and supernatant wastes. This approach estimates the mean concentration and density of each constituent in the left quadrants of Table 1.1 for saltcake and supernatant wastes in each tank. The procedure also provides the uncertainties associated with these mean estimates/predictions. To obtain distributions for the concentrations and densities, a normality assumption or other distribution assumption is necessary to specify the shape of the distributions.

Section 3.2.1 discusses the purpose and procedure for grouping tanks for their saltcake and supernatant wastes. Section 3.2.2 describes the saltcake data and supernatant data used later in the ANOVA procedure to do estimation/prediction. Section 3.2.3 discusses a random-effect model and assumptions made to analyze variance, as well as the outcomes from this analysis.

3.2.1 Tank Grouping for Saltcake and Supernatant

Unlike sludge wastes, saltcakes and supernatants cannot be further partitioned into waste types due to the mixing nature of the transactions and evaporations. Therefore, the mechanism used to extend sample results from the tanks in the quadrant Q11 to the tanks in Q21 (see Table 1.1) has to be at a tank level, instead of a waste type level.

A tank grouping technique can provide an extending mechanism at a tank level for supernatant and saltcake wastes. It is believed if tanks have similar transaction history, the chemical compositions of these tanks are likely to be similar. A similar assumption can be made for only supernatant or saltcake wastes in the tanks. If the tanks can be grouped in such way so that the tanks in same group tend to have similar compositions in the supernatant or/and saltcake wastes, we may estimate these concentrations for the unsampled tanks using the sample data from the tanks within the same group.

The historical concentration estimates for the wastes tracked by SMM model in each tank are selected as classifiers for this tank grouping effort. This is because SMM model tracks both supernatant and supernatant concentrate (saltcake) wastes and, therefore, those historical estimates reflect the tank transaction history for the supernatant and saltcake portion of the wastes.

The same 17 constituents as listed in Table 3.1 are currently used as the discriminators. Alternative constituents more efficient for those two waste phases are still under investigation. A hierarchical cluster analysis technique is applied to these historical concentration estimates to achieve tank grouping. The resulting tank grouping can then be used in an ANOVA model to obtain concentration estimates and uncertainties for both supernatant and saltcake wastes in all 177 tanks.

3.2.2 Data of Saltcake and Supernatant

The data used to estimate the mean concentrations and densities for saltcake and supernatant should represent these portions of wastes in a tank.

For a supernatant, the data used are all sample measurements with samples labeled as liquid. This includes all supernatant and drainable liquid samples. The underlying assumption is that all liquid wastes are classified as SMM wastes. The assumption may be further modified so that supernatants contain only supernatant wastes since the drainable liquids contained in sludge layers may be considered as a part of the sludge.

For saltcakes, the samples are identified through the cluster analysis, which is described in Section 3.1.1, that is made on solid samples. The samples classified into the super groups 7 and 8 are considered to be saltcake samples since the two groups contain primarily the SMM waste types defined in the HDW model. Some samples in the miscellaneous groups are also considered to be saltcake samples if the sampled tanks contain primarily the saltcake wastes.

3.2.3 ANOVA Procedure

A univariate ANOVA technique is applied separately to saltcake and supernatant samples described in the previous section. The term "univariate" means that the analysis is conducted one constituent at a time without imposing any correlation structure among the constituents. The ANOVA model used to obtain predictions for mean concentrations or densities along with the associated uncertainties is

$$y_{ijk} = \mu + G_i + T_{ij} + E_{ijk} \quad (3.1)$$

where

- y_{ijk} = the concentration of a specific constituent or density of the k^{th} sample in tank j in group i
- μ = the overall mean concentration of the constituent across all tanks
- G_i = the random effect of group i and is assumed independently and identically distributed as a $N(0, \sigma_G^2)$ distribution for all groups
- T_{ij} = the random effect of tank j in group i and is assumed independently and identically distributed as a $N(0, \sigma_T^2)$ distribution for all tanks
- E_{ijk} = the within-tank random error term and is assumed independently and identically distributed as a $N(0, \sigma_E^2)$ distribution for all tanks.

A similar approach was used to predict T-200 series tank concentrations that had not yet been sampled (see Engel 1997 for details). A one-core sampling taken later from each T-200 series tank showed that more than 90% of the 68 estimated means (17 major constituents in four tanks) based on the sample measurements fell in the 95% confidence interval of the tank means produced through the model analysis.

In this ANOVA model, a normality assumption is made for the concentration distributions. This assumption is necessary because the analysis procedure is not available for such models if the underlying distribution assumption is different from normality. If, however, the sample data show that some concentration distributions severely deviate from the assumption, some data transformation techniques will be applied to the data to satisfy the assumption.

A Restricted Maximum Likelihood (REML) method is used to estimate the variance components in the model. The end products of this analysis are the mean concentration and associated uncertainty of saltcake and supernatant for each constituent in each tank.

3.3 Density and Specific Gravity by Waste Type

This section discusses tank waste sample density and specific gravity data. These data are of interest because sample density and specific gravity measurements allow calculations of mass inventories from analyte concentrations for given volumes. The following tables are intended to summarize what we

know about the density and specific gravity data available from TWINS/TCD as of the end of January 1998.

3.3.1 Definitions and Record Counts

Table 3.5 shows an overall tabulation of density and specific gravity records retrieved from TWINS/TCD from a special download performed in early February 1998 to capture density data available only after our November 1997 download. This table shows that several types of density measurements are carried in TWINS/TCD. They are defined as

- “Bulk density” applies to the entire sample from the core and includes interstitial liquids.
- “Density” applies to the solids portion of the sample after draining off liquids.
- “Solids density” applies to the solids portion of the sample after draining off liquids.

In addition, a few records are described as “density before centrifuge” and “density after centrifuge.” These are not considered in this section.

In TCD, samples are identified as coming from solids-phase samples or liquid-phase samples. Density measurements apply principally, but not always, to solid samples, and specific gravity measurements apply even more consistently to liquid samples. Density measurements are always reported in TCD in grams per milliliter (g/mL), and as such allow direct conversion of a concentration expressed on a mass basis (e. g., $\mu\text{g/g}$) to a volume basis. Specific gravity data are always unitless, being the ratio of the mass of a unit volume of the sample to the mass of the same volume of water at standard temperature and pressure, and can be applied directly to analyte concentrations already on a volume basis (e. g., $\mu\text{g/mL}$).

“Density” and “Solids density” can be treated the same way in the computations. “Bulk density” and “density” actually may not be measuring exactly the same physical entity, but for tank samples, they are probably practically interchangeable. However, not accounting for interstitial liquid in a solid sample may be a source of uncertainty in inventory estimates. Table 3.6 shows differences between aggregate “bulk density” and aggregate “density” means in solid samples, but it is not clear as to what these differences should be attributed to.

Table 3.5. Categorization of Density and Specific Gravity Records. Record counts include records with result value set to NA.

Analyte	Units	Number Solid Samples*	Number Liquid Samples*	Number Primary Samples	Number Duplicate Samples	Number Min- detection
Bulk density	g/mL	549	17	544	22	517**
Density	g/mL	222	78	252	48	0
Solids density	g/mL	22	0	17	5	0
Specific gravity	Unitless	3	944	499	448	378

* Includes Primary and Duplicate sample records, but not minimum detection limit records

** Bulk density minimum detection limit values are uniformly set to 0.5 in the TCD records.

Table 3.5 also tabulates the data by “result type”—pairs of Primary and Duplicate measurements. The table shows that only specific gravity measurements have, as a rule, both Primary and Duplicate measurements, which could, in principle, be used to estimate sampling error. The density measurements,

as a rule, do not have replicate samples. Although these values account for only about 80% of the pairs of specific gravity measurements, they can provide a means to estimate sample error.

3.3.2 Minimum Detection Limits

Density measurements generally do not have meaningful minimum detection limit values in TCD. Specific gravity data do have minimum detection limit values, which provide information on the expected precision of specific gravity measurements. The modal minimum detection limit is 0.001, and the maximal value is 0.10 specific gravity units. These data suggest that in most cases the uncertainty of inventory estimates due to uncertainties in specific gravity measurements will be of the order of 0.05% to 0.1%, with a few cases of 5 to 10%.

3.3.3 Descriptive Statistics

Tables 3.6 and 3.7 show descriptive statistics for all density and specific gravity measurements taken as a whole without regard to tank, spatial location, or existence of duplicate measurements. These tables provide a general idea of the distribution of these data. The minimum value of 0.37 for density in Table 3.6 remains unexplained.

Table 3.6. Solid Samples: Overall Descriptive Statistics for Density and Specific Gravity Measurements

Analyte	N	Min	1 st Q	Median	3 rd Q	Max	Mean	Stdv	Units
Bulk density	525	0.91	1.52	1.62	1.71	2.10	1.58	0.20	g/mL
Density	221	0.37	1.11	1.28	1.44	2.02	1.30	0.27	g/mL
Solids density	22	1.50	1.67	1.71	1.73	2.12	1.73	0.14	g/mL
Specific gravity	3	1.95	2.18	2.40	2.52	2.64	2.33	0.35	Unitless

Table 3.7. Liquid Samples: Overall Descriptive Statistics for Density and Specific Gravity Measurements

Analyte	N	Min	1 st Q	Median	3 rd Q	Max	Mean	Stdv	Units
Bulk density	17	0.94	1.04	1.36	1.47	1.59	1.27	0.28	g/mL
Density	78	0.52	1.15	1.26	1.30	1.48	1.20	0.17	g/mL
Solids density	0	NA	NA	NA	NA	NA	NA	NA	g/mL
Specific gravity	938	0.96	1.074	1.30	1.41	2.77	1.27	0.20	Unitless

Figure 3.8 shows plots for example distributions of bulk density and density data for the 224 super waste type. Each distribution is smoothed by the kernel smoother described above. This demonstrates that the density and specific gravity data can be formed into empirical distributions, which can be used for Monte Carlo simulations in order to better represent natural variability in these observations.

Figure 3.8. Histograms, Estimated pdf Curves, and Estimated cdf Curves Based on Sample Bulk Density and Sample Density Data for Super Group 224

3.3.1 Relating Density Measurements to Concentrations by Sample

Density and specific gravity data are most useful for estimating inventories when the measurements can be directly associated with analyte concentrations in a specific sample or region of each tank, as discussed in Section 2 on the overall approach. In order to determine the extent to which this condition is obtained for the data set, LOCIDs were constructed for each density and specific gravity measurement, as discussed in Section 3.1.1. The density and specific gravity LOCIDs were then matched against the analyte sample LOCIDs and the match results tabulated as shown in Table 3.8.

Table 3.8 shows how density and specific gravity observations are distributed between solid and liquid phase samples. When tabulated on the basis of sample location (LOCID), it is evident that the liquid samples are more likely to have a corresponding specific gravity measurement than the solid samples are likely to have a density measurement. Over 84% of liquid sample LOCIDs can be matched to a specific gravity measurement. In contrast, less than 39% of the solid sample LOCIDs can be matched to a corresponding density measurement.

Table 3.8. Number of Samples by LOCIDs for Which a Density or Specific Gravity Observation Exists

	Number solids samples	Number solid samples matched to density	Number liquids samples	Number liquid samples matched to specific gravity
Number samples	1578		376	
Bulk density	471	29.8%	7	1.8%
Density	146	9.3%	10	2.7%
Solids density	22	1.4%	0	N/A
Specific gravity	2	0.1%	318	84.6%
Taken together	613	38.8%	331	88.0%

Some density and specific gravity data exists for all the super waste types defined above except the FeCN super waste type. Table 3.9 shows the extent of representation for each super waste type for bulk density data. The samples in the SSMA super waste type appear to be much better represented by bulk density data, with 73% of samples matched to density values, than the other super waste types. Similar tables can be built for other classes of density data. Table 3.9 also shows that mean densities vary considerably between super waste types. The 224 waste type appears to have the lowest mean density at 1.24 g/mL. SSMA, SSMS, R, and MISC means lie between 1.63 and 1.75 g/mL.

Although obtaining sample-based matches may be somewhat problematic, taken as a whole, the coverage may not be as sparse as appears in Table 3.9 because samples are usually represented by one of bulk density, density, specific gravity, or some other type of measurement, so that the assignment of specific density values to samples is probably more complete than outlined in Table 3.10. In addition, even when a specific density match to a sample cannot be made, this may indicate that a sample density may have been measured for an entire segment, but other analytes were measured at subsegment resolution. This suggests that concentration data for several LOCIDs would be multiplied by the same density measurement taken at a coarser resolution to calculate the inventory for that tank. Each unmatched sample would have to be taken on a case-by-case basis.

Table 3.9. Bulk density coverage by waste type

Waste type	Number samples (LOCID)	Number Bulk density observations	Percent of waste type Samples represented	Mean Bulk Density (g/mL)	Standard deviation (g/mL)
1C2C	59	3	5%	1.40	0.19
224	120	24	20%	1.24	0.08
BYSlCk	74	23	31%	1.49	0.28
FeCN	37	0	0%	-	-
R	51	6	12%	1.76	0.22
RWZr	53	24	45%	1.45	0.16
SSMA	217	159	73%	1.65	0.09
SSMS	213	65	31%	1.71	0.11
MISC	260	83	32%	1.63	0.22
Totals	1084	387	36%	-	-

TWINS/TCD continues to actively receive new tank sample data, including density and specific gravity data that were not originally included in TCD. It is expected that as the data backlog is added to TWINS/TCD, the proportion of density-to-sample LOCID matches will increase.

4.0 Methodology to Estimate Waste-Volume Distributions

As discussed in Section 2, volume distributions for the three waste phases and the total tank waste need to be developed. The methods to estimate these volume distributions are described in the following four subsections.

4.1 Uncertainty Distribution of Total Waste Volume

Conceptually, we think of tank volumes as the volume of a cylinder, $V = \pi r^2 h$. However, the measurements for these parameters are not certain, and the volume is not flat on the top or bottom. Therefore, instead of using $V = \pi r^2 h$ to estimate the volume, we will 1) partition the tank into volume elements, "voxels", 2) estimate each probability distribution of the volume of each voxel, and 3) sum up the probability distributions to get a total volume probability distribution.

The tank contents will be partitioned into voxels that are sectors of concentric tracks centered at the tank center.

Each voxel will have a cross-sectional shape of a sector. The dimensions of the sector shall be approximately 1 square foot, approximately 1 foot on each side. The i^{th} tank's volume is

$$V_i = \sum_{\text{all voxels}} h_{ij} \otimes a_{ij} = \sum_{\text{all voxels}} (h_{ij}^t - h_{ij}^b) \otimes a_{ij} \quad (4.1)$$

where

h_{ij} = the height of the voxel

h_{ij}^t = the vertical distance of the top surface of the j^{th} voxel from the reference frame

h_{ij}^b = the vertical distance of the bottom surface of the j^{th} voxel from the reference frame

a_{ij} = the area of the horizontal cross-sectional area of the j^{th} voxel.

This small cross-section is considered flat on top and bottom, an approximation that is believed to be sufficiently close to the truth to support the approach.

It is important to identify another issue at this point. The concentrations and densities used will be based on analysis conducted on waste samples that have been handled so that any gas trapped in the waste would be released. Therefore, we have reduced the waste volume by the volume of trapped gas. So the total volume is more usefully estimated as

$$V_i = \left[\sum_{\text{all voxels}} h_i \otimes a_i \right] - V_i^{\text{gas}} = \left[\sum_{\text{all voxels}} (h_i^t - h_i^b) \otimes a_i \right] - V_i^{\text{gas}} \quad (4.1)$$

We need to estimate the probability distribution for each term. Factors that affect the terms are summarized in Table 4.1. We have constructed our measurement system around the assumption that there is an origin to a rectangular reference frame for each tank with a true horizontal reference plane in the vicinity of the top of the tank and a vertical centerline through the tank center.

The top surface of the waste is assumed to be measured in one or more locations with varying accuracy by ENRAF, Food Instrument Company (FIC), photo interpretation, or other means. Points across the top surface are estimated based on all of the location measurements and on the assumption that the surface is a continuous undulating surface. A kriging technique is used to combine these data.

Due to limitation associated with the available data, we may use a nominal surface shape (i.e.; flat, sloped, hill, funnel) to reflect the best estimate of the surface and appropriate level of uncertainty.

A similar approach is used for the bottom, but the waste surface is presumed to be much smoother. Tank bottoms were designed to be either flat or saucer shaped. We do not assume they follow either design perfectly. Allowance is made for bumps, warps, and bulges that may exist in the tank bottoms.

The diameters of the tanks are nominally 75 feet or 20 feet with stated tolerances on the design drawings. The voxels consist of vertical columns with horizontal cross-sections that are shaped as sectors with a radial measure approximately 1 foot in length. This length will vary proportionately to any deviation in total tank diameter. (For instance, if the probability distribution of a tank diameter is presumed to have a 1% chance of being less than 74.8 feet, then the radial measurement will also have a 1% chance of having a length of 74.8/75 feet.)

Considerable work has been accomplished in estimating the volume of gas trapped in the waste of several tanks. This work will be extended for use here. The volumes will be converted to temperature and pressure values estimated to be in the tank at the time the volume measurements were made.

Table 4.1. Error Sources Affecting Total Tank Waste Volumes

Term	Error Sources Affecting the Term	Description
h_{ij}^t	E_{1ir}^{vt}	Horizontal reference plane error associated with riser/data source r of tank i
	E_{2ir}^{vt}	Instrument calibration error for riser/data source r of tank i .
	E_{3ir}^{vt}	Noise for riser/data source r of tank i .
	E_{4i}^{vt}	Parameters for interpreting locations of top surface of waste in tank i .
	E_{sij}^{vt}	Deviation from location j of tank i , given information from all risers/data sources $r = 1, \dots, R$ and results of kriging program.
h_{ij}^b	E_{li}^{vb}	Horizontal reference plan error of estimated distance between tank reference frame and bottom reference frame.
	E_{4i}^{vb}	Parameters for interpreting locations of bottom surface of waste in tank i .
	E_{sij}^{vb}	Deviation from location j of tank i , results of kriging program.
a_i	E_{lij}^{va}	Deviation from nominal size of horizontal cross-section of voxel j in tank i .
V_i^{gas}	E_{li}^{vg}	Volume of gas

4.1.1 Comments on the Estimation of the Parameter for E_{1ir}^{vt} , Uncertainty in Absolute Riser Elevations

Since the calculation method for determining volumes relies directly on the tank-riser elevations and other reference points for other measurement systems, the uncertainty in those elevation measurements will affect final volume estimates. The report, "Waste Tank Risers Available for Sampling" (Lipnicki 1997) lists the riser elevations for many risers in each tank. Several riser elevation benchmarking studies were performed in the early 1990s. As a result, 209 cases spread between 100 tanks have multiple measurements for the elevation of the same riser. These data will be analyzed using statistical methods.

4.1.2 Comments on the Estimation of the Parameters for E_{2ir}^{vt} , Instrument Calibration Errors, and E_{3ir}^{vt} , Instrument Measurement Error (Noise)

Instrument calibration errors may occur when instruments are periodically replaced, cleaned, and/or recalibrated, causing a noticeable one-time change in waste level. These instruments are apparently more effective at measuring relative changes in surface level than absolute surface level. Unfortunately, the absolute surface level is what is used to calculate waste volumes. The calibration error can be estimated by identifying all calibration points in the surface level measurements.

Instrument measurement error (noise) is inherent to the measuring device and is not due to the calibration factors listed above.

These two sources of error can be estimated for data that come from ENRAF or FIC by fitting the surface level data for all 177 tanks to an ANOVA. We created a data set that contains 435,454 raw surface level measurements from 1/1/1990 through 1/16/1998 for all 177 tanks. Observations labeled as suspect and observations taken with the FIC in intrusion mode were removed from the data set. Observations taken with the FIC in intrusion mode are not true surface level observations because the FIC probe is lowered to a depth above the waste surface, and conductivity across the probe is not achieved unless the waste surface rises with the intrusion of water or other materials. The cleaned data set contains 309,141 surface level measurements for all 177 tanks. The ANOVA model fit for this data set will provide insight into the uncertainty associated with the error sources.

4.1.3 Comments on the Estimation of the Parameter for E_{1i}^{vb} , Error Source of the Difference in the Horizontal Reference Plane, Error of Estimated Distance Between Tank Reference Frame and Bottom Reference Frame

We expect to be able to estimate the error difference for each tank and estimate the uncertainty about it from design drawings and other tank-specific data sources.

4.1.4 Comments on the Estimation of the Parameters for E_{4i}^{vt} , E_{5i}^{vt} , E_{4i}^{vb} , E_{5i}^{vb} .

- E_{4i}^{vt} = Parameters for interpreting locations of top surface of waste in tank i.
- E_{5ij}^{vt} = Deviation from top location j of tank i, given information from all risers/data sources $r = 1, \dots, R$ and results of kriging program.
- E_{4i}^{vb} = Parameters for interpreting locations of bottom surface of waste in tank i.
- E_{5ij}^{vb} = Deviation from bottom location j of tank i, given information from data sources $r = 1, \dots, R$ and results of kriging program.

These parameters are associated with the spatial interpolation of a few measured data sources for the top of the waste to estimate the top of all voxels, and similarly, for the bottom of the waste surface. We will estimate the value of the parameters from the values of the measured data sources.

4.1.5 Comments on the Estimation of the Parameters for E_{1ij}^{va} , Voxel Horizontal Cross-sectional Area Uncertainty

A perfect cylinder 75 feet in diameter (450 inches in radius) would exhibit 2,754 gallons of volume per inch of height. Conversely, a tank with a volume conversion of 2,750 gallons per inch would represent a tank with a radius of 449 $\frac{3}{4}$ inches. If the construction tolerances allowed for ± 1 inch in radius, then a volume uncertainty of 24 $\frac{1}{2}$ gallons per inch would be introduced. This represents a volume error of approximately 0.9%. If the construction tolerances were ± 2 inches, the volume error would be 49 gallons per inch or 1.8%. The construction tolerances should be reported on the as-built drawings or construction plans. For the AN double-shell tank (DST) tank farm, the tolerance is listed as ± 2 inches (Drawing No. H-2-71975). Since the tank farms were constructed over a period of decades, it is highly conceivable that the construction tolerances were different for various tank farms.

4.1.6 Comments on the Estimation of the Parameters for E_{1ij}^{Va} , Volume of Gas Trapped in the Waste of Tank i

The current concentration estimates for tank wastes do not account for any gas trapped *in situ* within the waste matrix. Upon sampling, this gas is released before measuring waste density and analyte concentrations. When gases accumulate in the waste matrix, the waste is essentially diluted by the amount of trapped gas present, relative to the analyte concentrations measured in the laboratory. Although gas trapped in the waste matrix will be affected by changes in atmospheric pressure, this effect is small relative to the volume of gas. This parameter can be approximated using the total volumes of flammable gas estimated by Blaine Barton and Paul Whitney. For many tanks, this value will be very small.

4.2 Sludge Volumes by Waste Type

For the sludge portion of wastes, volume distribution in a tank needs to be developed for each waste type. Although volumes by waste type are very difficult to measure and, therefore, little measured data are available, the historical tank-waste transaction records can provide an avenue to obtain such estimates. In fact, point estimates of the sludge waste volumes by waste type have already been provided in the HDW model by the LANL team.

Recently, the LANL team has agreed to extend their work to furnish uncertainty estimates for these volumes. The uncertainty estimates, presumably centered at the current HDW model point estimates, would be developed based on transaction frequency of a tank and the amount of each transaction. With a distribution assumption, typically normal, volume distributions for saltcakes can be built using the point estimates as means and the uncertainty estimates as variances.

4.3 Supernatant Volumes

The supernatant volume estimates provided in Hanlon's report (Hanlon 1997) are used as the point estimates. Potential data sources for assessing the uncertainty associated with the supernatant volumes include gamma and/or neutron log data, waste-temperature measurements, and tank photos. These measurements may help to identify the border between supernatant and sludge/saltcake layers. The approach for developing volume distributions for supernatants is still under investigation.

4.4 Saltcake Volumes

The saltcake volumes are determined by the following equation:

$$\text{Saltcake Volume} = \text{Total Volume} - \text{Sludge Volume} - \text{Supernatant Volume} \quad (4.2)$$

This indicates that the saltcake volume in a tank is not independently distributed, but bounded by the other three volumes. Therefore, volume distributions for the saltcakes may not need to be built separately. However, a procedure is required to restrict this calculated quantity within a reasonable physical range. This procedure is currently under development.

5.0 Conclusion

The methodology presented in this paper is based on scientific principles, sound technical knowledge of the realities associated with the Hanford waste tanks, the Hanford "Best Basis" research, chemical analysis of samples from the tanks and historical data. It is difficult to estimate tank contents and uncertainty intervals for tanks and analytes for sampled and unsampled tanks and for measured and unmeasured analytes, but it is necessary to accomplish the Hanford mission. This methodology presents a reasonable approach that parallels the approach taken by the Hanford "Best Basis" team and uses sound mathematical principles.

The approach partitions the waste into three groups:

1. **Sludges** are residual solids that settled out of the waste slurries, often remaining in the same tank to which they were first transferred. Interstitial drainable liquids are considered to be a part of sludges. Four saltcakes are also included in this group, but easily identified: B, BY, T1, and R saltcakes.
2. **Saltcakes** are formed as precipitates from supernatant, often as a result of the cooling after the waste completes the evaporator process.
3. **Supernatants** are liquids that stand on top of sludge or saltcake layers in the tanks.

Each of these has a generated probability distribution based on the probability distributions for:

1. waste concentrations (micrograms [microCuries] of a specified analyte per gram of waste or per milliliter of waste),
2. density (grams of waste per liter, if concentrations were on a weight basis),
3. volume of waste.

The estimation of the three probability distributions uses direct or indirect measurements of the three quantities, whenever possible, and historical process records.

The development of this methodology has evolved considerably over the past few months to a more implementable approach. We anticipate more changes, and refinements will be necessary in the upcoming months as more investigations into the exact nature of the available data are completed.

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