

Standard Inventories of Chemicals and Radionuclides in Hanford Site Tank Wastes

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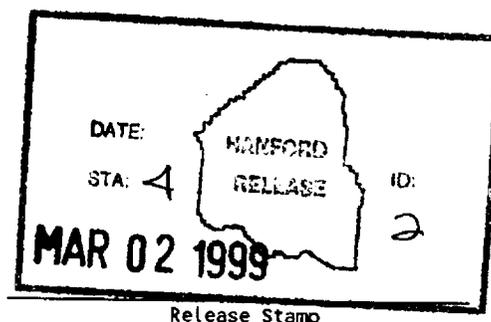
Abstract: This report presents work performed to date to establish standard global and tank-by-tank waste component inventory estimates. This report emphasizes the derivation of global inventory estimates (based primarily on process history) for 30 nonradioactive components and 46 radionuclides that account for approximately 99 percent of the total tank waste inventory. The global waste inventories presented herein complement and supplement inventories derived by summation of tank-by-tank estimates by providing an alternate basis for comparison. The tank-by-tank values are based primarily on sample analyses and are presently considered the best-basis inventories for data end users. Tank-by-tank best-basis inventories are accessible on the Tank Characterization Database. To ensure that inventory values resulting from these evaluations are technically defensible and reproducible, every attempt is being made to document the bases, i.e., originating sources of assumptions, data, and background information that support these values.

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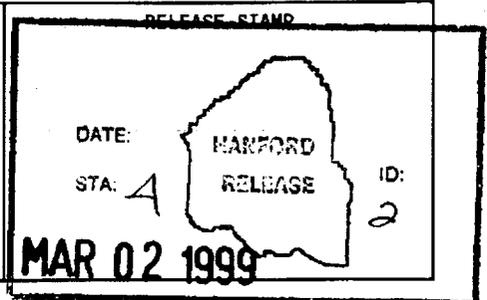
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STANDARD INVENTORIES OF CHEMICALS AND RADIONUCLIDES IN HANFORD SITE TANK WASTES

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EXECUTIVE SUMMARY

The Best-Basis Inventory program is chartered to develop a Best-Basis Inventory (Standard Inventory) of chemicals and radionuclides in wastes stored in the 177 Hanford Site underground tanks. The Best-Basis Inventory program has three major responsibilities:

- Develop and maintain estimates of the total (global) tank waste inventories based on process knowledge.*
- Establish and maintain tank specific Best-Basis Inventories based primarily on analytical data and process knowledge.*
- Reconcile conflicting information and establish a Tank Farm Total Best-Basis Inventory for tank waste.*

This report presents global inventory estimates for chemical and radionuclide components in the 177 Hanford Site underground storage tanks. The global inventories are an independent estimate of the total amount of each chemical or radionuclide component (e.g., metric tons of sodium, nitrate, aluminum, etc., and curies of $^{239/240}\text{Pu}$, ^{137}Cs , ^{90}Sr , etc.) presently stored in the underground waste tanks. Information used to establish global inventories originated from key process history records (e.g., essential material purchase records, various chemical flowsheets used in reprocessing of irradiated Hanford Site reactor fuels, and from calculations of radionuclide isotope generation and decay). The global inventory estimates are presented in Sections 5.0, 6.0, and 7.0.

Best-Basis Tank Inventories for each of the 177 single- and double-shell underground waste storage tanks on the Hanford Site have also been developed as part of the Best-Basis Inventory program. Best-Basis Tank Inventories are derived primarily from tank sample

analyses, process knowledge, and evaluation of the HDW Rev. 4 model (Agnew et al. 1997¹). The Best-Basis Tank Inventories are published in the respective Tank Characterization Report for each tank. In instances where a Tank Characterization Report has not been issued, the Best-Basis Tank Inventory is issued as a preliminary Tank Characterization Report. The Best-Basis Tank Inventories are also available in the Tank Characterization Database (LMHC 1998)². The Best-Basis Tank Inventories now serve as waste composition data for the TWRS process flowsheet modeling work, safety analyses, risk assessments, and waste retrieval, treatment, and disposal system design.

Additional details on the methodology for deriving global chemical and radionuclide inventories, as well as Best-Basis Tank Inventories, is presented in this report in Section 8.0.

Best-Basis Tank Inventories have been developed for 25 chemical and 46 radionuclide components. Global inventories are available for five chemicals in addition to the chemicals and radionuclides reported in the Best-Basis Tank Inventories. The chemical analytes selected represent over 99 weight percent of the tank contents, and the radionuclides represent over 99 percent of the activity.

The Best-Basis Tank Inventory summation (as of September 30, 1998) and the respective global values are shown in Tables ES-1 and ES-2. Thirteen of the tank summation and global chemical inventories match within 25 percent. For these 13 components, seven match within 10 percent (Table ES-2). More than 50 percent of the tank summation inventory is based on sample data for 19 of the 25 chemicals. Comparison of tank summation and global radionuclide inventories (Table ES-2) is less meaningful since the two inventories are often derived from the HDW Rev. 4 model and, therefore, are not truly independent of one another.

¹ Agnew, S. F., J. Boyer, R. A. Corbin, T. B. Duran, J. R. FitzPatrick, K. A. Jurgensen, T. P. Ortiz, and B. L. Young, 1997, Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 4, LA-UR-3860, Los Alamos National Laboratory, Los Alamos, New Mexico.

²LMHC, 1998, Best-Basis Inventory TWRS Planning Baseline as of 10/1/98, Tank Characterization Database, Internet at <http://twins.pnl.gov:8001/TCD/main.html>, Lockheed Martin Hanford Corporation, Richland, Washington.

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Table ES-1. Comparison of Global Inventory and Best-Basis Tank Inventory Summation for Nonradioactive Components (Metric Tonnes). (2 Sheets)

<i>Component</i>	<i>Best-Basis Tank Inventory summation¹</i>	<i>Global (process history-based) inventory</i>	<i>Corresponding document section for global inventories</i>
<i>Aluminum</i>	7,950	7,845	5.1
<i>Bismuth</i>	631	580	5.2
<i>Calcium</i>	319	214	5.3
<i>Carbonate</i>	9,390	4,830	5.4
<i>Chloride</i>	930	500	5.6
<i>Chromium</i>	670	785	5.7
<i>Fluoride</i>	1,190	1,360	5.8
<i>Hydroxide</i>	23,500	23,000	5.9
<i>Iron</i>	1,400	1,230	5.10
<i>Lanthanum</i>	51.3	51	5.11
<i>Lead</i>	84.0	279	5.12
<i>Manganese</i>	194	105	5.13
<i>Mercury</i>	1.81	2.1	5.14
<i>Nickel</i>	174	111	5.15
<i>Nitrite and Nitrate</i>	64,700	85,700	5.16
<i>Phosphate</i>	5,550	6,000	5.17
<i>Potassium</i>	874	481	5.18
<i>Silicon</i>	941	570	5.19
<i>Sodium</i>	48,800	54,200	5.20
<i>Sulfate</i>	3,330	5,000	5.21
<i>Strontium</i>	45.9	31.3	5.22
<i>Total organic carbon</i>	1,690	4,000	5.23
<i>U_{TOTAL}</i>	929	965	6.3
<i>Zirconium</i>	470	440	5.24

Table ES-1. Comparison of Global Inventory and Best-Basis Tank Inventory Summation for Nonradioactive Components (Metric Tonnes). (2 Sheets)

Component	Best-Basis Tank Inventory summation ¹	Global (process history-based) inventory	Corresponding document section for global inventories
<i>Minor components</i>			
Cadmium	NR ²	8.2	7.3.1
Cerium	NR ²	8.8	5.5
Silver	NR ²	8.93	7.3.2
Thorium	NR ²	25.6	7.3.3
Tungsten	NR ²	15.9	7.3.4

¹As of September 30, 1998

²NR = Not reported. Only small quantities of these chemicals are present in the waste and analytical data are limited.

Table ES-2. Comparison of Global Inventory and Best-Basis Tank Inventory Summation for Radioactive Components (Decayed to January 1, 1994). (3 Sheets)

Radionuclide	Best-Basis Tank Inventory summation (Ci) ¹	Global (process history-based) inventory (Ci)	Corresponding document section for global inventory
³ H	2.45 E+04	3.40 E+04	6.1
¹⁴ C	3.87 E+03	4.81 E+03	6.1
⁵⁹ Ni	8.76 E+02	9.34 E+02	6.1
⁶⁰ Co	1.99 E+04	1.23 E+04	6.1
⁶³ Ni	8.64 E+04	9.20 E+04	6.1
⁷⁹ Se	7.11 E+02	7.73 E+02	6.1
⁹⁰ Sr	5.86 E+07	7.16 E+07	6.1 and 6.2
⁹⁰ Y	5.86 E+07	7.16 E+07	6.1
⁹³ Zr	3.45 E+03	3.63 E+03	6.1
^{93m} Nb	2.53 E+03	2.69 E+03	6.1
⁹⁹ Tc	2.83 E+04	3.26 E+04	6.1
¹⁰⁶ Ru	1.26 E+05	1.04 E+05	6.1

Table ES-2. Comparison of Global Inventory and Best-Basis Tank Inventory Summation for Radioactive Components (Decayed to January 1, 1994). (3 Sheets)

Radionuclide	Best-Basis Tank Inventory summation (Ci) ¹	Global (process history-based) inventory (Ci)	Corresponding document section for global inventory
^{113m} Cd	1.66 E+04	1.69 E+04	6.1
¹²⁵ Sb	2.52 E+05	2.08 E+05	6.1
¹²⁶ Sn	1.17 E+03	1.19 E+03	6.1
¹²⁹ I	9.91 E+01	6.30 E+01	6.1
¹³⁴ Cs	8.68 E+04	8.89 E+04	6.1
¹³⁷ Cs	5.29 E+07	4.64 E+07	6.1 and 6.2
^{137m} Ba	5.01 E+07	4.39 E+07	6.1
¹⁵¹ Sm	2.60 E+06	2.75 E+06	6.1
¹⁵² Eu	1.45 E+03	1.48 E+03	6.1
¹⁵⁴ Eu	1.94 E+05	1.47 E+05	6.1
¹⁵⁵ Eu	1.75 E+05	1.36 E+05	6.1
²²⁶ Ra	5.83 E-02	6.31 E-02	6.1
²²⁷ Ac	8.75 E+01	8.76 E+01	6.1
²²⁸ Ra	7.75 E+01	7.71 E+01	6.1
²²⁹ Th	1.80 E+00	1.81 E+00	6.1
²³¹ Pa	1.56 E+02	1.56 E+02	6.1
²³² Th	4.41 E+00	2.11 E+00	6.1
²³² U	1.32 E+02	1.23 E+02	6.1
²³³ U	5.01 E+02	4.76 E+02	6.1
²³⁴ U	3.47 E+02	3.46 E+02	6.1
²³⁵ U	1.41 E+01	1.45 E+01	6.1
²³⁶ U	1.19 E+01	9.57 E+00	6.1
²³⁷ Np	1.84 E+02	1.41 E+02	6.1
²³⁸ Pu	2.69 E+03	2.77 E+03	6.1
²³⁸ U	3.10 E+02	3.22 E+02	6.1 and 6.3
²³⁹ Pu	5.52 E+04	3.91 E+04	6.1
²⁴⁰ Pu	1.11 E+04	8.93 E+03	6.1

Table ES-2. Comparison of Global Inventory and Best-Basis Tank Inventory Summation for Radioactive Components (Decayed to January 1, 1994). (3 Sheets)

<i>Radionuclide</i>	<i>Best-Basis Tank Inventory summation (Ci)¹</i>	<i>Global (process history-based) inventory (Ci)</i>	<i>Corresponding document section for global inventory</i>
²⁴¹ Am	1.06 E+05	6.99 E+04	6.1
²⁴¹ Pu	1.66 E+05	2.29 E+05	6.1
²⁴² Cm	1.71 E+02	7.70 E+01	6.1
²⁴² Pu	1.02 E+00	1.16 E+00	6.1
²⁴³ Am	1.76 E+01	9.34 E+00	6.1
²⁴³ Cm	3.28 E+01	1.00 E+01	6.1
²⁴⁴ Cm	7.82 E+02	2.42 E+02	6.1

¹As of September 30, 1998.

Appendix K presents an expanded discussion of the difference between the Best-Basis Tank summation and global inventory values and also includes a discussion of recently identified issues and biases that may impact resolution of discrepancies between the two sets of inventory values. As additional data are obtained, and increased historical data become available, it is expected that the Best-Basis Tank summation and global inventory differences can be fully reconciled for most major waste components.

In the interim, it is recognized that users require a single Tank Farm Total Best-Basis Inventory value by analyte. Since the Best-Basis Tank Inventory summations are in use by the Tank Waste Remediation System program, it is recommended that these same values be used by other end users of the inventory data. The global inventories presented in this report should be viewed as supplemental data that provides an alternate basis for comparison.

Development and maintenance of the Best-Basis Tank Inventories and the resulting Tank Farm Total Best-Basis Inventories is an on-going effort. As new waste sample and analytical data become available, respective Best-Basis Tank Inventories are typically adjusted

and reported in a revised Tank Characterization Report and in the Tank Characterization Database. Values may also be revised based on new or refined process knowledge. Specific data users may wish to use the revised individual tank data as needed for specific study purposes. Users are referred to the Tank Characterization Database for the most current Best-Basis Inventory data set. For users needing to fix on an inventory data set for an extended period, the Best Basis Inventory TWRS Planning Baseline as of October 1, 1998, has been established as a separate query on the Tank Characterization Database. Current plans call for update to this TWRS Planning Baseline Inventory on an annual basis.

It is recognized that the Best-Basis Inventory, as reported in Tables ES-1 and ES-2, will not satisfy all users' needs. The Best-Basis Inventory data set has already been expanded to include uncertainty estimates and wash/leach factors that are now available on the Tank Characterization Database in the Best-Basis Inventory/Uncertainty Data and Best-Basis Inventory/Wash and Leach Factors queries, respectively.

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STANDARD INVENTORIES OF CHEMICALS AND RADIONUCLIDES IN HANFORD SITE UNDERGROUND TANK WASTES

1.0 INTRODUCTION

After the primary mission at the Hanford Site changed from plutonium production to environmental restoration, the U.S. Department of Energy (DOE) established the Tank Waste Remediation System (TWRS) in 1991 to safely manage and dispose of the radioactive wastes stored in underground tanks. Key activities for the TWRS include overseeing tank farm operations and identifying, monitoring, and resolving the safety issues associated with those operations and with the tank wastes. Disposal activities involve designing equipment, processes, and facilities for retrieving wastes, and processing waste into a form that is suitable for long-term storage/disposal. Safety analyses, engineering evaluations, risk assessments, and regulatory issues connected with these activities require information about the chemical and physical properties of tank wastes. To support these activities, chemical information usually is required in the form of tank waste component inventories, on either a global (total) or tank-specific basis.

There are multiple sources of tank waste inventory information, often with inconsistent data. A task was initiated in fiscal year (FY) 1996 to establish a best-basis standard inventory for chemicals and radionuclides in Hanford Site tank waste. The goal is to resolve differences among the many reported inventory values and to provide a consistent inventory basis for all waste management and disposal activities. This report summarizes the work performed to establish standard tank component inventories on a global basis based on process knowledge; i.e., the global inventory.

Inventories have also been derived on an individual (tank-by-tank) basis as part of the Standard Inventory task. Individual tank inventories are based primarily on analyses of tank waste samples. Chemical and radionuclide data from the individual tanks (or the total tank farm inventories based on summation of the individual tank values) are presently considered the Best-Basis Inventories to support all present users of inventory data. Individual tank inventories are available in the Tank Characterization Database (LMHC 1998).

The emphasis of this report, however, is to estimate the global inventory for an analyte based nearly exclusively on process history. This independent estimate of tank waste inventories is derived primarily from essential material purchase records, from various chemical flowsheets used in reprocessing of irradiated Hanford Site reactor fuels, and from calculations of radionuclide isotope generation and decay. The global inventory complements and supplements the summation of the Best-Basis Inventories for the individual tanks by providing an alternative basis for comparison. As additional analytical data are obtained, and increased historical data become available, it is expected that the summed tank-by-tank inventories and global (process history based) inventory differences will be fully reconciled for most major waste components.

1.1 BACKGROUND

Total tank waste inventory information was previously reported in several sources including the following:

- Estimated Inventory of Chemicals Added to Underground Waste Tanks, 1944 Through 1975 (Allen 1976)
- Hanford Defense Waste Final Environmental Impact Statement (DOE 1987)
- Tank Remediation System, Hanford Site, Washington, Final Environmental Impact Statement (DOE 1996)
- Chemical and Radionuclide Inventory for Single and Double-Shell Tanks (Shelton 1996)
- The *Hanford Tank Content Estimate* (HTCE) documents (Brevick et al. 1997a, 1997b, 1997c, and 1997d)
- The Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 3 (Agnew et al. 1996) and the Hanford Tank Chemical and Radionuclide Inventories: HDW Model Rev. 4 (Agnew et al. 1997a), which report predictions of the Hanford Defined Waste (HDW) model developed by Los Alamos National Laboratory (LANL)
- National databases, such as the Federal Facility Compliance Act Database and Integrated Database (DOE 1997).

Each of the inventory reports listed contains the best inventory knowledge available at the time of publication. For example, inventories reported in the *Single-Shell and Double-Shell Tank Waste Inventory Data Package for the Tank Waste Remediation System Environmental Impact Statement* (Golberg and Guberski 1995) are based on those established by Allen (1976) but also include those chemicals used in process operations after 1975. The inventories reported by Shelton (1996), in support of TWRS process flowsheet modeling work, correspond with DOE (1987) values with two exceptions: (1) TWRS double-shell tank (DST) inventories were revised to reflect sampling data, and (2) TWRS single-shell tank (SST) component inventories for aluminum, chromium, phosphate, and chloride were revised as the result of a recent (1995) evaluation of these inventories. Finally, the HDW model predictions were based on historical data and do not directly correspond to any of the inventory reports. Not surprisingly, the various inventory sources provide inconsistent inventory values.

Several attempts have been made over the years to distribute component inventories among individual waste tanks. The distribution is performed by: (1) estimating component inventories using results from sample analyses, or (2) predicting component inventories based on process knowledge and historical information. The result is that tank-specific inventory sources also often report inconsistent inventory values.

The major sources of tank-specific inventories include the following:

- Agnew et al. (1997a), which reports predictions of the HDW model developed by LANL
- Tank Characterization Reports (TCRs)
- The HTCE documents (Brevick et al. 1997a, 1997b, 1997c, and 1997d)
- Radionuclide and Chemical Inventories for the Single-Shell Tanks (Van Vleet 1993a) and Radionuclide and Chemical Inventories for the Double-Shell Tanks (Van Vleet 1993b)
- Multiple electronic databases, e.g., Tank Characterization Database (TCD) (LMHC 1998), Braun Database (Forehand 1995), and ICF-Kaiser Hanford Company Database (Brevick et al. 1995).

The present effort to establish a best-basis inventory on a global and tank-specific basis involves a thorough review of all pertinent information sources to identify errors, biases, inconsistencies, and missing information. The data sources typically include sample analyses, process flowsheets, waste transaction records, reactor fuel data, and essential material records. The process used to estimate a best-basis inventory is described in Sections 5.0, 8.0, and Appendix J of this report.

This report presents work performed to establish standard global component inventory estimates. This work includes: global inventory estimates for 25 nonradioactive components that account for approximately 99 percent of the total tank waste inventory, global inventory estimates for an additional four (4) minor chemical components, and global inventory values for 46 radionuclides. Tank-by-tank best-basis inventories that include the same suite of chemicals and radionuclides for each of the 177 Hanford Site underground storage tanks are published in TCRs and revised TCRs issued in fiscal year (FY) 1998, and as preliminary TCRs. As previously mentioned, the individual tank waste inventories are also available in the TCD (LMHC 1998), and have been designated as the official source for the tank inventory values. To ensure that inventory values resulting from these evaluations are technically defensible and reproducible, every attempt was made to document the bases, i.e., originating sources of assumptions, data, and background information, that support the inventory values.

1.2 DOCUMENT OVERVIEW

Information in this report is presented as follows:

- Section 1.0 provides background information regarding tank inventories.
- Section 2.0 provides an overview of the Standard Inventory task plan and shows where the global component inventory work fits within this plan.
- Section 3.0 summarizes data requirements that served as the foundation for the global inventory work.
- Section 4.0 introduces the chemical processes that generated waste inventories, the types and amount of fuel processed, and the system inventory losses.
- Section 5.0 presents evaluations and global best-basis inventory values for individual components.
- Section 6.0 presents the methods used to establish global inventories for 46 radionuclides.
- Section 7.0 evaluates the sources of minor chemical components that contribute to the tank waste inventories and presents the global best-basis inventories for four minor components.
- Section 8.0 defines details of the methodology used to determine the tank-by-tank best-basis inventories for each of the 177 Hanford Site underground waste storage tanks.
- Section 9.0 lists the references used in this document.
- Appendix A provides a summary of the users' data needs for chemical and radionuclide inventories.
- Appendix B presents details of fuel processing history at the Hanford Site, including production data and fuel processing rates for the various separations facilities.
- Appendix C defines the flowsheet bases for the bismuth phosphate (BiPO_4) process wastes. This information is the standard basis for use in defining global and tank-by-tank inventories.
- Appendix D defines the flowsheet bases for the Reduction and Oxidation (REDOX) process wastes. This information is the standard basis for use in defining global and tank-by-tank inventories.

- Appendix E provides calculations supporting the global aluminum inventory evaluation (Section 5.1).
- Appendix F provides calculations supporting the global lead inventory evaluation (Section 5.12).
- Appendix G provides sample fuel activity records, supporting data and assumptions referred to in the global radionuclide inventory evaluation (Section 6.0).
- Appendix H defines the basis for corrosion product (iron, nickel, and chromium) inventories.
- Appendix I defines limitations on content of the Standard Inventory.
- Appendix J contains guidance for preparing tank-by-tank inventory evaluations (i.e., best-basis inventories).
- Appendix K discusses the reconciliation status of the sum of the individual tank best-basis inventories with the global tank waste inventories.

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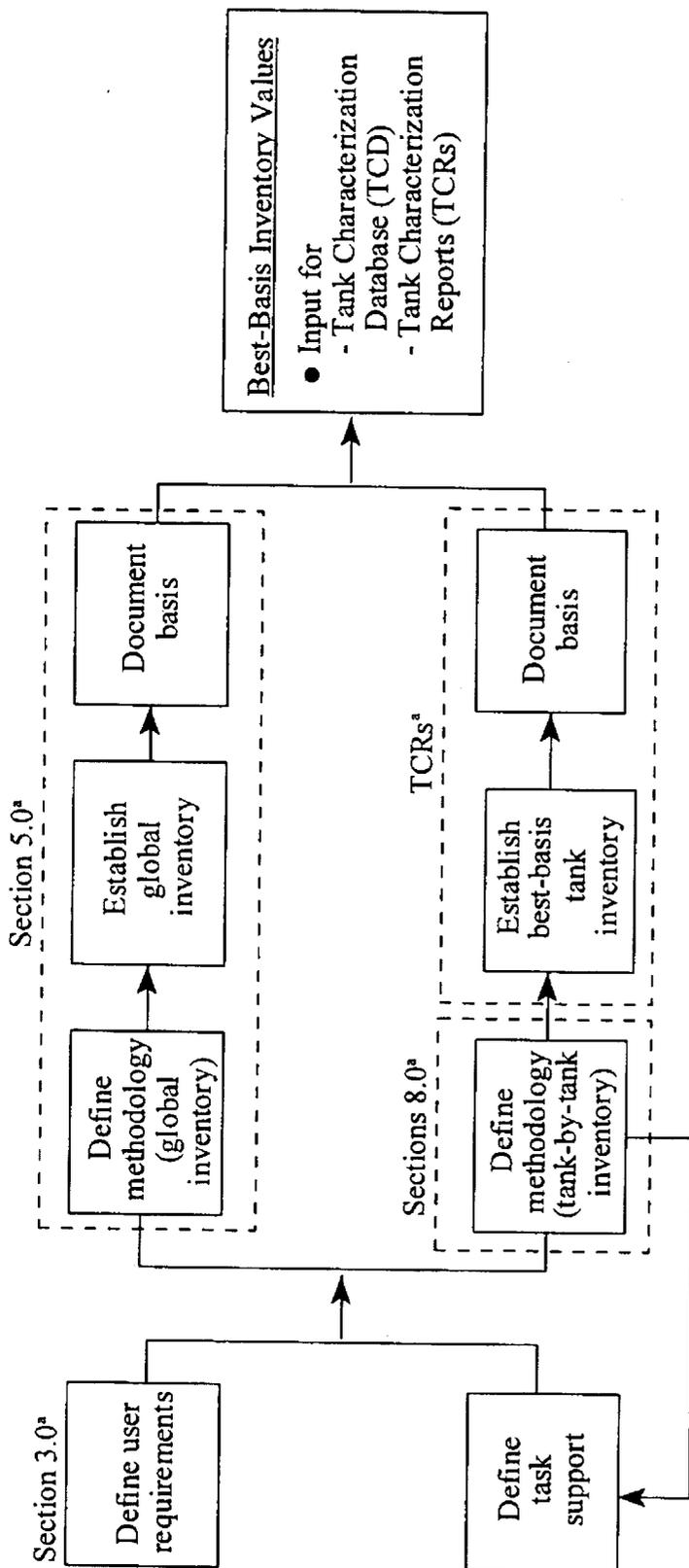
2.0 STANDARD INVENTORY TASK OVERVIEW

The primary objective of the Standard Inventory task is to establish best-basis waste inventory estimates that provide consistent data for a multitude of activities related to the safe storage of the tank waste and its eventual treatment and disposal. Additional objectives within the work scope included ensuring that this information is incorporated into the TCD (LMHC 1998), documented in a published report, and maintained in a structured and controlled manner.

The approach used to implement the Standard Inventory task objectives is defined in the *Work Plan for Defining a Standard Inventory Estimate for Wastes Stored in Hanford Site Underground Tanks* (Hodgson and LeClair 1996) and is schematically depicted in Figure 2.0-1. Specific activities within this plan involved the following:

- Identification of waste components and characteristics, i.e., data requirements, that are important to waste management and disposal activities. The results from this activity are defined in Appendix A and further summarized in Appendix I and Section 3.0.
- Development and documentation of the methodology used to evaluate both global and tank-by-tank inventories. The global and tank-by-tank inventory evaluation methodologies are defined in Sections 5.0 and 8.0, respectively.
- Determination of global inventory values for specified waste components by evaluating the bases, i.e., assumptions, historical data, and background information, that support these values. The results of this effort are documented in Section 5.1 through Section 5.24. This effort complements the independent determination of tank-by-tank waste summation inventories (described below) which are presently considered the best-basis inventories for end-user planning purposes.
- Determination of tank-by-tank waste inventories from evaluating available sampling data and historical information. The results of this effort are documented in each of the respective TCRs or preliminary TCRs, and are presently considered the best-basis inventories with official access in the TCD. Appendix K discusses reasons for variation between the individual tank best-basis inventories and the global tank waste inventories.
- Publication of the tank-by-tank inventory results in a form that makes the data easily accessible to data users, makes it easy to update the data set, and also allows for archiving of the data and the associated pedigree (usually published as a supporting document). The TCD (LMHC 1998) was recognized as an existing resource for providing these capabilities.
- Development of a mechanism for maintenance and update of the Standard Inventory. The results of this effort are discussed later in this section.

Figure 2.0-1. Schematic of Task Plan.



TCRs = Tank Characterization Reports

^a Refers to location where applicable documentation can be located.

2.1 STANDARD INVENTORY MAINTENANCE

For the Standard Best-Basis Inventory values to remain useful, they must be periodically updated and modified to reflect new information on tank contents. As sample analysis results, modeling data, and other information become available, they must be evaluated and reconciled with existing data in much the same fashion that the initial Standard Inventory values were derived. Revised Standard Inventory values and their associated pedigree (supporting documentation) must then be approved by the change control process and incorporated into the TCD. In instances where the prompt for update is new sample analysis results, the best-basis evaluation will also be documented in the respective TCR. Tank-by-tank inventories may change during the year based, e. g., on new sample data and other fundamental work. This "current" tank waste best-basis inventory will be accessible by TWINS queries on the TCD. In addition, for consistency among data users in preparing strategic documents, an annual reconciliation of the summed inventories will be completed at the beginning of the fiscal year. Subsequent data will not be entered into the TWRS Planning Baseline inventory in the TCD until the following annual update.

A Standard Inventory change control process was developed during Standard Inventory User and Policy Workshops conducted in July 1996 (LeClair 1996). Procedures and policies define the protocol to be followed for proposing changes to the inventory, approving changes to the inventory, and updating the TCD to report revised Standard Inventory values. Responsibilities, authorities, methodology application, quality assurance practices, and database configuration control are also addressed by these procedures and policies.

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3.0 DATA REQUIREMENTS

The primary objective of the Standard Inventory task is to provide reliable waste inventory data that will benefit several safety, waste management, and disposal related activities. Examples of such activities include the following:

- Performance of safety analyses of tank farm operations and of tank wastes
- Performance of risk assessments associated with storage and/or processing of tank wastes
- Design, development, and implementation of waste retrieval, treatment, and disposal systems
- Conduct of performance assessments to evaluate long-term potential doses and risks from waste form disposal systems
- Obtaining regulatory permits.

To ensure that this task encompassed data needs for all the above-listed activities, an initial effort in October 1995 involved reviewing existing Data Quality Objective (DQO) documents and contacting cognizant personnel to identify specific data requirements. The results from this effort are contained in Appendix A, Summary of Data Requirements.

3.1 STANDARD INVENTORY CONTENT LIMITATIONS

The data requirements defined in Appendix A are quite extensive and, since available resources were limited, it was necessary to focus this task on a subset of the Appendix A data requirements. A discussion of the content limitations and the resulting set of chemicals and radionuclides addressed by the Standard Inventory task is provided in Appendix I. This subset of chemicals and radionuclides was reviewed with data users during the Standard Inventory User and Policy Workshop conducted in July 1996 (LeClair 1996).

3.2 EXPANSION OF THE STANDARD INVENTORY CONTENT

As originally conceived, the Standard Inventory would be the inventory data set having most value to the largest number of users. As a second objective, the Standard Inventory must also be updated and modified on a regular basis so that it reflects the most current information available on tank contents. The Standard inventory will also be updated and modified in response to evolving user needs. This second objective imposes some limit on the size of the Standard Inventory data set, therefore, it is recognized that it will not meet every need of all users.

As currently defined, the standard inventories are single values per analyte for the whole of the 177 tanks and for the whole of each tank (tank-by-tank inventory). The need to include some measure of precision (e.g., confidence interval, error bars, uncertainty estimates, etc.) on each of these analyte inventories has already been recognized and was implemented in the TCD early in fiscal year 1999.

Several other user needs have been identified over the course of the Standard Inventory task including reporting of chemical compound inventories, reporting of the soluble and insoluble fractions of the analyte waste inventory by tank, and reporting of the analyte inventories by liquid and solid phase for each tank. Soluble and insoluble fractions were recently incorporated as part of the best-basis inventory via the TCD. Some of the program-specific needs can often be extracted from the data presented in the best-basis documentation or within the sample analytical data reported in the TCR for each tank. As additional program needs are identified, it is anticipated that the Standard Inventory will be expanded to report the needed data within the TCD, but such data may not be included in the best-basis inventory discussion of each TCR.

5.0 BEST-BASIS GLOBAL INVENTORY EVALUATIONS

Global component inventories for SST and DST wastes were initially developed as composition data for TWRS flowsheet modeling, safety analysis, and risk assessment. Subsequent to development of the global inventories the individual tank best-basis inventories were developed and the summation of the best-basis inventories were established as the preferred baseline inventory. The global inventories are currently considered a complementary source of information to the best-basis inventory summation.

The global inventories were primarily developed from key historical records, e.g., essential material purchase records, from various chemical flowsheets in the processing of irradiated Hanford Site reactor fuels, and from calculations of radionuclide generation and decay. These sources and previous inventory estimates often provide conflicting inventory estimates. The methodology for resolving these differences is depicted schematically in Figure 5.0-1.

Data quality for the work described in this section was maintained by adhering to the following steps:

1. Critically examine all input data and assumptions used in the TWRS Inventory and HDW model inventory calculations to identify any discrepancies, errors, missing information, etc.

Identify waste types that contributed to the majority of the component inventory.

Evaluate process stream compositions from chemical flowsheets.

Review amount of fuel processed, waste transfers, e.g., crib discharges, purchase records.

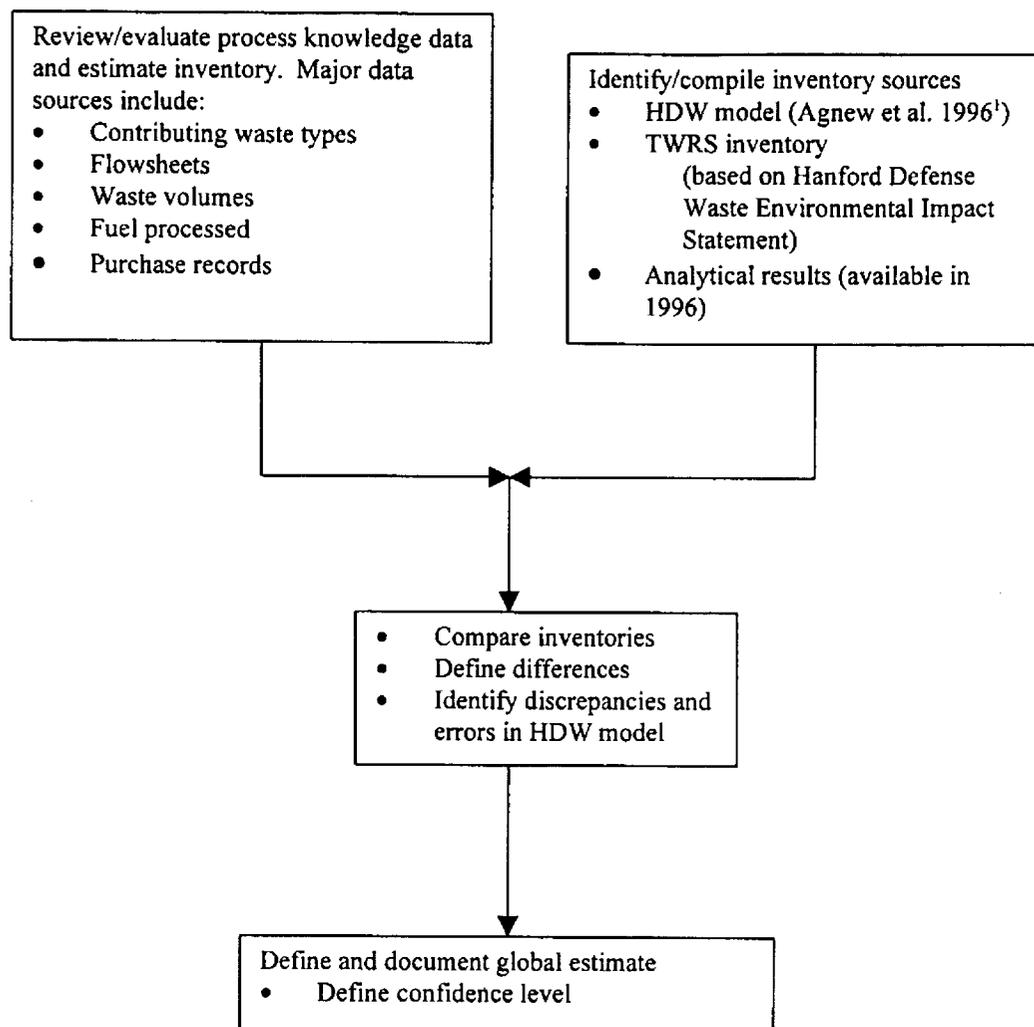
Review/evaluate tank waste sample analyses.

2. Document all bases and obtain agreement among the majority of technical experts that the revised input data are sound.

Retracing the evolution of data has proved challenging, and every attempt is being made to document original sources of data and information to ensure that inventory values resulting from these evaluations are technically defensible and reproducible. The following Sections 5.1 through 5.24 present global inventory evaluations for 25 chemicals.

Several sections provide tables that total the various mass contributions for a chemical component. At the author's discretion, these totals are often rounded to two or three significant figures. The summation of individual values may not be exactly equal to the rounded value presented.

Figure 5.0-1. Methodology for Developing Global Inventory Values for Major Components.

**Legend**

HDW = Hanford Defined Waste

TWRS = Tank Waste Remediation System

¹The HDW model inventories reflect the global values reported in HDW model Rev. 3 (Agnew et al. 1996) rather than the global values from HDW model Rev. 4 (Agnew et al. 1997). This is due to the fact that most of the global inventory evaluations reported in Sections 5.1 through 5.24 in Section 5.0 were conducted in fiscal year 1996 and used HDW Rev. 3 model as the basis for comparison.

5.1 ALUMINUM

The primary sources of aluminum (Al), a major constituent in tank wastes, were: (1) the Al cladding on the irradiated fuel (greater than 90 wt% of the fuel processed at the Hanford Site was aluminum-clad), and (2) the Al added as aluminum nitrate nonahydrate (ANN) - $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ as a salting agent in the REDOX solvent extraction process. Smaller sources of aluminum were the aluminum canisters used to contain the early New Production Reactor (NPR) (N Reactor) fuels processed at the REDOX Plant in 1965 and 1966, ANN salting agent for the Plutonium Finishing Plant (PFP) solvent extraction system, and aluminum added as ANN to complex fluoride ion, thereby reducing the corrosion of the stainless steel process vessels and piping. The PUREX Plant used ANN for this purpose during thorium fuel processing and zirconium-clad fuel decladding (Zirflex process), and all plants used ANN when fluoride ion was used in flushes.

The total mass of Al in the tank waste is important since the amount is so large that it will greatly impact the volume of the final waste glass products. The only larger mass metallic component is sodium (see Table 5.0-1). Aluminum is relatively widespread throughout the waste tanks. Aluminum from the bismuth phosphate process decladding waste was combined with the first decontamination cycle (1C) wastes. Solids from this waste stream, including aluminum solids, preferentially settled in the first tank of the 1C cascades. When the 1C supernatants were later concentrated, more solids crystallized out in the evaporator bottoms tanks. Aluminum concentrations are high in almost all the REDOX process waste tanks (primarily S, SX, and U farm tanks), both from the ANN additions and the fact that the REDOX Plant cladding wastes and concentrated solvent extraction wastes were not initially segregated in the waste tanks; wastes were segregated after 1954. The PUREX process aluminum cladding wastes were always routed to the C farm tanks; however, the waste supernatants were later concentrated in the In-Tank Solidification (ITS) systems (bottoms in BY farm) and the 242-T Evaporator (bottoms in the TX farm). Eventually the supernatants were further concentrated in the vacuum evaporator-crystallizers, 242-S for 200 West Area and 242-A for 200 East Area.

5.1.1 Identification of Aluminum Inventory Values

The reported global aluminum inventory values are 7,720 MT (HDW model, Agnew et al. 1996) and 4,960 MT (TWRS, Shelton 1996). A separate estimate is provided below to verify these values. The estimate details are shown in Table 5.1-1.

¹ The HDW model inventories reflect the global values reported in HDW model Rev. 3 (Agnew et al. 1996) rather than the global values from HDW model Rev. 4 (Agnew et al. 1997). This is due to the fact that most of the global inventory evaluations reported in Sections 5.1 through 5.24 in Section 5.0 were conducted in fiscal year 1996 and used HDW Rev. 3 model as the basis for comparison.

Pages 5-4 through 5-6 intentionally deleted.

6.0 RADIONUCLIDES

Determination of the radionuclide inventories involves consideration of the date the radionuclides were generated in the reactor, the particular process through which they were subsequently processed, and the decay rate and decay chain specific to each isotope. Section 6.1 provides the basis used to develop inventory estimates of the 46 key radionuclides. Most of these radionuclides were considered waste and were directly discharged to the various process waste streams. However, some of the isotopes were considered to have beneficial use. Most notable in this respect were the isotopes of Pu, U, ^{237}Np , ^{137}Cs , and ^{90}Sr . Due to the unique aspects of recovering these materials, a separate discussion of ^{137}Cs and ^{90}Sr disposition is provided in Section 6.2, and uranium is discussed in Section 6.3.

6.1 ESTABLISH MODEL-BASED GLOBAL INVENTORY FOR RADIONUCLIDES

Historically, global inventory values for Hanford Site waste tanks have been generated by reactor fuel activity codes with supplemental separations plant waste analysis data for certain radionuclides. Until recently, analysis of waste samples taken from the tanks has not been relied on to predict total quantities since not enough tanks have been sampled and comprehensively analyzed. Past calculational methods have been as follows:

- RIBD-II Code (Gumprecht 1968) for fission products plus hand calculations for activation products
- The TRAC model, which used factors derived from the RIBD code
- The RADNUC code (Schwarz 1995), which uses tables of Ci/MTU factors generated by ORIGEN2
- A simplified ORIGEN2 code calculation (Wittekind 1989) that scales annual radionuclide production proportional to annual fuel exposure (megawatt days).

Results from the RIBD Code and TRAC model methods were published as Engineering Support Data (RHO 1985a) for the *Hanford Defense Waste Final Environmental Impact Statement* (DOE 1987) and are also reported to the national Integrated Database Report (DOE 1994). Unfortunately, an adequate documentation trail for these global inventory data sets no longer exists, and the primary codes, RIBD and TRAC, are no longer operational on Hanford Site computers.

The RADNUC code has been successfully used to predict radionuclide inventories in fuel and PUREX process waste associated with N Reactor operations from 1972 through 1987. However, significant code modifications would be required before RADNUC could be used to predict contributions to the global inventory from pre-1972 operations.

The simplified ORIGEN2 method has been recently used to estimate global inventories (i.e., for all reactors, 1944 to present) and to predict radionuclide compositions for LLW (Schmittroth 1995). Results generated by the ORIGEN2 model should be relatively easy to document since the ORIGEN2 code, itself, is well documented and accepted.

However, for the task of generating tank-by-tank inventory estimates for radionuclides, neither the RADNUC code nor the simplified ORIGEN2 code alone can easily provide the month-by-month definition of waste stream activities that is needed. Even the task of modelling global radionuclide inventories requires complex modelling because global inventory values must agree with the sum of tank-by-tank inventory values and to make this agreement possible much of the month-by-month data describing separation plant production must be incorporated in the global calculation. Thus, the global calculation takes on the complexity of the tank-by-tank calculation.

The following sections describe the requirements for the overall modelling task and the model architecture that has been set up to carry out the global and tank-by-tank estimates. Tables of supporting data and assumptions are given in Appendix G of this report. Section 6.1.8 summarizes best-basis radionuclide inventory data for the tank system, listing values for the 46 key radionuclides.

6.1.1 Requirements for a Global Tank Radionuclide Inventory Model

The task of calculating global inventories for many radionuclides is more complicated than in the case of chemicals. Not only must radionuclide fill-data (additions to tanks) be determined but, in the case of radionuclides, the concurrent effects of decay and buildup also must be computed. The waste activity contribution of short-cooled, low-exposure fuel irradiated in the forties and early fifties is uniquely different from that of longer-cooled, higher-exposure fuel irradiated in the seventies. It is obvious that one or more computer codes or models is needed to support this type of calculation.

Specifically, the calculation must begin with a "fuel activity generation" model such as ORIGEN2 to calculate the curie levels of fission products, activation products and actinides in individual batches of fuel discharged from Hanford Site reactors. Modelling of curie levels in discharged reactor fuel needs to account for variables of: (1) production (tons of fuel sent to separations plants per month), (2) fuel type (uranium enrichment or ^{235}U concentration as well as cladding type), (3) impurity levels affecting the generation of activation products, and (4) fuel exposure (megawatt days per metric ton of fuel [MWd/MTU]).

The ORIGEN2 code or other code must account for the effects of radionuclide decay during variable fuel cooling periods in reactor storage basins and later during waste aging periods in storage tanks. The buildup of ^{241}Am in the fuel and waste is particularly sensitive to effects of fuel exposure and reactor-to-separations cooling time.

Next, the chemical separation of extractable elements (Tc, U, Th, Np, and Pu) and volatile elements (^{14}C , Kr, I, and Xe) must be modelled so as to predict the quantities of these radionuclides entering the system of 177 waste tanks.

Finally, a waste chemistry model must be employed to account for the effect of radionuclide solubilities and the loss of certain radionuclides from the tank system during the transfer of waste supernatants to cribs during the mid-fifties.

6.1.2 Requirements for a Tank-by-Tank Radionuclide Inventory Model

In addition to the global requirements, the overall model must calculate tank-fill data (separation plant output) for each of several waste streams and over relatively short-time intervals (monthly or shorter) so that the quantities of radionuclides entering specific receiving tanks can be properly accounted for. The model must also account for solubility effects on the transfer of certain radionuclides from tank-to-tank during the subsequent history of waste transfers.

6.1.3 Radionuclides to be Tracked

Forty-six key radionuclides have been identified for tracking in development of a best-basis inventory for Hanford Site tank wastes (see Table 6.1-1 and Appendix A of this report). Of these 46 radionuclides, five (^{14}C , ^{59}Ni , ^{63}Ni , ^{60}Co , and ^{125}Sb) are generated totally or in part by neutron activation of minor alloy or impurity elements present in the array of Hanford reactor fuels. The calculation of inventory values for these five radionuclides is therefore more uncertain due to the necessary approximations made in setting concentration parameters for these precursor elements in the fuels.

Another subgroup of key radionuclides (^{226}Ra , ^{227}Ac , ^{228}Ra , ^{229}Th , and ^{231}Pa) deserves mention. These are alpha decay chain daughters of parent radionuclides such as ^{232}Th , ^{233}U , ^{234}U , and ^{235}U . With the exception of ^{228}Ra , these daughter nuclides are expected to slowly buildup in Hanford tank wastes, not reaching peak activity levels until thousands of years in the future. (The ^{228}Ra will likely peak much sooner). Data users must recognize that the grossly approximate estimates for these five nuclides presented, herein, are of little value in the long term waste migration scenarios for which they are of concern. Further decay calculations derived from inventory values of the parent nuclides will be required to support long term release scenario evaluations. For this reason, no attempt has been made to refine the estimate of near-term activity for these five nuclides.

Table 6.1-1. Key Radionuclides to be Added to the Hanford Defined Waste Model.

^3H	^{129}I	^{233}U
^{14}C	^{134}Cs	^{234}U
^{59}Ni	^{137}Cs	^{235}U
^{60}Co	$^{137\text{m}}\text{Ba}$	^{236}U
^{63}Ni	^{151}Sm	^{237}Np
^{79}Se	^{152}Eu	^{238}Pu
^{90}Sr	^{154}Eu	^{238}U
^{90}Y	^{155}Eu	^{239}Pu
^{93}Zr	^{226}Ra	^{240}Pu
$^{93\text{m}}\text{Nb}$	^{227}Ac	^{241}Am
^{99}Tc	^{228}Ra	^{241}Pu
^{106}Ru	^{229}Th	^{242}Pu
$^{113\text{m}}\text{Cd}$	^{231}Pa	^{242}Cm
^{125}Sb	^{232}Th	^{243}Am
^{126}Sn	^{232}U	^{243}Cm
		^{244}Cm

6.1.4 Requirements for Output Reporting

Output from the overall model should provide both global and tank-by-tank inventories for the 46 key radionuclides with activity values decayed to January 1, 1994. In future versions the total elemental mass of these radionuclides also should be tracked and reported.

6.1.5 Model Architecture

One relatively straightforward way of generating global and tank-by-tank inventory values for these key radionuclides was to modify the Hanford Defined Waste (HDW) model to carry data fields for these 46 radionuclides. This existing model provides the calculational framework to identify receiver tanks for the various waste streams exiting the separations plants and to model subsequent tank-to-tank and tank-to-crib waste transfers and solubility effects.

Providing the HDW model with the necessary radionuclide input data is a Fortran code named DKPRO plus a number of spreadsheets, and existing databases linked together as illustrated by the data flow schematics of Figures 6.1-1 and 6.1-2, and described in Section 6.1.5.1.

An alternative way of generating global inventory values is also depicted in Figure 6.1-2. Here, the same fuel activity output file generated by the DKPRO code is coupled to a Hanford spreadsheet (named "SPLIT" for discussion purposes, here) which duplicates the role of the HDW model in accounting for the removal of certain radionuclides during fuel separations processing and waste fractionation operations. The spreadsheet SPLIT applies waste loss factors to individual fuel batches and calculates total inventory values accumulated in tank waste. Both of these model architectures are discussed, below, and compared.

6.1.5.1 ORIGEN2 and DKPRO Codes. At the center of Figure 6.1-1 is a box representing the newly created fortran code named DKPRO. The objective of the DKPRO code is to generate an output file of "fuel activity records"--curie values for 46 key radionuclides contained in batches of reactor fuel input monthly (or for shorter periods) to individual separations plants. The code, DKPRO, has the general capability to estimate radionuclide activity values in batches of reactor fuel (or fuel separations plant waste), using output from the ORIGEN2 code. However, to avoid an excessive number of ORIGEN2 calculations a "table interpolation technique" is used. Output from 15 ORIGEN2 runs, spanning various reactor fuel types and exposure levels, are used as a data library in DKPRO. The job of DKPRO is to read a second data file, the "Unified Separations Production File," and for each of approximately 1,300 fuel batch records (e.g., data on fuel type, fuel tonnage, exposure, separations date, and cooling time) calculate radionuclide activities in that batch of fuel by interpolating the ORIGEN2 output data library as a function of fuel type and exposure (MWd/MTU). (See Watrous and Wootan [1997] for further details of the ORIGEN2 and DKPRO code setup and for a discussion of how the Unified Separations Production File was generated.)

For the purpose of this radionuclide modelling task, Hanford Site fuels have been classified in six fuel types, as defined in Table 6.1-2.

Figure 6.1-1. Data Flow Schematic--Generation of Fuel Activity Records from ORIGEN2.

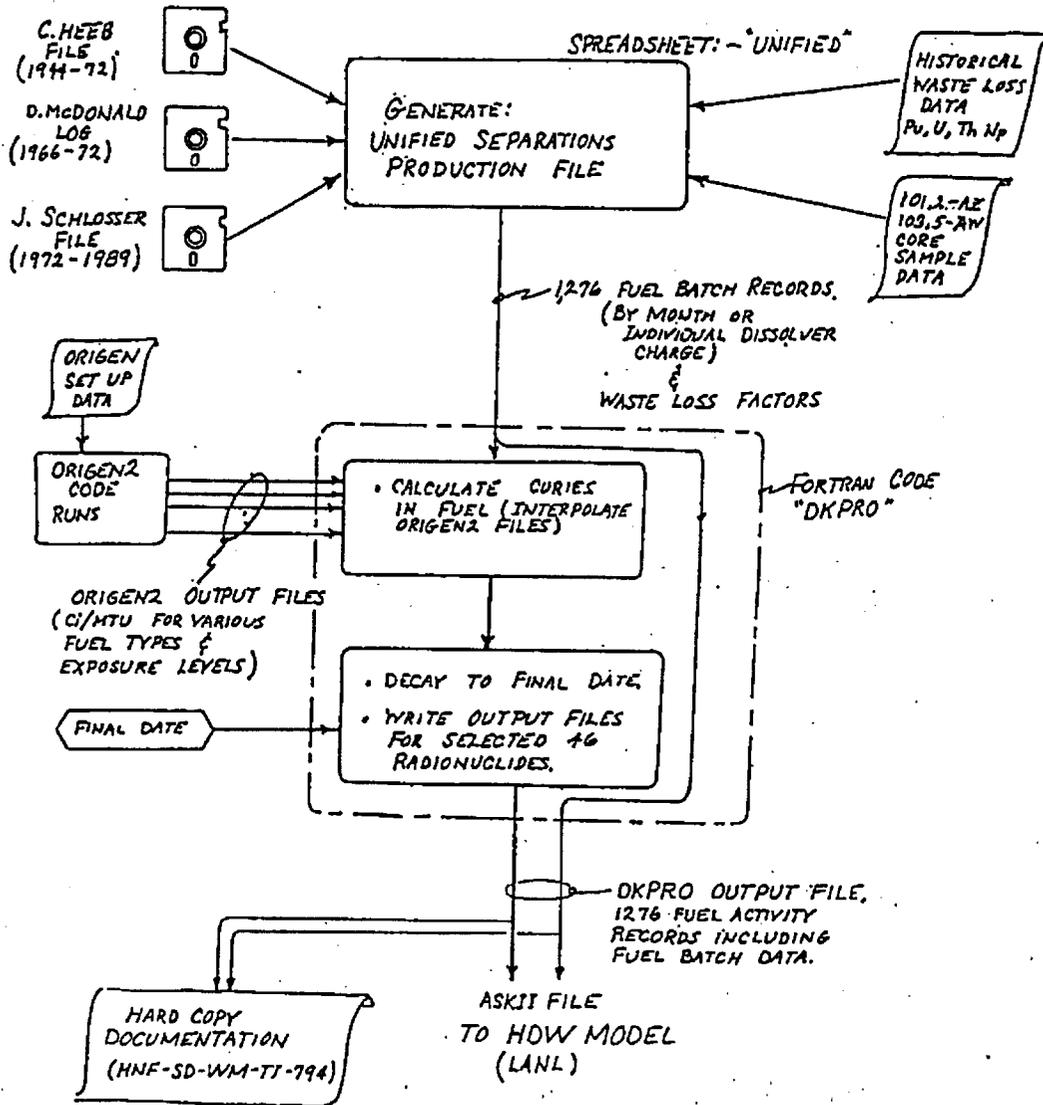


Figure 6.1-2. Data Flow Schematic--Generation of Tank Radionuclide Inventories.

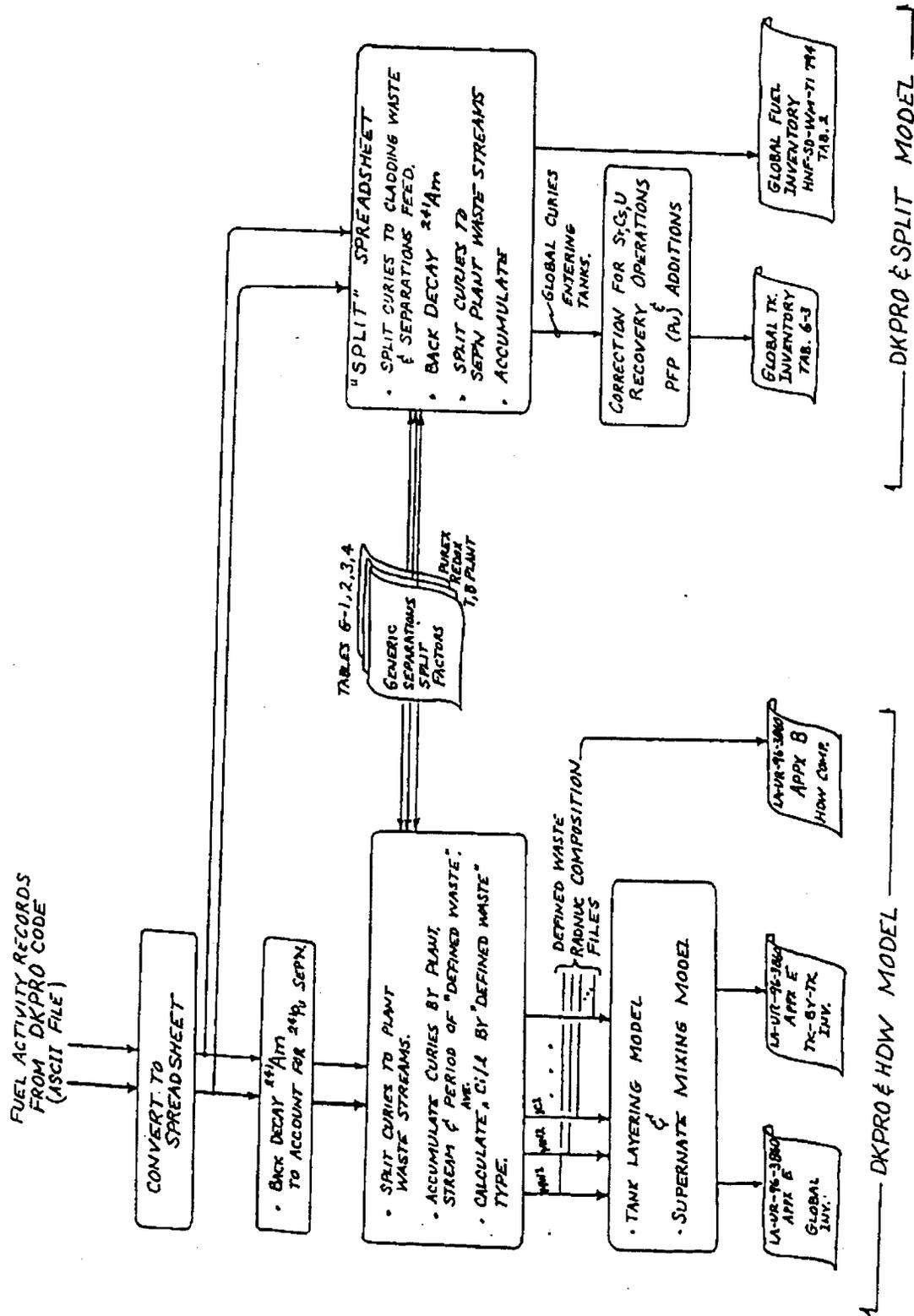


Table 6.1-2. Definition of Fuel Types and Fuel Codes for Radionuclide Inventory Modelling.

Code	Fuel definition
Al-0.71U	Aluminum clad, natural enrichment uranium (0.71 percent ²³⁵ U) -- either solid core or I&E. Single pass reactor fuel.
Al-0.94U	Aluminum clad, enriched uranium (0.94 percent ²³⁵ U). Single pass reactor fuel.
Zr-0.94U	Zircaloy®-clad, enriched uranium (0.94 percent ²³⁵ U). N Reactor fuel.
Zr-1.1U	Zircaloy®-clad, spike uranium (0.94 percent ²³⁵ U inner fuel tube and 1.25 percent ²³⁵ U outer fuel tube, together averaging 1.1 percent ²³⁵ U). N Reactor fuel.
Zr-2.1U	Zircaloy®-clad, co-product driver uranium (2.1 percent ²³⁵ U). N Reactor fuel.
Al-0.0Th	Aluminum-clad, thorium oxide target elements. Single pass reactors.

In this current version of the DKPRO code/HDW model interface, the DKPRO code calculates radionuclide decay from the date of fuel discharge to the "standard inventory reference decay date," January 1, 1994, accounting for the unique timing of each fuel batch. This "pre-decay" feature avoids the complexity of adding decay functions for all radionuclides in the LANL portion of the overall model. In general DKPRO also has the capability to account for the separation (i.e., the partial removal) of various radionuclides during fuel processing operations. In this current application, however, while DKPRO accounts for the total decay of radionuclides in fuel, it does not account for the chemical separation of routinely extracted elements: Tc (partial extraction), Th, U, Np, and Pu. Also, it does not account for the removal (to gaseous effluents, silver reactors, or process condensates to cribs) of volatile elements, including ³H, ¹⁴C, and ¹²⁹I. In effect, the "fuel activity file" output by this version of DKPRO represents the activity of fuels as if they were held from the date of reactor discharge to January 1, 1994, but without being processed through separations.

Thus, while fuel activity records for non-separated radionuclides can be used directly as values input to tanks, further data correction must be done (i.e., in the HDW or SPLIT models) to account for the separated radionuclides mentioned, above. A special correction is also required in the case of ²⁴¹Am to account for the removal of its parent, ²⁴¹Pu, at the fuel separation date. Note that this correction for ²⁴¹Am remaining in tank waste on January 1, 1994, is very sensitive to the fuel cooling time, the fraction of ²⁴¹Pu remaining in waste, and the waste aging period (separation date to January 1, 1994).

Output from the DKPRO code is an electronic file of 1,276 waste activity records of pre-decayed radionuclide values, each record representing the curies associated with a batch of fuel processed by month (or a shorter period) through individual separation plants. Within each record DKPRO generates two sub-records: (1) curies in the fuel cladding and (2) total curies in fuel cores to be fed to extraction operations. The calculation splitting activation products between cladding and fuel cores is based on target element concentrations in fuel cladding and cores. These data are presented in Appendix G of this report as Table G-5. The DKPRO output file also carries data fields giving batch-specific waste loss factors for extractable elements (U, Th, Np, and Pu) for use later in the HDW and SPLIT models.

Format of the DKPRO output file is explained in Watrous and Wootan (1997).

6.1.5.2 Hanford Defined Waste Model. During FY 1996, the HDW model (then under development at LANL) was modified to carry data fields for the 46 key radionuclides in addition to data fields for inert chemicals. Then, as indicated in Figure 6.1-2, a separations partitioning model was, in effect, added to the overall LANL model to account for the partial removal of U, Th, Np, and Pu during fuel separations processing, and to generate "defined waste compositions" (in terms of radionuclides) for various separations process waste streams. Waste stream split factors and separations loss factors for U, Th, Np, and Pu as used in the HDW model are described in Agnew et al. (1997a).

In FY 1996, modifications to the then existing HDW model were made to account for the solubility behavior of radionuclides. Many of the 46 key radionuclides are elements whose solubility was already modelled (e.g., Sr, Cs, U, Pu, and Am). Others of the key radionuclides are elements that are new to the HDW model (e.g., Se, Co, or Ru) and for which only a grossly approximate definition of solubility behavior has been programmed (i.e., the assumption of a 50/50 split of the radionuclide between sludges and supernatants).

The HDW model also accounts for the removal from tank inventory of all soluble radionuclides that were routed to cribs during tank volume recovery operations in the 1950's. Likewise, the loss of tritium (^3H) to evaporator and boiling tank condensate is accounted for in the model as is the separation of Sr-Y and Cs-Ba during waste fractionization operations in B Plant.

Results from the HDW model are given in Agnew et al. (1997a), in terms of tank by tank curie inventories and global inventories (see Appendix E of Agnew et al. 1997a). The global inventory results were also copied into Table 6.1-3 (under the heading DKPRO/HDW model), where they can be compared to other inventory data sources.

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Table 6.1-3. Global Radionuclide Inventory Comparisons.
(Curies remaining in 177 tanks - decayed to January 1, 1994) (2 Sheets)

Nuclides	Calculated by DKPRO/SPLIT model	Calculated by DKPRO/HDW model	From IDB Rev. 12 ^a	From TWRS FEIS ^b	From nuclear material accountability and samples ^c
³ H	3.40 E+04	7.16 E+04			
¹⁴ C	3.28 E+03	4.81 E+03	4.57 E+03	5.34 E+03	
⁵⁹ Ni	1.18 E+03	9.34 E+02			
⁶⁰ Co	1.33 E+04	1.23 E+04			
⁶³ Ni	1.17 E+05	9.20 E+04			
⁷⁹ Se	1.00 E+03	7.73 E+02			
⁹⁰ Sr	7.16 E+07	6.16 E+07	6.25 E+07	6.19 E+07	
⁹⁰ Y	7.16 E+07	6.16 E+07	6.25 E+07	6.19 E+07	
⁹³ Zr	4.74 E+03	3.63 E+03			
^{93m} Nb	3.48 E+03	2.69 E+03			
⁹⁹ Tc	2.47 E+04	3.26 E+04	3.21 E+04	3.21 E+04	
¹⁰⁶ Ru	1.14 E+05	1.04 E+05			
^{113m} Cd	2.24 E+04	1.69 E+04			
¹²⁵ Sb	2.19 E+05	2.08 E+05			
¹²⁶ Sn	1.53 E+03	1.19 E+03			
¹²⁹ I	4.57 E+01	6.30 E+01	2.98 E-01	3.83 E+01	
¹³⁴ Cs	8.89 E+04	8.82 E+04			
¹³⁷ Cs	4.64 E+07	4.71 E+07	3.95 E+07	4.01 E+07	
^{137m} Ba	4.39 E+07	4.46 E+07	3.74 E+07	3.79 E+07	
¹⁵¹ Sm	3.54 E+06	2.75 E+06	1.07 E+06		
¹⁵² Eu	1.89 E+03	1.48 E+03			
¹⁵⁴ Eu	1.74 E+05	1.47 E+05			
¹⁵⁵ Eu	1.60 E+05	1.36 E+05			
²²⁶ Ra	7.72 E-02	6.31 E-02			
²²⁷ Ac	1.08 E+02	8.76 E+01			
²²⁸ Ra	6.44 E+01	7.71 E+01			

Table 6.1-3. Global Radionuclide Inventory Comparisons.
(Curies remaining in 177 tanks - decayed to January 1, 1994) (2 Sheets)

Nuclides	Calculated by DKPRO/SPLIT model	Calculated by DKPRO/HDW model	From IDB Rev. 12 ^a	From TWRS FEIS ^b	From nuclear material accountability and samples ^c
²²⁹ Th	2.92 E+01	1.81 E+00			
²³¹ Pa	1.91 E+02	1.56 E+02			
²³² Th	2.11 E+00	4.36 E+00			
²³² U	1.19 E+02	2.59 E+02			1.23 E+02
²³³ U	4.60 E+02	9.94 E+02			4.76 E+02
²³⁴ U	2.78 E+02	8.50 E+02			3.46 E+02
²³⁵ U	1.17 E+01	3.66 E+01			1.45 E+01
²³⁶ U	7.69 E+00	1.61 E+01			9.57 E+00
²³⁷ Np	1.43 E+02	1.41 E+02			
²³⁸ Pu	2.77 E+03	2.08 E+03			1.45 E+03
²³⁸ U	2.59 E+02	9.06 E+02			3.22 E+02
²³⁹ Pu	3.91 E+04	4.70 E+04	2.64 E+04	2.64 E+04	3.59 E+04
²⁴⁰ Pu	8.93 E+03	9.87 E+03	6.69 E+03	6.70 E+03	8.97 E+03
²⁴¹ Am	8.07 E+04	6.99 E+04	1.04 E+05	1.04 E+05	
²⁴¹ Pu	2.29 E+05	1.67 E+05	1.03 E+05	1.00 E+05	1.47 E+05
²⁴² Pu	1.16 E+00	7.90 E-01	2.80 E-01		1.06 E+00
²⁴² Cm	1.10 E+02	7.70 E+01			
²⁴³ Am	4.19 E+01	9.34 E+00			
²⁴³ Cm	1.37 E+01	1.00 E+01			
²⁴⁴ Cm	3.14 E+02	2.42 E+02			

FEIS = Final Environmental Impact Statement

HDW = Hanford Defined Waste

IDB = Integrated Database

TWRS = Tank Waste Remediation System

^aIDB Data from Rev. 12 (DOE 1996a), Table 2.11, corrected to January 1, 1994

^bTWRS Final Environmental Impact Statement (DOE 1996b), Appendix A, corrected to January 1, 1994

^cAccountability data for period 1944-1971 from (ERDA 1977). Sample Data for 1972-1989 Production, from Tank Characterization Reports (DiCenso and Simpson 1994, Hodgson 1995a and b, and Ryan 1995). (No decay date adjustment due to long half lives.)

6.1.5.3 DKPRO/SPLIT Model. The combination of the DKPRO output file (1,276 fuel activity records) and the SPLIT spreadsheet serves as an alternate method for estimating global radionuclide inventories (but not tank-by-tank inventories). In its use of separations processing split factors for ^{14}C , ^{99}Tc , ^{129}I , the isotopes of U, Th, Np, and Pu, and material balance data for encapsulated ^{90}Sr , ^{137}Cs , recovered U and added Pu (from PFP operations), the DKPRO/SPLIT methodology is independent of HDW model assumptions. This methodology uses an alternate set of separations waste loss factors that are carried on the DKPRO output file. Likewise, an alternate set of separations waste split factors, given in Appendix G, are used to account for separations plant removal of ^{14}C , ^{99}Tc , and ^3H . These split factors are organized in three tables, one for each separation process (BiPO₄ [B and T Plants], REDOX, and PUREX), as shown in Tables G-2, G-3, and G-4. The separations partitioning model uses these defined factors to calculate the partitioning of fuel activity into the several waste streams discharged to tanks from the separations plants.

Like the modified HDW model, the SPLIT spreadsheet also calculates a decay correction for ^{241}Am to account for the timing of ^{241}Pu separation from each fuel batch and its effect on ^{241}Am buildup in the waste.

Unlike the HDW model, the DKPRO/SPLIT model does not account for losses of soluble radionuclides in tank waste leaks or supernate transfers to cribs in the early years. However, these effects are estimated to be minor. Global Inventory results from the DKPRO/SPLIT method are summarized in Table 6.1-3.

6.1.6 Comparison of Global Inventory Model Results

Global Inventory results from the above two alternative models are listed in Table 6.1-3 along with two previously published tank inventory data bases--the Integrated Data Base report, Rev. 12 (DOE 1996a) and the TWRS Final EIS (DOE 1996b).

For uranium and plutonium an additional source of global tank inventory information exists in a combination of accountability records and tank sample data. Table 6.1-4 illustrates the generation of global inventory values for uranium isotopes. Here, accountability data (based on waste stream concentrations measured at the separations plants) are used to represent the waste losses associated with production from 1944 through 1971, while tank sample data (for tanks 241-AZ-101, 241-AZ-102, 241-AW-103, and 241-AW-105) give a truer representation of waste losses associated with post 1971 production (zirconium clad fuel processing operations).

The estimate for plutonium is similar to that for uranium (waste stream measurements for the 1944 through 1971 period plus tank sample analyses for the post 1971 production period). However, the "accountability/sample" value for Pu also accounts for the addition of 142 kg of Pu in waste from Plutonium Finishing Plant (PFP) operations, added in the period 1981 and later.

Table 6.1-4. Calculation of Global Uranium Isotopic Inventory from Accountability Records and Tank Samples.

1 Nuclide	2 Ci/g	3 Curies by DKPRO/ SPLIT ^a	4 Curies (44-71) from ERDA 77-44	5 Curies (72-89) from TCRs	6 Calculated Activity*		8 Total curies ^g	9 Mass U ^h (g)
					Curies (44-71)	Curies (72-89)		
²³² U	21.4	1.1918 E+02	-	-	1.2338 E+02 ^d	-	1.2338 E+02	5.7652 E+00
²³³ U	0.009639	4.5980 E+02	476 ^b	-	476 ^d	-	4.7600 E+02	4.9383 E+04
²³⁴ U	0.006249	2.7832 E+02	-	-	3.2250 E+02 ^e	2.3880 E+01 ^f	3.4638 E+02	5.5430 E+04
²³⁵ U	2.2 E-06	1.1689 E+01	13 ^b	-	1.3544 E+01 ^e	1.0029 E+00 ^f	1.4547 E+01	6.7286 E+06
²³⁶ U	6.5 E-05	7.6861 E+00	-	-	8.9062 E+00 ^e	6.5946 E-01 ^f	9.5656 E+00	1.4785 E+05
²³⁸ U	3.4 E-07	2.5890 E+02	300 ^b	-	3.0000 E+02 ^e	2.2213 E+01 ^f	3.2221 E+02	9.5840 E+08
Total U mass (g):		7.7566 E+08	-	6.655 E+07 ^c				9.6538 E+08

^aUranium isotopic estimate from DKPRO/SPLIT model, Table 6.1-3.

^bAvailable curie data from accountability records for the waste accumulation period (1944 through 1971). Value for ²³³U (400 Ci) from ERDA 77-44 has been corrected to 476 Ci to match original measured waste stream losses of 49.4 kg ²³³U.

^cTank sample data for 1972 to 1989 production period is summed from Tank Characterization Reports (TCR) (DiCenso and Simpson 1994, Hodgson 1995a and b, and Ryan 1995).

^dCurie value for ²³²U is estimated from ²³³U (Column 4), in ratio to Column 3 curies.

^eCurie values for ²³⁴U, ²³⁵U, and ²³⁶U are estimated from ²³⁸U (Column 4), in ratio to Column 3 curies.

^fCurie values for ²³⁴U, ²³⁵U, ²³⁶U, and ²³⁸U are estimated from U mass (Column 5), in ratio to Column 3 curies. Specific activity constants from Column 2 are first used to calculate the equivalent mass for Column 3 curies (7.7566 E+08 g U).

Example for ²³⁴U:

$$2.388 \text{ E}+01 \text{ Ci} = (2.7832 \text{ E}+02 \text{ Ci}) \cdot (6.655 \text{ E}+07 \text{ g U}) / (7.7566 \text{ E}+08 \text{ g U})$$

^gTotal curies are the sum of Columns 6 and 7.

^hIsotopic mass and total mass are converted from Column 8, using specific activity constants from Column 2.

*Values filled by Ratio to DKPRO Curies in column 3.

The Table 6.1-3 comparison of DKPRO/HDW model results to DKPRO/SPLIT model results shows minor discrepancies for most radionuclides (differences of about 20 percent or less) and somewhat larger discrepancies for those elements that have experienced extraction or volatilization during fuel separations operations or waste fractionation processing. This model-to-model comparison is also illustrated by Figure 6.1-3, which shows an X-Y scatter plot of DKPRO/HDW versus DKPRO/SPLIT model results. Here, it can be seen that (relative to the DKPRO/SPLIT model) the DKPRO/HDW model predicts significantly higher values for ^3H , ^{129}I , ^{232}Th , and all U isotopes. These differences are largely a result of the DKPRO/HDW model's not accounting for volatilization losses or using conservatively high waste loss factors. For example, in the case of ^{233}U , detailed accountability records for the 1966 and 1970 Thoria fuel processing campaigns (Isochem 1967, Jackson and Walser 1977) indicate that a global total of 49.4 kg of ^{233}U (476 Ci) was lost to waste tanks. This value compares well with ^{233}U results of the DKPRO/SPLIT model (460 Ci). In contrast, the DKPRO/HDW global estimate for ^{233}U (994 Ci) exceeds the accountability value by a factor of 2.1.

Comparison of both model results to previous inventories (i.e., to IDB and TWRS FEIS reports) shows close agreement between the DKPRO/HDW result for ^{90}Sr and previous inventories, whereas the DKPRO/SPLIT model predicts 15 percent more. Comparison for ^{137}Cs shows that DKPRO/HDW results agree closely with DKPRO/SPLIT results and both exceed previous inventories by about 13 percent to 17 percent.

For most radionuclides the discrepancies between both models and previous inventory lists are so minor (generally less than 20 percent) as to make it difficult to judge which model may be closer to reality. However, the relatively larger discrepancies for tritium, ^{129}I , Th, U, and Pu isotopes invite a closer analysis.

- Tritium values estimated by the DKPRO/SPLIT model attempt to account for losses of ^3H to separations process condensates and atmospheric emissions, whereas the DKPRO/HDW model assumes that 100 percent of ^3H in fuel is routed to tank waste. Thus, it is reasonable that DKPRO/SPLIT tritium values should be lower (and a more realistic prediction) than DKPRO/HDW results.
- The ^{99}Tc inventory value from the DKPRO/SPLIT model is 76 percent of the value predicted by the DKPRO/HDW model. This difference occurs because the DKPRO/SPLIT model accounts for an estimated 24 percent of technetium being extracted and routed to the uranium product in PUREX operations and for 37 percent being extracted in REDOX operations. The DKPRO/HDW model, on the other hand, assumes no loss of Tc during fuel separations processing. Due to the uncertainties in the Tc extraction factors used in the DKPRO/SPLIT model, the ^{99}Tc inventory predicted by the DKPRO/HDW model (32,600 curies) is judged to be the more reliable and conservative bounding value.
- Iodine values estimated by the DKPRO/SPLIT model attempt to account for losses of ^{129}I to atmospheric emissions, solid waste burials (spent silver reactors) and silver reactor flushes that were routed to cribs. The model assumes split factors for iodine (fraction routed to tank waste) of 0.4, 0.4, 0.4, and 0.8 for T, B, REDOX, and PUREX plants, respectively. In effect an overall split factor of 0.71 results for

the combined four plant throughput. In contrast, the DKPRO/HDW model makes the more conservative and simplifying assumption that essentially all of the ^{129}I in reactor fuel was routed to waste tanks (a split factor of 1.0). Due to the uncertainties in factors used in the DKPRO/SPLIT model, the ^{129}I inventory predicted by the DKPRO/HDW model (63 curies) is judged to be the more reliable and conservative bounding value.

- Although the DKPRO/HDW model's prediction for ^{232}Th at first appears to be the higher and more conservative value, the DKPRO/SPLIT model's value of 2.11 curies (roughly equivalent to 25 metric tons elemental thorium) is judged to be the more realistic, being based on accountability measurements made during the two thorium production campaigns in 1966 and 1970.
- Comparison of model results for uranium isotopes (from Table 6.1-3) to accountability/sample data (from Table 6.1-4) shows an interesting spread of values: In comparison to the accountability/sample curie values for ^{238}U (equivalent to a global inventory of 960 MTU), the DKPRO/SPLIT model predicts a value that is slightly lower (80 percent), while the DKPRO/HDW model predicts a value that is significantly higher by a factor of 2.8. The HDW result for ^{238}U , being a factor of 2.8 greater than previous estimates, has no basis in sample evidence to suggest that a correction of this magnitude could be real. (The apparent overprediction of the HDW model is thought to be due to the model's use of a conservative factor for fraction of metal waste not recovered.)
- For the plutonium isotopes, ^{239}Pu , ^{240}Pu and ^{242}Pu , DKPRO/SPLIT model results and accountability/sample derived values compare rather closely (a difference of 9 percent or less). This remarkably good agreement tends to support the validity of DKPRO/SPLIT model results for major Pu isotopes, and implies an equivalent Pu mass inventory range of 620 to 670 kg. DKPRO/HDW model results, being 20 to 30 percent higher, are likely less accurate due to relatively simplistic assumptions used in modelling. [Note that both DKPRO/SPLIT and accountability/sample inventory results include an estimated contribution from Plutonium Finishing Plant (PFP) waste that is equivalent to 142 kg of Pu.]

For the shorter half-lived Pu isotopes, ^{238}Pu and ^{241}Pu , the ratio comparison of DKPRO/SPLIT results over accountability/sample derived values is not as good, being factors of 1.9 and 1.6, respectively. Based on the understanding that the reported accountability values for ^{238}Pu were not actually measured but were calculated by ratio from earlier fuel activity codes, it seems reasonable to conclude that DKPRO/SPLIT values are likely the more accurate in regard to these minor isotopes. Therefore, DKPRO/SPLIT values are judged to be the best basis for a global Pu inventory.

6.1.7 Validation of Model Results from Tank Heat Measurements

Accuracy of DKPRO/HDW model predictions for total (global) ^{90}Sr and ^{137}Cs was tested by comparison of calculated equivalent decay heat to a combination of measured and independently estimated decay heat values for all tanks. "Measured" decay heat for 162 tanks (derived from a previous 1995 study that analyzed tank temperature data) (Kummerer 1995) was combined with engineering estimates for 15 other tanks (not measured in the 1995 study) to give a total tank decay heat value of 690 kilowatts (as of ca. 1994). Heat balance calculation results are summarized in Table 6.1-5.

Table 6.1-5. Comparison of Measured Tank Decay Heat to Model Predicted Heat.

Calculation of measured/estimated tank decay heat (as of ca. 1994)	
Measured Decay Heat in 162 Tanks (Watts) ^a	5.05 E+05
Calculated Decay Heat in AZ-101,2 (Watts) ^b	1.26 E+05
Estimated Decay Heat in 6 unmeasured DSTs (Watts) ^c	3.96 E+04
Estimated Decay Heat in 7 unmeasured SSTs (Watts) ^d	1.60 E+04
Total Decay Heat in 177 Tanks (Watts)	6.87 E+05
Model based tank decay heat (as of 1/1/1994)	
Cs/Sr Decay Heat Predicted from DKPRO/SPLIT Model (Watts)	6.87 E+05
Cs/Sr Decay Heat Predicted from DKPRO/HDW Model (Watts)	6.35 E+05

^aData from (Kummerer 1995).

^bBased on ^{137}Cs and ^{90}Sr predicted by the DKPRO/SPLIT model for 241-AZ-101, 241-AZ-102 tanks (PUREX throughput, 1983-1989).

^cEstimated by ratio to measured heat in 15 tanks in AN, AP, and AW farms. (Data from Kummerer 1995)

^dEstimated by ratio to measured heat in 33 tanks in TX, SX, and U farms. (Data from Kummerer 1995).

In comparison to this measured/estimated decay heat total of 687 kilowatts, the DKPRO/HDW model predicts a total decay heat of 635 kilowatts--a difference of only 8 percent, while the DKPRO/SPLIT model predicts a total decay heat of 687 kilowatts. (In this latter comparison, the perfect agreement is likely coincidental and should probably not be viewed as evidence that the DKPRO/SPLIT model is more valid than the DKPRO/HDW model.) This close comparison provides additional confirmation to the accuracy of the HDW model (for global values of non-extracted radionuclides). The close agreement also tends to validate precursor codes and databases such as the ORIGEN2 code (as set to model fuel activity for Hanford's single-pass reactors and N Reactor) and our historical Separations Production data file.

6.1.8 Selection of Model-Based Global Radionuclide Inventory

From the comparisons made in sections 6.1.6 and 6.1.7, above, it is judged that DKPRO/HDW model global results are sufficiently accurate to serve as the global inventory values for the majority of radionuclides (i.e., non-extracted elements). However, for extractable or volatile radionuclides (^3H , ^{90}Sr , and its daughter, isotopes of Cs and their daughters, ^{232}Th , and isotopes of Pu) it is judged that results of the DKPRO/SPLIT model are generally a more realistic estimate of global tank inventories. For isotopes of uranium it is judged that the accountability/sample inventory as generated in Table 6.1-4 should be considered more valid than predictions of the DKPRO/SPLIT model and more correct than the unsupported high loss predictions of the DKPRO/HDW model.

Accordingly, the global radionuclide inventory given in Table 6.1-6 has been composed from DKPRO/SPLIT results (for ^3H , ^{90}Sr , ^{90}Y , ^{134}Cs , ^{137}Cs , $^{137\text{m}}\text{Ba}$, ^{232}Th , and all Pu isotopes), accountability/sample results (for isotopes of U) plus DKPRO/HDW model results (for all other key radionuclides).

Table 6.1-6. Global Radionuclide Inventory. (2 Sheets)

Nuclide	Curies remaining in 177 tanks	Data source
	(Decayed to January 1, 1994)	(From DKPRO/HDW model unless stated otherwise)
^3H	3.40 E+04	Based on DKPRO/SPLIT model
^{14}C	4.81 E+03	
^{59}Ni	9.34 E+02	
^{60}Co	1.23 E+04	
^{63}Ni	9.20 E+04	
^{79}Se	7.73 E+02	
^{90}Sr	7.16 E+07	Based on DKPRO/SPLIT model
^{90}Y	7.16 E+07	Based on DKPRO/SPLIT model
^{93}Zr	3.63 E+03	
$^{93\text{m}}\text{Nb}$	2.69 E+03	
^{99}Tc	3.26 E+04	
^{106}Ru	1.04 E+05	
$^{113\text{m}}\text{Cd}$	1.69 E+04	
^{125}Sb	2.08 E+05	
^{126}Sn	1.19 E+03	
^{129}I	6.30 E+01	
^{134}Cs	8.89 E+04	Based on DKPRO/SPLIT model
^{137}Cs	4.64 E+07	Based on DKPRO/SPLIT model
$^{137\text{m}}\text{Ba}$	4.39 E+07	Based on DKPRO/SPLIT model
^{151}Sm	2.75 E+06	
^{152}Eu	1.48 E+03	

Table 6.1-6. Global Radionuclide Inventory. (2 Sheets)

Nuclide	Curies remaining in 177 tanks	Data source
	(Decayed to January 1, 1994)	(From DKPRO/HDW model unless stated otherwise)
²²⁶ Ra	6.31 E-02	
²²⁷ Ac	8.76 E+01	
²²⁸ Ra	7.71 E+01	
²²⁹ Th	1.81 E+00	
²³¹ Pa	1.56 E+02	
²³² Th	2.11 E+00	Based on DKPRO Waste Loss Factor.
²³² U	1.23 E+02	Based on Accountability/Sample Data.
²³³ U	4.76 E+02	Based on Accountability/Sample Data.
²³⁴ U	3.46 E+02	Based on Accountability/Sample Data.
²³⁵ U	1.45 E+01	Based on Accountability/Sample Data.
²³⁶ U	9.57 E+00	Based on Accountability/Sample Data.
²³⁸ U	3.22 E+02 (or 958 MT as defined in Figure 6.3-1)	Based on Accountability/Sample Data.
²³⁷ Np	1.41 E+02	
²³⁸ Pu	2.77 E+03	Based on DKPRO/SPLIT Model.
²³⁹ Pu	3.91 E+04	Based on DKPRO/SPLIT Model.
²⁴⁰ Pu	8.93 E+03	Based on DKPRO/SPLIT Model.
²⁴¹ Pu	2.29 E+05	Based on DKPRO/SPLIT Model.
²⁴² Pu	1.16 E+00	Based on DKPRO/SPLIT Model.
²⁴¹ Am	6.99 E+04	
²⁴² Cm	7.70 E+01	
²⁴³ Am	9.34 E+00	
²⁴³ Cm	1.00 E+01	
²⁴⁴ Cm	2.42 E+02	

HDW = Hanford Defined Waste.

Note that for ³H, ¹²⁹I, Th, and U isotopes, global values as estimated in Table 6.1-6 may not necessarily match the sum of the tank-by-tank values given in Agnew et al. (1997a), Appendix E or the sum of best-basis values for all 177 tanks. These expected discrepancies are to be reconciled in future work.

6.1.9 Summary of Errata and Accuracy Issues for Table 6.1-6

Comparisons made in the sections, above, show that best-basis global inventory results (as given in Table 6.1-6) are reasonably accurate for major radionuclides such as ⁹⁰Sr, ¹³⁷Cs, ²³³U, and plutonium--those radionuclides that can be compared to some independent measurement. The basic assumption being made in this methodology is that if the ORIGEN2

code and historical Separations Production data file can, together, accurately predict inventory values for certain major radionuclides, then values for all other (unmeasured) radionuclides can be predicted with equal accuracy. Although this assumption is generally true, there are a few radionuclides (activation products and extractable elements) for which model predictions will be more uncertain.

In the current version of the DKPRO/HDW model, several simplifying assumptions have been made that affect the accuracy of global and tank-by-tank inventory estimates for the following radionuclides:

- Losses of Tritium to process condensate and to atmosphere at the four separations plants can vary markedly, depending on the degree of waste stream evaporation/concentration. The global inventory value given on Table 6.1-6 is based on tritium split factors of 1.0, 1.0, 0.78 and 0.014 for T, B, REDOX, and PUREX plants, respectively (fraction of fuel activity routed to tank waste) as derived from process flowsheets. The reader needs to recognize possible inaccuracies in the tritium inventory associated with the use of these assumed split factors.
- ORIGEN2 code values for ^{14}C (Ci/MTU) are based on an assumed constant level of nitrogen impurity in uranium fuel cores of 20 ppm. In reality, the nitrogen level varied with time, ranging from highs of 90 ppm in the late 1940's to lows of about 8 ppm during the 1970's. A second simplifying assumption is made regarding losses of ^{14}C (to atmosphere) during fuel dissolution: it is assumed that 100 percent of ^{14}C in fuel is routed to waste tanks. (In actuality, 30 to 40 percent of the ^{14}C was measured leaving the plant in the dissolver offgas during PUREX campaigns from 1983 through 1987.) For lack of any historical operating data, the same 100 percent assumption was also applied to fuel processed through T, B, and REDOX separations plants.

Consequently, global inventories for ^{14}C given in Table 6.1-6 may be conservatively high.

- Activation products, ^{59}Ni and ^{63}Ni , are based on an assumed constant 800 ppm of Ni in aluminum cladding--an average value for all aluminum clad fuel types. (Beginning in 1959, fuel cladding (aluminum alloys) contained nickel levels of 9,000 to 13,000 ppm.) Thus, while HDW model results for global $^{59}\text{Ni}/^{63}\text{Ni}$ may be fairly representative of our total tank inventory, individual tank model results may be significantly distorted, being high in tanks filled before 1959 and low in tanks filled after 1959.
- The ORIGEN2 estimates for activation product, ^{60}Co , assume a cobalt impurity level of 10 ppm in cladding and 0 ppm in core uranium. Since actual levels of cobalt in uranium are unknown but probably greater than zero, it is predictable that the DKPRO output results for ^{60}Co are low.
- The ORIGEN2 code's prediction of ^{237}Np is based on an assumed constant level of

400 ppm ^{236}U in initial 0.947 percent enriched uranium input to the Hanford reactors. In actuality, the ^{236}U level varied from nearly zero to over 800 ppm in enriched uranium irradiated during the 1980's. This buildup of ^{236}U over time was the result of a closed fuel cycle, which in effect exposed the same uranium to multiple irradiation cycles. The ^{237}Np production can be expected to vary somewhat with the ^{236}U level, but a direct proportionality is not expected (due to the multiple neutron capture paths involved in ^{237}Np production). In addition, the efficiency of Np recovery in the REDOX and PUREX processes has been variable over the years and incompletely documented. Consequently, the global and tank-by-tank estimates for ^{237}Np contain a higher degree of uncertainty than do most other radionuclides.

One problem internal to the ORIGEN2 code is now known which affects the accuracy of curie values listed here for ^{79}Se :

- Curie values for ^{79}Se calculated by ORIGEN2 are suspected of being high by a factor of about 8 due to the use of an erroneous half-life value in the ORIGEN2 data library.

6.1.10 Tank-By-Tank Accuracy Issues

Radionuclide inventory values for individual tanks are predicted by the DKPRO/HDW model. Section 6.1.9, above, discusses some of the accuracy issues that are associated with the use of the DKPRO/HDW model for estimating global radionuclide inventory values. When using the DKPRO/HDW model for individual tank inventory values, another set of model assumptions, the solubility factors, come into play to add additional uncertainty to the inventory results. The degree of error between model results and actual tank contents can only be evaluated by comparison to actual core sample results.

To illustrate the degree of model error for individual tanks, scatter plots have been prepared in which available sample-based tank inventory values (for selected radionuclides) are plotted versus DKPRO/HDW model results for the same set of tanks. Inventory values for an additional set of approximately 20 tanks (derived from engineering estimate methods) are also compared on the same plots. Each plot also contains a 1:1 correspondence line on which the scatter points should lie if there is no error in the model and no error in the measured value.

Comparisons for ^{90}Sr , ^{137}Cs , ^{239}Pu , and ^{241}Am are shown in Figures 6.1-4 through 6.1-7. Plots for ^{90}Sr and ^{137}Cs (Figures 6.1-4, and 6.1-5) are similar in that DKPRO/HDW model values greater than about 100,000 Ci per tank show a moderate correlation with measured or estimated values. The somewhat even distribution of points on either side of the 1:1 correspondence line also indicates that there is no major bias in either the model or measured values. However, for tanks containing less than about 100,000 Ci the scatter becomes much worse, indicating relative errors of several orders of magnitude are possible.

The comparison for ^{239}Pu inventory values (Figure 6.1-6) indicates a similarly weak to moderate correlation where errors of up to a factor of 40 may generally be possible with rare tanks being in error by several orders of magnitude.

The comparison for ^{241}Am (Figure 6.1-7) reveals a weak correlation and a strong indication that the analytical procedure for ^{241}Am may be biased high. It is unfortunate that model or sample accuracy for an important alpha emitter such as ^{241}Am cannot be characterized better than that indicated by Figure 6.1-7. Future work should be aimed at better understanding any shortcomings in the ^{241}Am analytical methods being used.

Scatter plot comparisons for ^{60}Co , ^{99}Tc , and ^{155}Eu have been analyzed but are not shown for reason of too few data points. The comparison for ^{60}Co , however, does show a strong indication that the DKPRO/HDW model is predicting low. This situation may be due to the fact that ORIGEN2 model runs lacked realistic setup values for Co impurities present in Hanford uranium fuel cores.

Figure 6.1-4. Model/Sample/Estimate Correlation for Strontium-90.

Sr-90 -- Curies per Tank @ 1/1/94

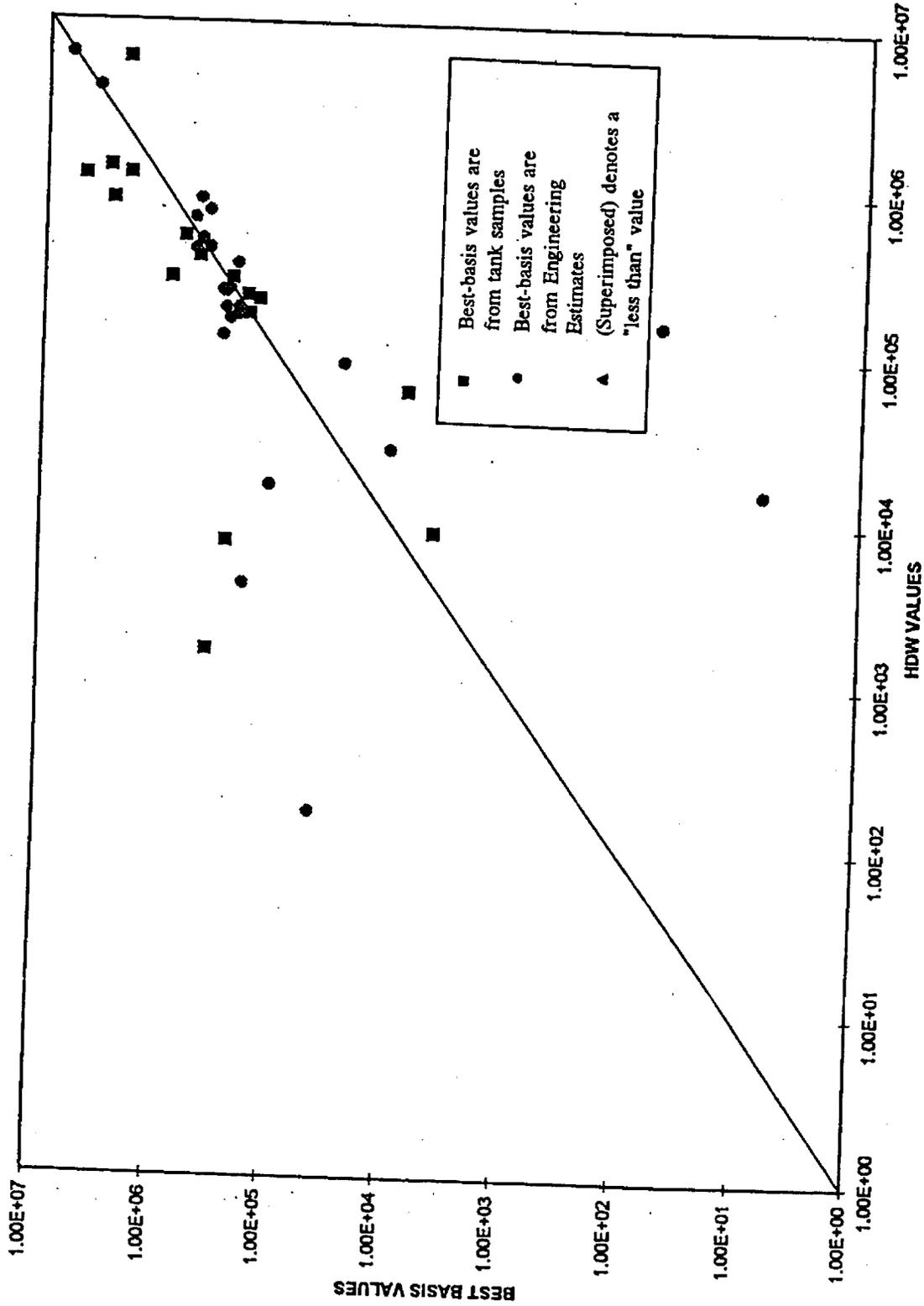


Figure 6.1-5. Model/Sample/Estimate Correlation for Cesium-137.

Cs-137 -- Curies per Tank @ 1/1/94

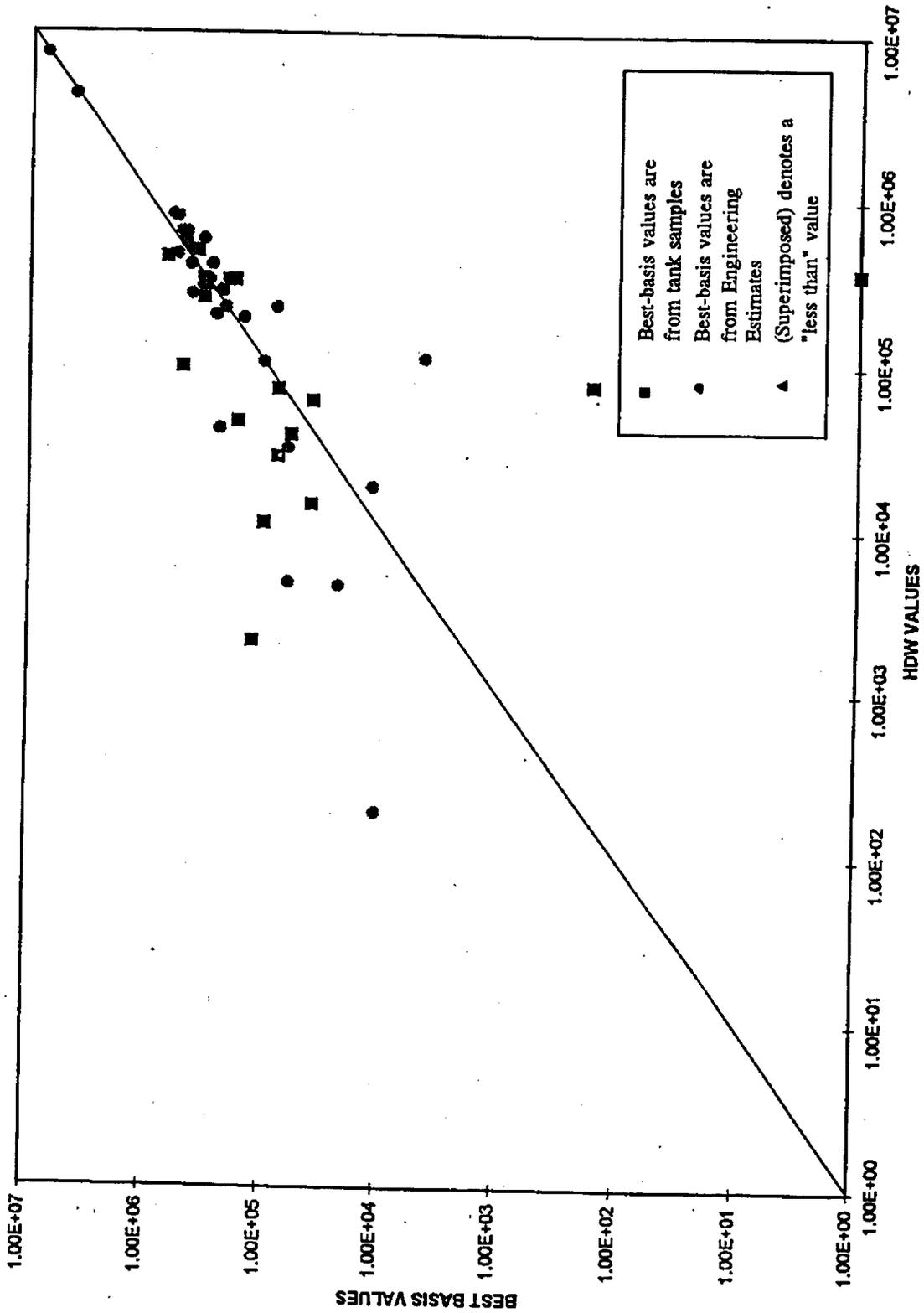


Figure 6.1-6. Model/Sample/Estimate Correlation for Plutonium-239.

Pu-239 -- Curies per Tank @ 1/1/94

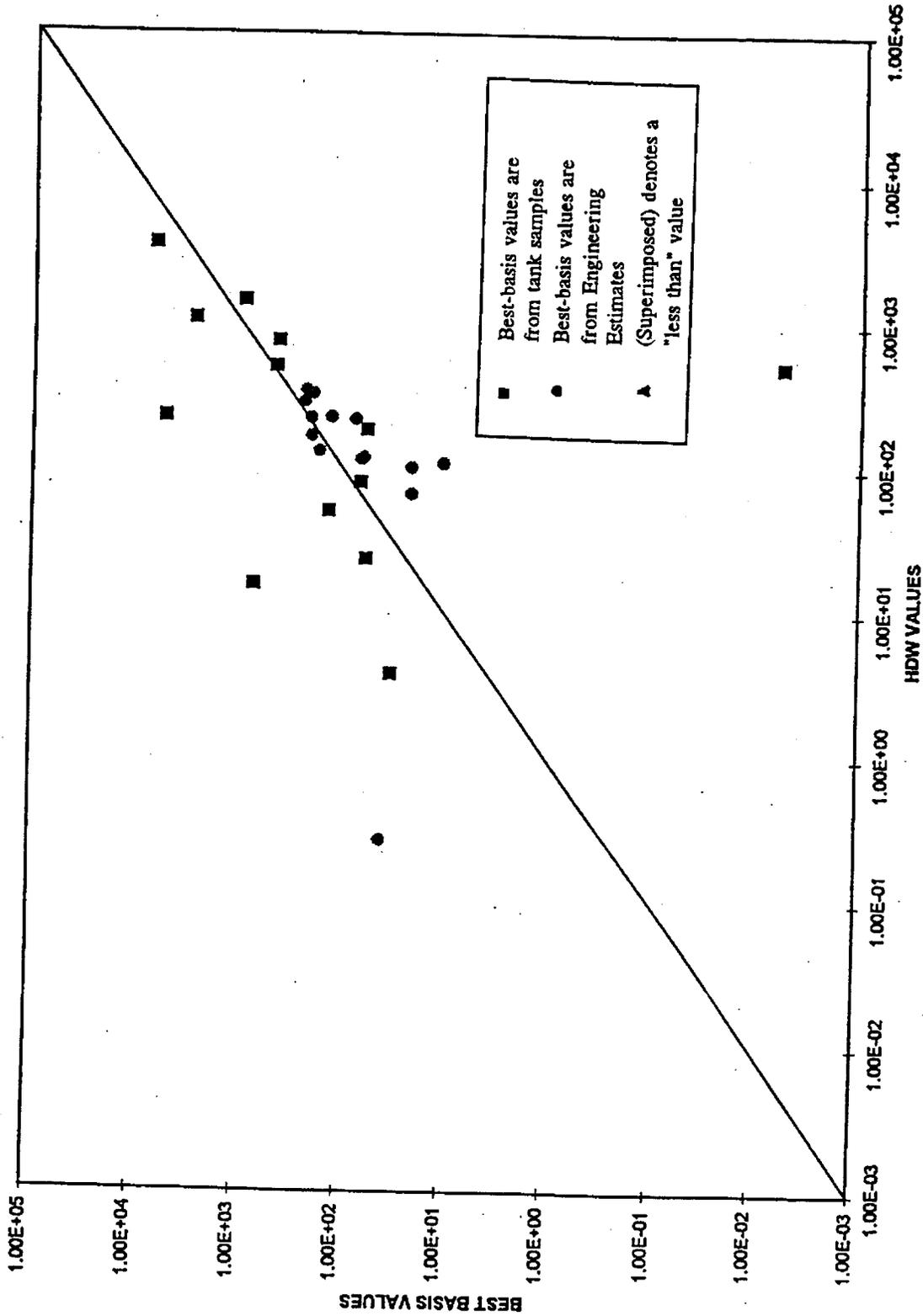
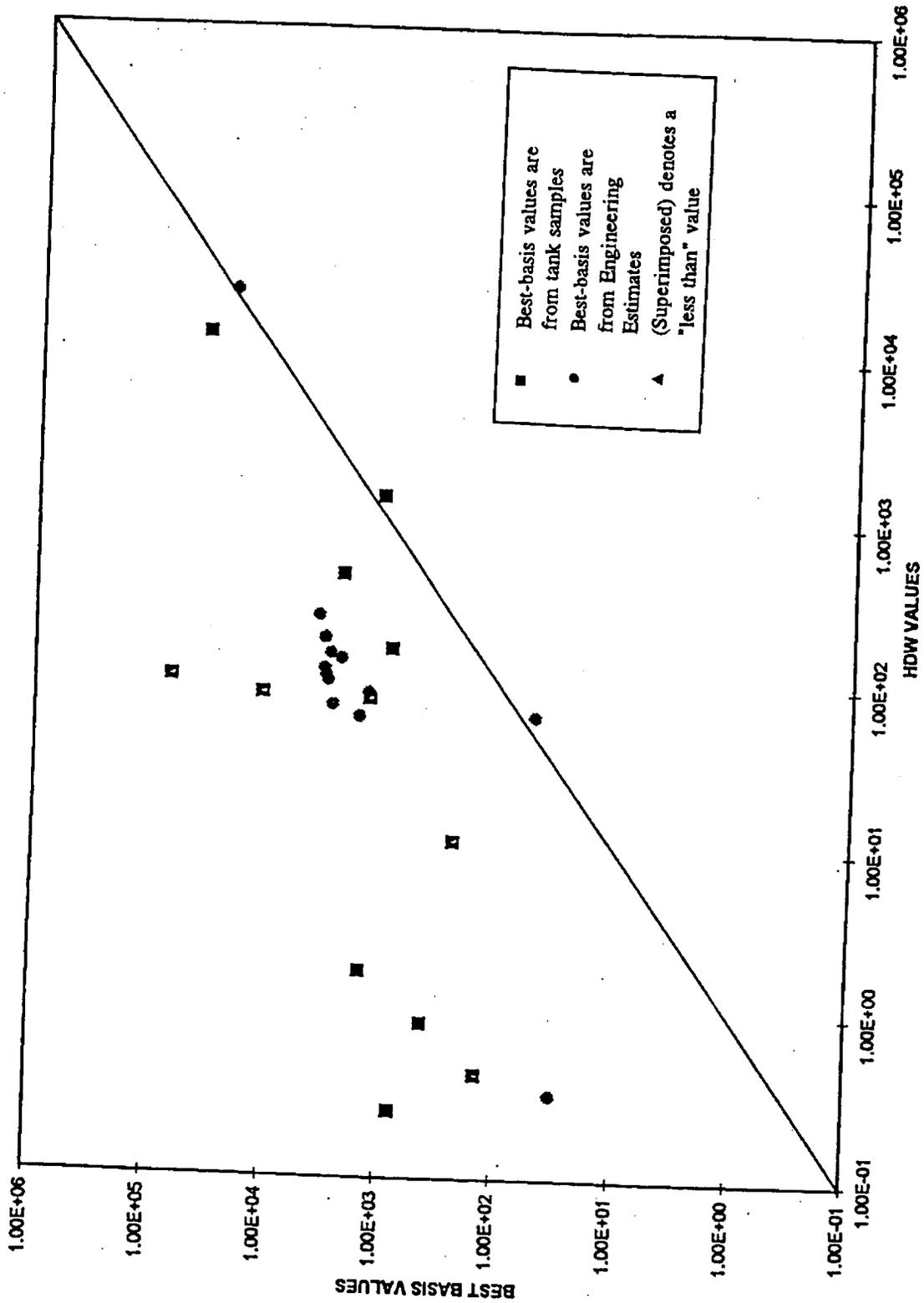


Figure 6.1-7. Model/Sample/Estimate Correlation for Americium-241.

Am-241 - Curies per Tank @ 1/1/94



6.2 CESIUM AND STRONTIUM

The two radionuclides, ^{137}Cs and ^{90}Sr , along with certain others, e.g., ^{99}Tc , ^{79}Se , and the actinide elements, are the most important radioactive constituents of Hanford Site tank wastes.

Because of their approximate 30-year half lives, ^{137}Cs and ^{90}Sr contribute not only almost all the decay heat associated with the tank wastes, but also most of the radioactivity (curie) content of the tank wastes. Systems for disposing of retrieved Hanford Site tank wastes all involve removal of the bulk of the ^{137}Cs in such wastes. Removal of ^{90}Sr from some retrieved wastes may also be necessary or desirable to obtain LLW that can be disposed of in near-surface facilities. For economic reasons, i.e., costs involved in designing, constructing, and operating a ^{137}Cs removal facility, it is particularly important to establish a high-quality best estimate of the global inventory of ^{137}Cs in the 177 underground tanks at the Hanford Site.

The principal objective of the analyses and evaluations reported here is to estimate the global inventory of ^{137}Cs and ^{90}Sr currently in Hanford Site tanks. However, information was also obtained relating to the range of inventories of ^{137}Cs and ^{90}Sr that might eventually be transferred to the tanks as the result of future decontamination and decommissioning activities in the Hanford Site B Plant.

6.2.1 Identification and Comparison of Cesium-137 and Strontium-90 Inventory Values

Currently reported global inventories of ^{137}Cs and ^{90}Sr (from two data sources) in Hanford Site tanks are listed in Table 6.2-1. For comparison purposes, it is convenient to consider separately the amounts of these two radionuclides in DSTs and SSTs. A significant amount of analytical data for wastes in all the individual DSTs allows a reasonable estimate of the global inventory of ^{137}Cs and ^{90}Sr in the DSTs. Preliminary comparisons indicate that the DST global inventories of ^{137}Cs and ^{90}Sr established from analytical data differ by about 25 percent from inventories predicted by the HDW (Rev. 3) model. However, the SST inventory of ^{137}Cs predicted by the HDW (Rev. 3) model is over twice that reported in the present TWRS inventory. Also, the HDW model predicts the ^{90}Sr inventory of the SSTs to be about 25 percent less than that shown in the TWRS inventory. The following subsections describe new material balance calculations to establish SST inventories of ^{137}Cs and ^{90}Sr . Best-basis estimates of the global inventories of ^{137}Cs and ^{90}Sr are provided in Section 6.2.3.

Table 6.2-1. Currently Reported Inventory Values for Cesium-137 and Strontium-90 in Hanford Site Tanks.

Tanks	¹³⁷ Cs, MCi ^a		⁹⁰ Sr, MCi ^a	
	TWRS data	HDW (Rev. 3) model data ^b	TWRS data	HDW (Rev. 3) model data ^b
Double-shell tanks	25.4 ^c	20.2	11.4 ^c	16.3
Single-shell tanks	9.5 ^d	20.5	41.2 ^d	33.4
All	34.9	40.7	52.6	49.7

HDW = Hanford Defined Waste

TWRS = Tank Waste Remediation System

^aDecayed to January 1, 2000.

^bAgnew (1995), Agnew et al. (1996).

^cShelton (1995a) describes the sources and bases for the TWRS data for double-shell tanks and single-shell tanks.

^dShelton (1995a and b), Boldt et al. (1996).

6.2.2 Cesium-137 and Strontium-90 Inventories Evaluation

Historically, not all the ¹³⁷Cs and ⁹⁰Sr originally introduced into the Hanford Site tanks remained there. A large fraction of both radionuclides was removed from 1968 to 1978 and eventually encapsulated as ¹³⁷CsCl and ⁹⁰SrF₂, respectively. Other fractions were discharged to soil columns, sent to other DOE (then Atomic Energy Commission) facilities, or disposed of as solid waste. Still other fractions were removed from the underground tanks and then later reintroduced into the tanks. For all these material transactions, there is some degree of uncertainty concerning the amounts of ¹³⁷Cs and ⁹⁰Sr transferred. Propagation of these uncertainties complicates estimation of the current tank inventories of ¹³⁷Cs and ⁹⁰Sr.

6.2.2.1 Material Balance Considerations. One method for estimating the inventory of ¹³⁷Cs and ⁹⁰Sr in the SSTs is through material balance calculations. Such calculations thus allow estimation of the global inventory of radiostromtium and radiocesium in all the tanks. Two key assumptions underlie the material balance approach to estimating the ¹³⁷Cs and ⁹⁰Sr content of the SSTs.

- The total amounts of ¹³⁷Cs and ⁹⁰Sr generated in Hanford Site reactors are known from computer code calculations.
- The ¹³⁷Cs and ⁹⁰Sr contents of the DSTs are known from analytical data.

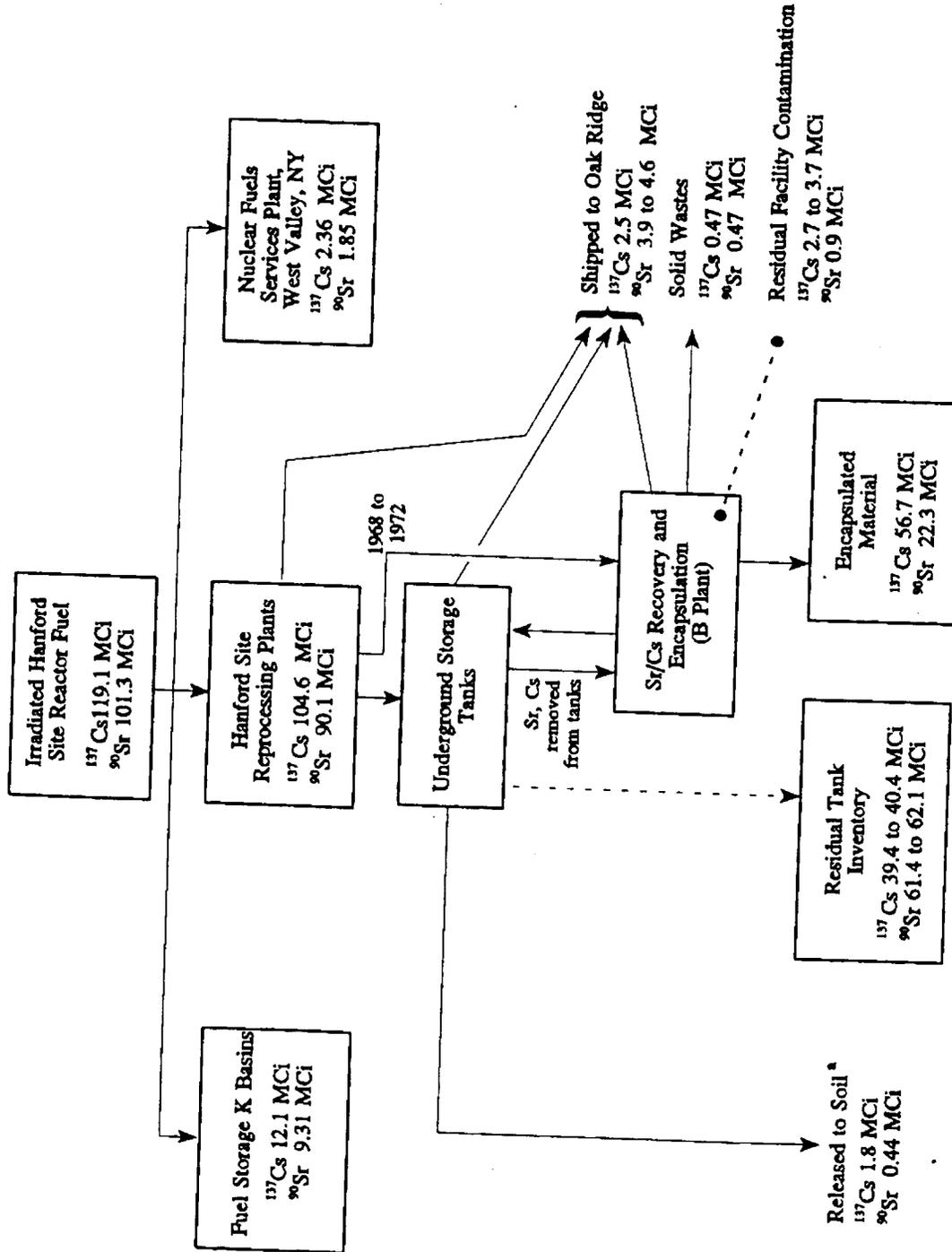
Figure 6.2-1 illustrates the various components of the Hanford Site ^{137}Cs and ^{90}Sr material balances. Table 6.2-2 summarizes best available values for certain of the material balance components cited in Figure 6.2-1. The accompanying Tables 6.2-3, 6.2-4, and 6.2-5 (and text) support information shown in Table 6.2-2 and also indicate sources of data. The validity and utility of global SST inventories of ^{137}Cs and ^{90}Sr derived from the material balance calculations presented in Table 6.2-2 are examined in a later section.

Estimates for the amounts of ^{137}Cs and ^{90}Sr in all fuel processed through the BiPO_4 , REDOX, and PUREX Plants from 1944 through 1989 have recently been generated by the DKPRO code (Schmittroth 1996), and alternatively by the RADNUC code in combination with the RIBD code. Schmittroth's results were obtained using the newly-developed computer code DKPRO (Version 1) which uses Ci/MTU values from the ORIGEN2 code. Outputs of the RIBD and RADNUC codes were combined to calculate the amounts of ^{137}Cs and ^{90}Sr in fuel reprocessed in the Hanford Site separations plants. The RIBD code is preferred to calculate production of ^{137}Cs and ^{90}Sr from 1944 to 1971 while the RADNUC code is preferred for the best estimate of the amounts of ^{137}Cs and ^{90}Sr generated from 1972 through 1989. Watrous and Wootan (1997) data are generated from a later version of the DKPRO code using data for single pass reactors specifically modelled by ORIGEN2. Results (activity in all fuel decayed to January 1, 2000) obtained by these codes are as follows:

^{137}Cs :	DKPRO (Version 1)	= 103.5 MCi
	RIBD/RADNUC	= 102 MCi
	DKPRO (1997)	= 104.6 MCi
^{90}Sr :	DKPRO (Version 1)	= 90.3 MCi
	RIBD/RADNUC	= 93.2 MCi
	DKPRO (1997)	= 90.1 MCi

These three sets of data agree within 3 percent. Agnew et al. (1996) notes that, from the slight differences in thermal fission yields and in half-lives, the inventory of ^{90}Sr after some 35 to 40 years of decay would be expected to be about 15 percent less than the inventory of ^{137}Cs . In the case of the DKPRO-1997 results, the ^{90}Sr inventory is 86 percent of the ^{137}Cs inventory. DKPRO-1997 results are selected for use in Figure 6.2-1 and Table 6.2-2. Data shown in Table 6.2-3 are used to compute the total quantities of ^{137}Cs and Sr^{90} listed in Figure 6.2-1 and Table 6.2-2.

Figure 6.2-1. Estimated Disposition of Cesium-137 and Strontium-90 Generated In Hanford Site Reactors.



All values are decayed to January 1, 2000

* Includes amounts disposed of as solid waste and liquid waste as tank leaks and liquids sent to cribs and trenches.

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Previously, Agnew et al. (1996) estimated that 94 MCi of ^{137}Cs and 78.4 MCi of ^{90}Sr (decayed to January 1, 2000) were introduced into the Hanford Site tanks. Peterson (1996) recently reported that 97 MCi of ^{137}Cs and 82 MCi of ^{90}Sr were sent to the underground tanks. The DKPRO-1997 estimate of 90.1 MCi of ^{90}Sr is thus 8.1 to 11.7 MCi higher than the previous estimates. The DKPRO-1997 estimate of 104.6 MCi of ^{137}Cs is 7.6 to 10.6 MCi higher than the previous estimates. Because they are based on more accurate estimates of fuel irradiation history, ^{137}Cs and ^{90}Sr inventories derived by the DKPRO-1997 code are considered more reliable than prior computer code calculations.

Note that in the table (above), in Figure 6.2-1, and in all subsequent tables of this Section (6.2), curie values for ^{90}Sr and ^{137}Cs are based on a decay date of January 1, 2000, which is different from the decay date basis (January 1, 1994) assumed in Tables 6.1-3 and 6.1-6. The January 1, 2000, decay date was chosen for Section 6.2 for ease of comparison to many existing data sources.

Table 6.2-2. Simplified Hanford Site Underground Tank Cesium-137 and Strontium-90 Material Balance.

Material balance component	MCi ^a	
	^{137}Cs	^{90}Sr
Generated in Hanford Site reactors	119.1	101.3
In fuel not reprocessed at the Hanford Site ^b	14.5	11.2
In fuel input to Hanford reprocessing plants	104.6	90.1
Output to capsules ^d	56.7	22.3
Output to other DOE facilities ^d	2.5	3.9 to 4.6
Output to solid waste disposal ^d	0.47	0.47
Output to facility contamination ^d	2.7 to 3.7	0.9
Output in releases to soil ^e	1.8	0.44
Total tank inventory remaining ^f	39.4 to 40.4	61.4 to 62.1
Inventory in double-shell tanks ^e	25.4	11.4
Inventory in single-shell tanks ^f	14.0 to 15.0	50.0 to 50.7

^aAs of January 1, 2000

^bSee Table 6.2-3

^cSee Table 6.2-4

^dSee Table 6.2-5

^eSee Table 6.2-1

^fCalculated value from material balance analysis.

6.2.2.2 Cesium-137 and Strontium-90 Not Reprocessed at the Hanford Site. Not all the fuel irradiated in Hanford Site reactors was reprocessed at the Hanford Site. As noted in Tables 6.2-2 and 6.2-3, part of the irradiated fuel was shipped to the Nuclear Fuels Services Company plant in West Valley, New York. Another part of the irradiated fuel is still stored in water basins at the K East and K West areas at the Hanford Site. The amounts of ^{137}Cs and ^{90}Sr , decayed to January 1, 2000, in the fuel not processed at the Hanford Site, are listed in Tables 6.2-2 and 6.2-3. These amounts of ^{137}Cs and ^{90}Sr were not introduced into the Hanford Site reprocessing plants.

Table 6.2-3. Inventory of Cesium-137 and Strontium-90 in Reactor Fuel Not Reprocessed at the Hanford Site.

Fuel disposition	Radionuclide content, MCi ^a	
	^{137}Cs	^{90}Sr
Shipped to Nuclear Fuel Services Co. Facility ^b	2.36	1.85
Stored in the Hanford Site K Reactor Basins ^c	12.1	9.31
Total	14.46 ^d	11.16 ^d

^aAs of January 1, 2000

^bBoldt et al. 1996, Myers 1995

^cBoldt et al. 1996, Willis and Praga 1995

^dRounded to 14.5 and 11.2, respectively, in Table 6.2-2.

Boldt et al. (1996) provides considerably more detail concerning fuel irradiated in Hanford Site reactors but not subsequently reprocessed at the Hanford Site. The ^{137}Cs and ^{90}Sr inventories shown in Table 6.2-3 are those cited by Boldt et al. (1996).

6.2.2.3 Cesium-137 and Strontium-90 Removed from Underground Tanks. Contrary to most other fission products, not all the ^{137}Cs and ^{90}Sr originally introduced into the underground Hanford tanks remained there. A major fraction (Table 6.2-4) of these two radioisotopes was transferred to the Hanford B Plant for recovery and subsequent conversion to $^{137}\text{CsCl}$ and $^{90}\text{SrF}_2$ and eventual encapsulation. During the period 1968 to 1972, acidic high-level wastes were transferred directly from the PUREX plant to B Plant. Other smaller fractions of ^{90}Sr and ^{137}Cs were separated directly from the PUREX Plant, the Strontium Semi-Works, and from tank farms.

Table 6.2-4. Inventory of Cesium-137 and Strontium-90 Removed from Tanks.

Transaction	MCI ^a	
	¹³⁷ Cs	⁹⁰ Sr
Released to soil	1.8	0.44
Sent to the Hanford Site B Plant	75.5	40.4
Total	77.3	40.8

^aAs of January 1, 2000.

Boldt et al. (1996) provides full details of the removal of ¹³⁷Cs and ⁹⁰Sr from the underground tanks to the underlying soil or to Oak Ridge National Laboratory. Data for the amounts of ¹³⁷Cs and ⁹⁰Sr (decayed to January 1, 2000) transferred to the Hanford Site B Plant are provided by a recent evaluation (Gehrke 1996).

B Plant ¹³⁷Cs and ⁹⁰Sr Material Balance Considerations. In one approach to calculating global inventories by material balance, the important data are the amounts of ¹³⁷Cs and ⁹⁰Sr transferred from tanks to B Plant and the amounts returned from B Plant to the underground tanks. Estimates for these input/output transfer values are given in Table 6.2-5.

Table 6.2-5. Material Balance for Cesium-137 and Strontium-90 in B Plant and Waste Encapsulation and Storage Facility^a.

Material balance component	MCI ^b	
	¹³⁷ Cs	⁹⁰ Sr
Input		
Transferred to B Plant	75.5	40.4
Output		
Encapsulated	56.7	22.3
Offsite Cask Shipments	2.5	3.9 to 4.6
Solid Waste Disposal	0.47	0.47
Returned to Tanks	1.8 to 11.0	0.9 to 5.5
Residual Facility Contamination	2.7 to 3.7	0.9
Total output	64.2 to 74.4 (85 to 99%) ^c	28.5 to 33.8 (70 to 84%) ^c

^aData from Gehrke (1996).

^bDecayed to January 1, 2000.

^cOf input to B Plant.

For completeness, Table 6.2-5 also lists the quantities of ¹³⁷Cs and ⁹⁰Sr (decayed to January 1, 2000) stored in an encapsulated form in the Hanford Waste Encapsulation and

Storage Facility as well as material balance data for other fractions of radiocesium and radiostrontium transferred from and remaining in the B Plant. The information presented in Table 6.2-5 derives from a recent analysis by Gehrke (1995, 1996).

As indicated in Table 6.2-5, the exact amounts of ^{137}Cs and ^{90}Sr returned from B Plant to the underground tanks are very uncertain. Gehrke (1996) states, "The range is my estimate of 1 to 5 percent losses during 3 cycles of processing, based on the flowsheets used. The wide range reflects the appropriate uncertainty that exists for this stream. The waste loss estimates are probably no better than -50 to +200 percent."

Gehrke thinks that the uncertainty associated with other entries in Table 6.2-5 is less, e.g., +10 percent for encapsulated material, +20 percent for feed and cask shipments, and +50 percent for residual facility contamination. According to Gehrke, the information for the amounts of cesium and strontium in the feed to the B Plant came from a chart (author unknown) prepared in 1985 after shutdown of processing operations in B Plant. Note, also, that estimates for the sum of output streams from B Plant do not equate to the input stream estimates. Consequently, an estimate of ^{90}Sr and ^{137}Cs remaining in underground tanks that is calculated from estimates of "transfers to" and "returns from" the B Plant will contain significant uncertainty.

Fortunately, an alternative estimate of ^{90}Sr and ^{137}Cs remaining in underground tanks can be made by using estimates of other B Plant output streams (given in Table 6.2-5) in a material balance as itemized in Table 6.2-2 and illustrated in Figure 6.2-1. In this alternative, a system envelope is drawn around "Reprocessing plants," "Underground Storage Tanks," and "B Plant operations" (refer to Figure 6.2-1). This method allows the Residual Tank Inventory to be calculated by difference (using the more certain estimates for the "Encapsulated Inventory," and "Inventory Shipped to Oak Ridge") without having to account for the more uncertain "Removed from tanks" and "Returned to tanks" streams.

6.2.2.4 Comparison with Hanford Defined Waste Estimates. Table 6.2-6 shows a comparative accounting for ^{137}Cs and ^{90}Sr , as estimated by three models.

Table 6.2-6. Comparison of Cesium-137 and Strontium-90 Accounting by Model.

	¹³⁷ Cs decayed to January 1, 2000 (MCi) ^a		
	HDW model (Rev. 3) ^b	DKPRO/HDW model (Rev. 4) ^c	DKPRO/SPLIT model ^d (1997)
In reprocessed fuel	94	104.6	104.6
Released to soil	1.3	1.2	1.8
Shipped to other facilities ^e	0	2.5	2.5
Solid waste	0	0	0.5
Residual B Plant contamination	0	3.2	2.7-3.7
Encapsulated	53	57.5	56.7
Remaining in tanks	40.7	41.0	40.4-39.4 ^f
Site accounted	95.0	105.4	104.6
Site unaccounted	-1.0	-0.8	0
	⁹⁰ Sr Decayed to January 1, 2000 (MCi) ^a		
In reprocessed fuel	78	90.1	90.1
Released to soil	0.6	0.97	0.4
Shipped to other facilities ^e	0	4.3	3.9-4.6
Solid waste	0	0	0.5
Residual B Plant contamination	0	0.9	0.9
Encapsulated	23	22.5	22.3
Remaining in tanks	49.7	53.3	62.1-61.4 ^g
Site accounted	73.3	82.0	90.1
Site unaccounted	+4.7	+8.1	0

HDW = Hanford Defined Waste

^aAll decayed to January 1, 2000

^bAgnew et al. 1996. Agnew provided a table showing the ¹³⁷Cs and ⁹⁰Sr accounting used in Revision 3 of the HDW model.

^cAgnew et al. (1997a)

^dData taken from Tables 6.2-2 through 6.2-5 of this report

^eOak Ridge National Laboratory and Pacific Northwest National Laboratory.

^fCalculated by material balance (Fuel - releases and products). Range corresponds to "Residual B Plant" range.

^gCalculated by material balance (Fuel - releases and products). Range corresponds to "other facilities" range.

It is convenient to consider ¹³⁷Cs data in Table 6.2-6 separately from that for ⁹⁰Sr. The following differences between the three model predictions are noticeable.

- The HDW Rev. 3 estimate of the amount of ^{137}Cs in the waste discharged to the Hanford Site tanks is 10.6 MCi less than the value estimated by the DKPRO-1997 code. The HDW estimate apparently derives from data provided by the TRAC model (Jungfleish 1984) while the data used in the later models is from the DKPRO-1997 code (Watrous and Wootan 1997).
- The HDW Rev. 3 estimate of the amount of encapsulated ^{137}Cs (decayed to January 1, 2000) is approximately 4 MCi less than the values used in later models. The DKPRO/SPLIT model value derives from the recent memorandum report by Gehrke (1996).
- The HDW Rev. 3 model does not take into account ^{137}Cs transferred to other sites, solid waste, or residuals remaining in facilities. This omission coupled with a low input value (94 MCi in waste sent to tanks) results in the models prediction of 40.7 MCi ^{137}Cs remaining in tanks--a value which is coincidentally close to the prediction of the later models.
- Comparison of the later models (DKPRO/HDW (Rev. 4) versus DKPRO/SPLIT) indicates close agreement in nearly all transaction values. The DKPRO/HDW (Rev. 4) model does predict 0.6 MCi more ^{137}Cs as remaining in tanks than does the DKPRO/SPLIT model--a relative difference of only 1.5 percent.

For ^{90}Sr global inventories (see Table 6.2-6), the HDW Rev. 3 model predicts only 78 MCi of ^{90}Sr were added to the tanks as a result of fuel reprocessing, whereas the later DKPRO/HDW and DKPRO/SPLIT models predict 90.1 MCi. Other highlights for the ^{90}Sr global inventories are as follows:

- No account is taken in the HDW Rev. 3 model of ^{90}Sr known to have been shipped to other facilities, ^{90}Sr disposed in solid waste or that remaining in facility residues.
- Table 6.2-6 shows 4.7 MCi of ^{90}Sr are unaccounted for in the HDW Rev. 3 model predictions. All or most of the missing ^{90}Sr likely represent material sent to the Oak Ridge and the Pacific Northwest National Laboratories.
- Accounting values used in the later models are approximately the same, with the exception that the DKPRO/HDW (Rev. 4) model predicts about 8.8 MCi less ^{90}Sr remaining in tanks than does the DKPRO/SPLIT model (53.3 MCi versus 62.1 MCi). This difference also shows up as 8.1 MCi being unaccounted for in the DKPRO/HDW (Rev. 4) model's balance. It appears that this imbalance is related to the problem of "missing ^{90}Sr " discussed in Agnew et al. (1997a).

6.2.3 Best-Basis for Cesium-137 and Strontium-90 Global Inventory Values

Four different estimates of the total inventory of ^{137}Cs and ^{90}Sr in DSTs and SSTs are compared in Table 6.2-7.

Table 6.2-7. Comparison of Tank Inventory Estimates for Cesium-137 and Strontium-90.

	¹³⁷ Cs decayed to January 1, 2000 (MCi)			
	TWRS data	HDW Rev. 3	DKPRO/HDW (Rev. 4)	DKPRO/SPLIT model
DST inventory	25.4	20.2	20.3	25.4
SST inventory	9.5	20.5	20.7	14.0 to 15.0
Total inventory	34.9	40.7	41.0	39.4 to 40.4
	⁹⁰ Sr decayed to January 1, 2000 (MCi)			
DST inventory	11.4	16.3	15.2	11.4
SST inventory	41.2	33.4	38.1	50.0 to 50.7
Total inventory	52.6	49.7	53.3	61.4 to 62.1

DST = Double-shell tank
 HDW = Hanford Defined Waste
 SST = Single-shell tank
 TWRS = Tank Waste Remediation System.

The analysis in Table 6.2-6 indicates that about 8 MCi of ⁹⁰Sr are not accounted for in the HDW Rev 4 model's inventory balance. To correct this imbalance in Table 6.2-7, the model's estimate of 38.1 MCi ⁹⁰Sr in SSTs would need to be increased to 46.9 MCi to bring the all-tank total to the value of 62.1 MCi. Even then the ratio of ⁹⁰Sr between single shell and double shell tanks would be inconsistent with the sample-derived estimate of the DKPRO/SPLIT model. (Inconsistencies in the SST/DST ratio are presumably due to shortcomings of solubility functions within the HDW model.) For these reasons, the results of the DKPRO/SPLIT model are used as the global inventory.

Table 6.2-8 summarizes current best-basis estimates of the inventories of ¹³⁷Cs and ⁹⁰Sr in the DSTs and SSTs as well as the global inventories of these radionuclides in all the Hanford Site tanks. The inventories, as of January 1, 2000, of ¹³⁷Cs and ⁹⁰Sr in the 28 DSTs are established, from analytical data, as 25.4 and 11.4 MCi, respectively. Future activities, i.e., more analytical data for ¹³⁷Cs and ⁹⁰Sr contents of the SSTs and validated modeling predictions, will likely result in some revision to the best-basis global inventory data shown in Table 6.2-8.

Table 6.2-8. Global Best-Basis Estimates of Cesium-137 and Strontium-90 Inventories in Hanford Underground Tanks.

Tanks	MCi ^a	
	¹³⁷ Cs	⁹⁰ Sr
Double-shell tanks ^b	25.4	11.4
Single-shell tanks	15.0	50.7
Total	40.4	62.1

^aDecayed to January 1, 2000.

^bAnalytical data.

The best-basis global inventory (Table 6.2-8) of ¹³⁷Cs in the Hanford Site SSTs is 15.0 MCi versus the 9.5 MCi value presently carried in the TWRS estimate. This additional 5.5 MCi of ¹³⁷Cs results as a consequence of an increase of 10.6 MCi in estimated fuel activity offset by a more complete accountability for ¹³⁷Cs losses or transfers to other facilities, solid waste and residual facility contamination.

The best-basis global inventory of ⁹⁰Sr is 62.1 MCi (decayed to January 1, 2000). Of this total, 11.4 MCi are in the DSTs and 50.7 MCi in the SSTs. The former value is identical to that in the present TWRS database and derived from analyses of wastes in each of the 28 DSTs. The best-basis global inventory, 50.7 MCi listed in Table 6.2-8 for the SSTs is 9.5 MCi more than the 41.2 MCi carried in the present TWRS database. The additional ⁹⁰Sr originally introduced into the Hanford Site tanks is discussed in this report (see Table 6.2-2).

Best-basis global inventory values given in Table 6.2-8 (40.4 MCi of ¹³⁷Cs and 62.1 MCi of ⁹⁰Sr at a decay date of January 1, 2000) are equivalent with values given in Table 6.1-6 (46.4 MCi and 71.6 MCi, respectively), allowing for the six year difference in decay date bases.

6.3 URANIUM

Another radionuclide present in tank waste is uranium--significant because of its long half-life and importance to the acceptability of near-surface waste disposal alternatives, and because it is a major component in total waste oxides. Waste core samples have not all been analyzed for uranium isotopic makeup or even for total uranium mass. Therefore, we need to calculate inventories for many of the tanks based on historical fuel throughput and separations plant operating data.

The modelling prediction of uranium inventories in tank wastes is somewhat more uncertain than for fission products since U is one of the several "extractable" fuel components--meaning that another piece of information, the U separation factor, is needed.

The following sections discuss (1) the fuel sources and separation plant history contributing to the tank waste inventory of uranium, and (2) a material balance for U inventory

data.

6.3.1 Uranium Fuel Sources and Separation History

From 1944 through 1971 aluminum clad fuels were processed through three separations processes (BiPO₄, REDOX, and PUREX). In the BiPO₄ process (T and B Plant operations, 1944 through 1956), 100 percent of the uranium in fuels was routed to tank waste and later recovered in a sludge sluicing operation. Recovered sludge was then dissolved and processed through a TBP solvent extraction process for uranium separation and purification, with a low percentage loss (possibly 1 to 2 percent) of uranium being returned from the TBP process to the tanks. However, it is the somewhat larger fraction of uranium bearing sludge that was not recovered in sluicing operations (typically about 5 percent) that contributes to the bulk of the tank U inventory from BiPO₄ operations. Consequently, our knowledge of this portion of the global tank uranium inventory is uncertain to the degree that historical records accurately describe the actual quantities of sludge remaining in tanks and the uranium concentration of those sludges.

Aluminum clad fuels plus a small quantity of zirconium clad fuels were processed through the REDOX and PUREX plants from 1952 through 1972. For the aluminum clad fuel these separation processes recovered a high percentage of the uranium, sending typically less than one percent to the combined cladding waste and extraction waste streams. Our knowledge of the resulting tank uranium inventory contribution is based on process waste analyses as reported through nuclear material accountability records. The quality of these waste sample analyses is thought to be relatively good since (unlike the case of zirconium clad fuel) acidic extraction waste streams contained few solids (i.e., most of the uranium in the waste stream was in solution and thus was amenable to a representative sample and analysis). Fortunately, uranium in the alkaline cladding waste solutions, while not easily measured, was a small part of the total uranium waste loss.

It should be noted that during 1966 and again in 1970, Thoria target elements were also processed through the PUREX plant for the recovery of ²³³U. Waste streams from these two processing campaigns were routed to C-farm tanks and are responsible for contributions of ²³³U and ²³²U to the global uranium inventory.

Zirconium clad fuels processing in the PUREX plant during the post 1971 time period suffered greater uranium waste losses due to (1) the attack of decladding solutions on fuel core uranium (up to 6 percent of the uranium was observed to react), and (2) the inefficiency of plant centrifuges to recover uranium solids from the decladding streams before their transfer to underground waste tanks. Measurement of U losses in Zirflex decladding streams was also uncertain due to the difficulty in getting a representative sample of the solid/liquid slurry. Likewise, the measurement of U in extraction wastes was also believed to be biased low due to the presence of U bearing solids in the waste. Uranium losses to post-1971 zirconium cladding waste streams and extraction waste streams, fortunately, have been contained in only four tanks (241-AW-103, 241-AW-105, 241-AZ-101, and 241-AZ-102). All four of these tanks have been core sampled with results indicating that, indeed, uranium inventory values are significantly greater than would be indicated by plant waste stream accountability records. Thus, for waste from zirconium clad fuel processing, an alternative and more representative

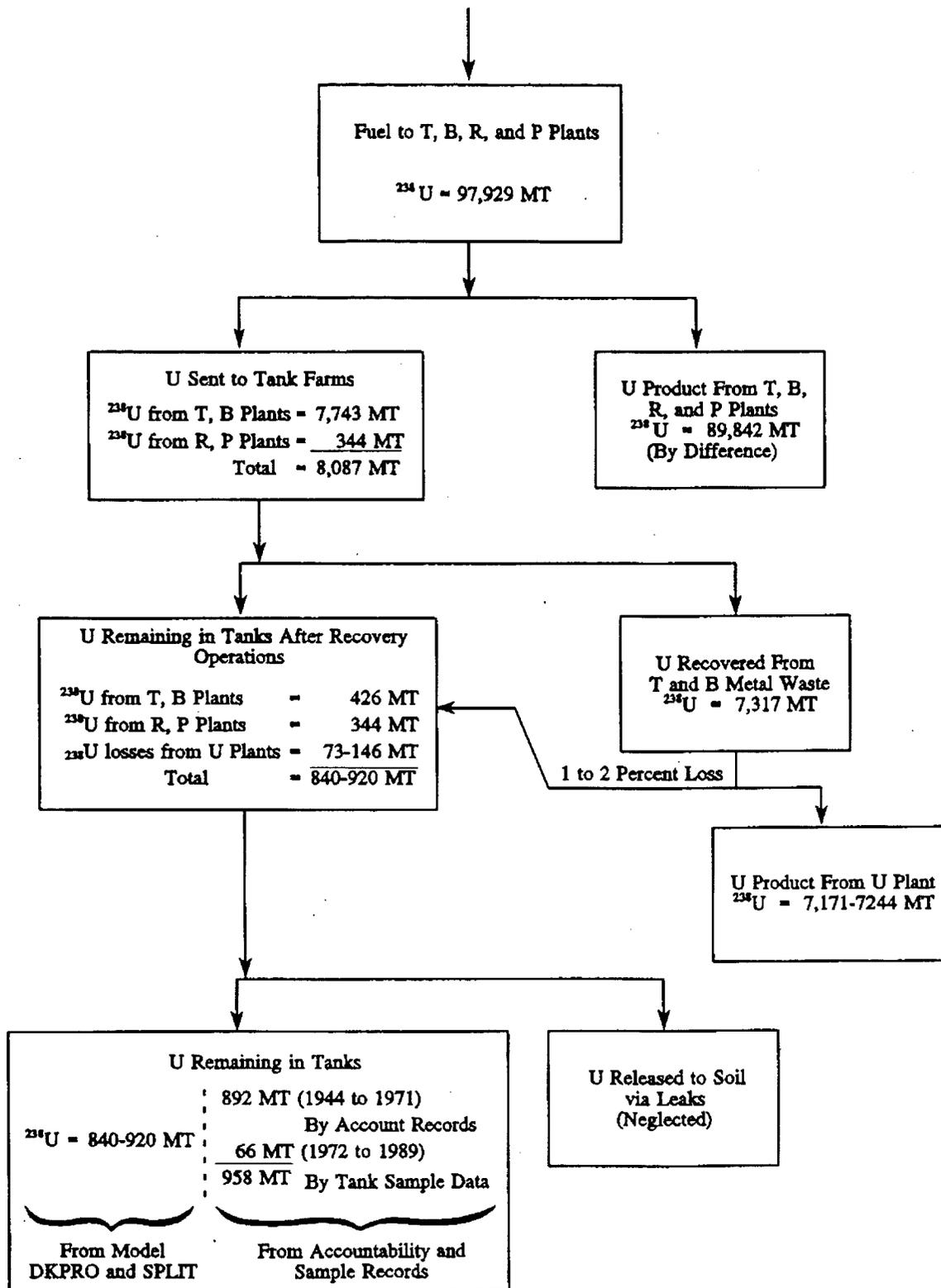
inventory data source is available in core sample results for these four tanks.

6.3.2 Calculation of Global Uranium Inventory

The material balance for uranium, from fuels to tank residuals, is summarized by Figure 6.3-1 in units of metric tons of ^{238}U . Of 97,929 MT ^{238}U in fuels processed through T, B, REDOX, and PUREX plants (based on fuel activity records given in Watrous and Wootan [1997]), Figure 6.3-1 indicates that 7,743 MT ^{238}U were sent to tanks from T/B Plant operations while 344 MT ^{238}U were sent to tanks from REDOX/PUREX operations--this latter U loss value being based on REDOX/PUREX monthly average waste loss factors which are used in the DKPRO/SPLIT model. The material balance then accounts for an assumed 94.5 percent of the T/B uranium being recovered (factor derived from McDonald (1959), and used in the DKPRO/SPLIT model), resulting in an estimate of 7317 MT ^{238}U being sent to Uranium Recovery (UR) extraction operations at U Plant. Uranium losses from UR operations are estimated to have been 1 to 2 percent, which equates to 73 to 146 MT returned to tanks. Residual ^{238}U left in tanks from T/B operations is then estimated as $(7,743 - 7,317 = 426)$ MT. This combined with the UR waste losses $(73 - 146 \text{ MT})$ plus the 344 MT sent to tanks from REDOX/PUREX operations sums to a total ^{238}U tank inventory of 840 to 920 MT for the assumptions used in the DKPRO/SPLIT model.

Figure 6.3-1 alternatively shows the ^{238}U tank inventory as derived from accountability records plus recent tank core samples (also see Table 6.1-4). Here, accountability records for operations 1944 through 1971 (as reported in ERDA 1977) indicate that 892 MT ^{238}U remain in all tanks after accounting for recovery of T/B uranium and measured waste losses from REDOX and PUREX. This alternative inventory method then estimates ^{238}U losses from PUREX operations (1983 through 1989) from core sample results for tanks 241-AZ-101, 241-AZ-102, 241-AW-103, and 241-AW-105 (66 MT ^{238}U), recognizing that core sample results are likely more representative than waste stream samples measured during plant operations. The alternative method then estimates the total ^{238}U tank inventory as the sum of these two sub-inventories $(892 + 66 = 958)$ MT ^{238}U .

Figure 6.3-1. Estimated Disposition of ²³⁸U Processed Through Hanford Separations Plants.



revertain/FIG6-3-1.WPG

6.3.3 Best-Basis for Uranium-238 Global Inventory Value

As discussed in Section 6.3.2, the 958 MT ^{238}U value derived from accountability and tank sample data sources is judged to be the preferred best-basis value. This value for ^{238}U is equivalent to a total uranium tank inventory of 965 MTU as derived in Table 6.1-4.

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Table D2-3. REDOX Process Waste Concentrations Predicted from Flowsheets.
(2 Sheets)

REDOX Coating Waste Concentrations by Flowsheet											
Species	REDOX Flowsheet Number							HDW	HDW	Allen 1976	Allen 1976
	No. 4 (molar)	No. 5 (molar)	No. 6 (molar)	No. 7 (molar)	No. 8 ^a (molar)	No. 9-Al ^a (molar)	No. 9-Zr ^a (molar)	CWR1 (molar)	CWR2 (molar)	Basis-Al (molar)	Basis-Zr (molar)
Al	0.80	1.30	1.91	2.56	2.31	2.32	0.33	2.0	0.8	1.2	---
Bi	0	0	0	0	0	0	0	0	0	---	---
C ₂ O ₄ ²⁻	0	0	0	0	0	0	0	---	---	---	---
Cr	0	0	0	0	0	0	0	0.0030	0.0030	---	---
F	0	0	0	0	0	0	3.4	0	0	---	0.071
Fe	0	0	0	0	0	0	0.014	0.015	0.015	---	---
I	0	0	0	0	0	0	0	---	---	---	---
K ^b	0	0	0	0	0	0	0	0.0022	0.0034	---	0.01
Mn ^b	0	0	0	0	0	0	0	0	0	---	---
Na	2.6	3.9	4.8	5.3	5.9	5.8	6.2	3.9	1.8	3.74	0.72
NO ₂	0.60	1.0	1.4	1.5	1.4	1.5	0.26	1.4	0.28	0.9	---
NO ₃	0.81	0.59	0.80	0.26	1.3	1.1	2.0	0.88	0.89	0.6	0.02
Si	0.0091	0.015	0.027	0.088	0.079	0.079	0	0.030	0.000	0.02	---
SO ₄	0	0	0	0	0	0	0	0.013	0.013	---	---
U	0.0012	0.0013	0.0056	0.011	0.0057	0.0057	0.017	0.019	0.018	---	0.0005
Zr	0	0	0	0	0	0	0.49	0	0	---	0.1
Issue Date	7/51	8/55	10/60	1/65	1/65	9/66	9/66				

HDW = Hanford Defined Waste

REDOX = Reduction and Oxidation

^aIncludes Tank Farm salt solution recycled to provide nitrate ion for Al coating removal or Al fuel bucket dissolution

^bAlthough not shown on the flowsheets after Flowsheet No. 4, KMnO₄ additions continued for a number of years, possibly until September 1959.

Despite this limitation, important conclusions can be drawn from the comparison of the REDOX process flowsheets. The flowsheets indicate that the HLW concentrations for major components (Na, Al, and NO_3) were relatively constant, even though the waste volume generation rate (volume/MTU) varied substantially over the plant's operating life. In contrast, the minor components were fairly constant on the basis of a MTU processed.

Actual essential material usage rates are available for aluminum nitrate nonahydrate, nitric acid and sodium hydroxide (Jenkins and Foster 1978). These usage rates were converted to kgs of chemical per MTU processed and compared to solvent extraction wastes predicted from the six flowsheets (Table D2-4). Even though the flowsheet implementation dates are not exactly known, significant process rework is evident.

The waste concentrations for Al, NO_3 , and Na were also calculated from the essential material usage (Jenkins and Foster 1978) and the waste volumes reported for 1955 through 1965 (Roberts et al. 1992) for comparison with the flowsheet waste predictions (Table D2-5). The actual plant data indicates relatively constant waste concentrations for the three major waste constituents (Al, Na, and NO_3), although concentrations for all three species are slightly more dilute than predicted from the flowsheets (Table D2-3). The agreement between the estimated concentrations based on plant data and the flowsheet predictions is good.

From an operating perspective, the concentration of sodium, aluminum, and nitrate ion would be expected to remain fairly constant regardless of the flowsheet being used or the amount of process recycle. The waste concentrator would be operated to a specific gravity, which essentially determined by the nitrate ion concentration since the atomic weights of aluminum and hydrogen are relatively low. Aluminum nitrate nonahydrate contributes roughly 70 percent of the nitrate ion, so its concentration will also be relatively constant. Since four moles of caustic are required to neutralize the aluminum and convert it to sodium aluminate, the caustic demand is also fixed and the waste sodium is also fairly constant. Improvements in plant operating efficiency are reflected in the reduction of waste volume transferred to the underground storage tanks, not a change in concentration for these three components.

APPENDIX K

RECONCILIATION OF GLOBAL INVENTORY AND TANK SUM VALUES

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APPENDIX K

RECONCILIATION OF GLOBAL INVENTORY AND TANK SUM VALUES

The primary purpose of this report (HNF-SD-WM-TI-740) is to document the process knowledge based estimate of the total (global) inventories of key analytes and radionuclides in Hanford Site underground storage tanks. Subsequent to preparation of this estimate, individual tank-by-tank sample based estimates have been developed and reported in their respective Tank Characterization Reports (TCRs) and in the Tank Characterization Database (TCD). The tank-by-tank inventories were derived by use of both sample data and process knowledge. The summation of tank-by-tank values is the standard inventory in use by the Tank Waste Remediation System (TWRS) program and is the inventory recommended for general use.

The process knowledge based global values presented in Sections 5, 6, and 7, are considered to be a secondary source to the summation of tank-by-tank inventories. The global inventories are substantially improved relative to previous inventory estimates (published prior to 1996) and provide an independent basis for comparison to the summed tank-by-tank values.

K1.0 COMPARISON OF TANK SUMMATION VALUES AND GLOBAL VALUES

The standard inventory activity has produced a global inventory estimate and specific best basis inventories for 177 individual tanks. The summation of individual tank best-basis inventories and the global inventory estimates do not necessarily match due to differences in the way that the estimates were developed (see Section 8.0 of this document). Table K-1 compares the summation of individual tank best-basis inventories to the global inventory as of September 30, 1998. The second column shows the sum of individual tank values that consist of values based on samples (S)¹, engineering assessment (E), or the Hanford Defined Waste (HDW) model (M), or a combination of these bases. Table K-1 also shows the fraction of the best-basis inventory that is derived from sample data. Data from tank sampling may be utilized directly, or may be modified based on process knowledge and engineering judgement. Thus the sample data in this column include the inventory from composite sources, i.e., sample/engineering assessment (S/E) values and sample/HDW model (S/M) values. The values in the last column include only the inventory that is directly from the HDW model (M values). The balance of the tank summation inventories is from engineering assessment (E) values and engineering assessment/HDW model (E/M) values. The engineering assessment (E and E/M) contributions are not shown in Table K-1.

¹S, E, M, and combinations thereof, are designations used in the best-basis inventories to indicate the basis for a value: S = sample based, E = engineering assessment based, and M = Hanford Defined Waste model based. See Appendix J for further clarification of the inventory bases.

Table K-1. Comparison of the Summation of Individual Tank Best-Basis Inventories to the Global Waste Inventory. (5 Sheets)

Analyte	Best-basis tank summation inventories ^a	Global inventory (process-history based)	Ratio of tank sum inventory to global inventory (%)	Percent of tank sum based on sample data ^b	Percent of tank sum based on HDW Model ^b
Chemicals (MT)					
Ag	NR	8.93	NR	NR	NR
Al	7,950	7,845	101	61.1	0.01
Bi	631	580	109	76.6	0.75
Ca	319	214	149	53.1	8.56
Cd	NR	8.20	NR	NR	NR
Cl	930	500	186	59.7	1.61
CO ₃	9,390	4,830	194	62.8	7.17
Ce	NR	8.80	NR	NR	NR
Cr	670	785	85.4	59.0	1.21
F	1,190	1,360	87.8	67.9	0.92
Fe	1,400	1,230	114	77.7	4.75
Hg	1.81	2.10	86.1	11.1	0
K	874	481	182	69.6	0.88
La	51.3	51.0	101	82.1	0.10
Mn	194	105	185	66.8	0.77
Na	48,800	54,200	90.0	63.2	0.15
Ni	174	111	157	74.3	3.52
NO ₂	12,400	NR	NR	NR	NR
NO ₃	52,300	NR	NR	NR	NR
NO ₂ and O ₃	64,700	85,700	75.5	55.1	0.75
OH _{TOTAL}	23,500	23,000	102	(c)	(c)
Pb	84.0	279	30.1	53.6	8.74
PO ₄	5,550	6,000	92.6	61.3	0.19

Table K-1. Comparison of the Summation of Individual Tank Best-Basis Inventories to the Global Waste Inventory. (5 Sheets)

Analyte	Best-basis tank summation inventories ^a	Global inventory (process-history based)	Ratio of tank sum inventory to global inventory (%)	Percent of tank sum based on sample data ^b	Percent of tank sum based on HDW Model ^b
Si	941	570	165	61.8	7.48
SO ₄	3,330	5,000	66.7	57.6	1.51
Sr	45.9	31.3	147	58.8	0
Th	NR	25.6	NR	NR	NR
TOC	1,690	4,000	42.2	61.7	1.01
U _{TOTAL}	929	965	96.3	65.2	7.61
W	NR	15.9	NR	NR	NR
Zr	470	440	107	98.8	0.08
Radionuclides (Ci)					
³ H	2.45 E+04	3.40 E+04	72.2	0.68	86.8
¹⁴ C	3.87 E+03	4.81 E+03	80.6	3.81	93.0
⁵⁹ Ni	8.76 E+02	9.34 E+02	93.8	1.09	92.6
⁶⁰ Co	1.99 E+04	1.23 E+04	162	45.3	13.8
⁶³ Ni	8.64 E+04	9.20 E+04	93.9	1.08	92.0
⁷⁹ Se	7.11 E+02	7.73 E+02	92.0	0.48	85.4
⁹⁰ Sr	5.86 E+07	7.16 E+07	81.9	59.6	6.94
⁹⁰ Y	5.86 E+07	7.16 E+07	81.9	59.6	6.94
⁹³ Zr	3.45 E+03	3.63 E+03	95.1	0	85.8
^{93m} Nb	2.53 E+03	2.69 E+03	94.0	0	89.6
⁹⁹ Tc	2.83 E+04	3.26 E+04	86.9	30.6	60.8
¹⁰⁶ Ru	1.26 E+05	1.04 E+05	121	39.1	0.35
^{113m} Cd	1.66 E+04	1.69 E+04	98.5	0	74.6
¹²⁵ Sb	2.52 E+05	2.08 E+05	121	2.09	8.24
¹²⁶ Sn	1.17 E+03	1.19 E+03	98.4	0	82.2

Table K-1. Comparison of the Summation of Individual Tank Best-Basis Inventories to the Global Waste Inventory. (5 Sheets)

Analyte	Best-basis tank summation inventories ^a	Global inventory (process-history based)	Ratio of tank sum inventory to global inventory (%)	Percent of tank sum based on sample data ^b	Percent of tank sum based on HDW Model ^b
¹²⁹ I	9.91 E+01	6.30 E+01	157	42.6	44.7
¹³⁴ Cs	8.68 E+04	8.89 E+04	97.7	0.14	1.89
¹³⁷ Cs	5.29 E+07	4.64 E+07	114	51.9	2.95
^{137m} Ba	5.01 E+07	4.39 E+07	114	51.9	2.95
¹⁵¹ Sm	2.60 E+06	2.75 E+06	94.5	0	86.4
¹⁵² Eu	1.45 E+03	1.48 E+03	97.9	0	68.1
¹⁵⁴ Eu	1.94 E+05	1.47 E+05	132	25.4	31.9
¹⁵⁵ Eu	1.75 E+05	1.36 E+05	129	14.6	42.6
²²⁶ Ra	5.83 E-02	6.31 E-02	92.5	0	95.8
²²⁷ Ac	8.75 E+01	8.76 E+01	99.9	0	100
²²⁸ Ra	7.75 E+01	7.71 E+01	101	0	96.2
²²⁹ Th	1.80 E+00	1.81 E+00	99.6	0	95.8
²³¹ Pa	1.56 E+02	1.56 E+02	99.8	0	99.8
²³² Th	4.41 E+00	2.11 E+00	209	0	92.1
²³² U	1.32 E+02	1.23 E+02	107	64.8	11.6
²³³ U	5.01 E+02	4.76 E+02	105	64.7	11.6
²³⁴ U	3.47 E+02	3.46 E+02	100	66.6	7.71
²³⁵ U	1.41 E+01	1.45 E+01	97.1	67.2	7.8
²³⁶ U	1.19 E+01	9.57 E+00	124	76.8	7.15
²³⁷ Np	1.84 E+02	1.41 E+02	131	31.7	47.1
²³⁸ Pu	2.69 E+03	2.77 E+03	97.0	89.7	8.81
²³⁸ U	3.10 E+02	3.22 E+02	96.2	65.4	4.15
²³⁹ Pu	5.52 E+04	3.91 E+04	141	78.5	12.0
²⁴⁰ Pu	1.11 E+04	8.93 E+03	125	79.2	10.5

Table K-1. Comparison of the Summation of Individual Tank Best-Basis Inventories to the Global Waste Inventory. (5 Sheets)

Analyte	Best-basis tank summation inventories ^a	Global inventory (process-history based)	Ratio of tank sum inventory to global inventory (%)	Percent of tank sum based on sample data ^b	Percent of tank sum based on HDW Model ^b
²⁴¹ Am	1.06 E+05	6.99 E+04	152	49.7	7.27
²⁴¹ Pu	1.66 E+05	2.29 E+05	77.3	90.0	8.53
²⁴² Cm	1.71 E+02	7.70 E+01	222	59.4	7.55
²⁴² Pu	1.02 E+00	1.16 E+00	87.8	80.4	7.74
²⁴³ Am	1.76 E+01	9.34 E+00	188	15.2	2.29
²⁴³ Cm	3.28 E+01	1.00 E+01	328	74.4	3.73
²⁴⁴ Cm	7.82 E+02	2.42 E+02	323	75.4	3.99

HDW = Hanford Defined Waste

NR = Not reported

^aOnly three significant figures reported.

^bValues in the best-basis inventories are either based on sample data, engineering assessment, or from the HDW model. Sample data in this table contain S, S/E, and S/M values.

^cThe total hydroxide value is calculated by charge balance; includes oxides as hydroxides, not including CO₃, NO₃, NO₂, PO₄, SO₄, and SiO₃.

As of September 30, 1998, seven of the tank summation and global chemical inventories matched within 10 percent and 13 match within 25 percent out of a total of 25 chemical inventories. This is an improvement from September 1, 1997 when only four chemicals matched within 10 percent and seven matched within 25 percent. In 1998 more than 50 percent of the tank summation inventory is based on sample data for 19 of the 25 chemicals. Individual best-basis tank inventories were not developed for 5 chemicals (for which globals were estimated) that were determined to be present in small quantities and for which little analytical data exist.

The radionuclide inventories in many cases appear to reconcile very well. This conclusion is misleading. The tank summation of radionuclide inventories are often derived from model values and therefore are not truly independent from the global radionuclide inventories. This is the result of having limited analytical data for many of the radionuclides. With the exception of uranium and a few other radionuclides, those tank summation and global inventories that reconcile within 10 percent are at least 85 percent or more based on HDW model values.

The best-basis tank inventories and the global inventory for nonradioactive components were independently developed from different sources of information (See Section 8.0 of this report). Each of these methods may contain several possible sources of error. However, consideration of all sources is necessary to establish a standard inventory.

The global inventories, presented in Sections 5.0, 6.0, and 7.0, were developed from process knowledge. Although there are numerous reports and records on fuel fabrication, and reactor and separations plant operation, these records are highly variable in content. Many of the flowsheets and process records were not retained to the present. Often the data are not adequately correlated. In some cases the profusion of data must be screened and approximations made to facilitate interpretation. In other cases only very limited data are available. In general the global inventories are based on the nominal operating conditions for the facility. Typically process flowsheets and material consumption records were used to estimate separation plant waste discharges. These records did not capture times when the plants experienced high losses or were engaged in startup or shutdown activities. Uncertainty is also introduced by losses to cribs and to tank leaks. Although losses to cribs were estimated for some of the components there are large uncertainties in the quantity of chemicals sent to the cribs. Some uncertainty also exists in the radionuclide content of the reactor fuel elements due to uncertainty in the reactor neutron flux and the fission product yield. The global inventories, discussed in earlier sections of this report, contain numerous caveats and assumptions with respect to the uncertainty of the values.

The individual tank specific best-basis inventories are primarily derived from sample data and tank specific process history. When sample data exist for a tank the best-basis inventory is generally derived directly from these data. When sample data do not exist or data are considered suspect, the best-basis inventory is generally derived from a group of tanks, that process history indicates contain the same waste type. Tank sampling data contain uncertainties with respect to analytical error and the statistical distribution inherent in the analysis of a heterogeneous material. Results from a task to evaluate and quantify these factors indicate that the uncertainty limits are generally quite broad (Ferryman et al. 1998). In addition there are less quantifiable uncertainties introduced by analytical bias, poor sample recovery, and lack of samples from the very bottom of the tanks. Interpretive errors in the tank history are also possible owing to the imprecision of tank farm records with respect to waste type and volume. These uncertainty factors have not been evaluated.

For several of the tanks, it was possible to validate the sample data by comparison to separations plant process flowsheets through the tank process history. Validation by this method was impractical for the majority of the tanks due to their complex waste receipt and transfer history.

K2.0 ASSESSMENT OF DIFFERENCES BETWEEN TANK SUMMATION AND GLOBAL VALUES

Following are observations and assessments of differences between tank summation inventory values and global (process history) based values listed in tables K-1 and K-2.

K2.1 CHEMICAL ANALYTES

The best-basis (tank-by-tank summation) values are generally derived from sample data and engineering assessments. The global inventory estimates for chemicals are primarily derived from process flowsheets, essential material records and other process records.

Aluminum. The best-basis (tank-by-tank summation) inventory for aluminum is considered to be confirmed by the close agreement with the global value.

Bismuth. The best-basis (tank-by-tank summation) inventory for bismuth is considered to be confirmed by the close agreement with the global inventory.

Calcium. The best-basis (tank-by-tank summation) inventory for calcium is considered to be more accurate than the global calcium value. The assessment of calcium sources (process usage estimate of 123 MT) only partially accounts for the measured tank-by-tank summation inventory of 314 MT. The global estimate derived from 1996 sample data projected a Ca inventory of 214 MT. This value is superseded by the current tank-by-tank summation. Several anecdotal leads such as damaged grout linings in cells, or calcium impurities in untreated flush water have been suggested as the basis for the difference between the standard inventory and the global inventory but upon evaluation these sources do not fully account for the differences.

Carbonate. The best-basis (tank-by-tank summation) value for carbonate is significantly higher than the global estimate and is considered to be more accurate than the global estimate. An adequate basis for estimating carbonate resulting from absorption of CO₂ from the atmosphere or from organic carbon degradation has not been included in the global inventory estimate.

Chloride. The best-basis (tank-by-tank summation) value for chloride is significantly higher than the global estimate. The tank-by-tank summation value is expected to be more accurate than the global estimate. The global chloride inventory is based on extrapolation from the limited sample data available in 1996. The major identified source of chloride is as an impurity in sodium hydroxide. The tank-by-tank summation value can be readily justified by this source and is considered to be the more accurate estimate.

Chromium. The best-basis (tank-by-tank summation) value for chromium is generally confirmed by the global inventory evaluation. Most of the global inventory estimate is attributed to chemical usage by the REDOX Plant. The global inventory assessment notes that there were frequent REDOX flowsheet modifications related to the ruthenium oxidation step. A sensitivity

analysis concluded that the REDOX Plant consumed 703 to 895 MT of chromium. Consumption at B and T Plant, and allowances for corrosion of stainless steel increase this to 785 to 980 MT. The lower value (785 MT) was in agreement with the HDW model result and was selected as the global value. The uncertainty in the chromium usage at REDOX and in the corrosion rate of stainless steel may account for the small difference between the global and best-basis value.

Fluoride. The best-basis (tank-by-tank summation) value for fluoride is slightly lower than the global inventory estimate. Fluoride is determined by ion-chromatography of water soluble samples. Acid digested and KOH fusion samples cannot be analyzed by ion chromatography. Sludge and saltcake samples can contain water insoluble fluoride compounds that are not measured by ion-chromatography. Additional investigation is warranted.

Hydroxide. The best-basis (tank-by-tank summation) value and the global value for hydroxide are both calculated as the amount of hydroxide required to achieve anion/cation charge balance. The hydroxide inventory is not an indication of alkalinity or free hydroxide as most of the hydroxide may be present or bound in multivalent metal hydrates. The free hydroxide found in supernatants is expected to be an order of magnitude less than the best-basis inventory value.

Iron. The iron best-basis (tank-by-tank summation) value is considered to be confirmed by the global inventory estimate.

Lanthanum. The best-basis (tank-by-tank summation) value for lanthanum is considered to be accurate based on the close agreement with the global estimate.

Lead. The lead global value is based on very limited experimental measurements of the thickness and composition of the intermetallic lead layer on uranium metal coupons. These measurements were used to estimate lead in fabricated aluminum clad fuel. The best-basis inventory for lead (tank-by-tank summation) established by sample analysis is not unrealistic when the uncertainties in the global (process knowledge) based estimate are considered. Solubility data collected by Agnew (1994) indicates that nearly all the lead would precipitate with the cladding waste solids; however, the cladding waste samples of several tanks exhibit lead concentrations well below those needed to support the global estimate. Thus, the tank summation value is considered the best estimate.

Manganese. The best-basis inventory (tank-by-tank summation) indicates substantially more manganese than was identified by the global inventory estimate. The primary use of manganese was as KMnO_4 for ruthenium oxidation in the REDOX process headend. Over the life of the plant there were frequent REDOX flowsheet modifications related to the ruthenium oxidation step. Best-basis inventories for both potassium and manganese are higher than the process knowledge based estimates indicating that KMnO_4 usage was probably higher than indicated from available records. The tank summation value is considered to be the best estimate.

Mercury. The best-basis inventory (tank-by-tank summation) of mercury is derived from sample values and a process knowledge distribution of the global value. Because of changes to the sample data in FY 1998, the tank-by-tank summation no longer agrees with the global value.

Also there is conflicting process knowledge on which process streams would contain mercury. The best-basis inventory assumes, consistent with the HDW model, that mercury was used to suppress radionuclide volatilization during decladding operations up through 1969. This assumption routes the mercury to cladding wastes. Process knowledge summarized in Section 5.14 of this report states that mercury was used to catalyze nitric acid dissolution of certain aluminum alloy fuels and was used to suppress radioiodine volatilization during nitric acid dissolution of short-cooled uranium metal fuel. This dispositioned the mercury to the high-level waste streams. The current assessment is that the HDW model assumption is wrong and that the process knowledge assessment in Section 5.14 is correct. A future update of the process knowledge based distribution of mercury is planned.

Nickel. The best-basis inventory (tank-by-tank summation) is believed to be on the order of 30-40% high, i.e. the global inventory may be more accurate. The discrepancy is due to individual tank-by-tank estimates that are based on sample results likely contaminated from sample fusion in nickel alloy crucibles. A re-evaluation of the sample data is being addressed in FY 1999.

Nitrate and Nitrite. Comparison of the best-basis (tank-by-tank summation) value to the global inventory implies that the tank summation nitrate and nitrite inventory may be low. This discrepancy, if it exists, may be due to (1) estimates for TX Farm which is essentially unsampled, (2) errors in the existing saltcake templates, and/or (3) uncertainties in the interpretation of sample liquid fractions. Data improvement plans include standardizing the interpretation of liquid sample data and updating of waste type templates. Conversely, the global inventory may be high due to uncertainties in flowsheet implementation, product rework and startup allowances, and the amount of chemicals sent to cribs.

Phosphate. The best-basis (tank-by-tank summation) value for phosphate is considered to be confirmed by the global inventory.

Potassium. The best-basis (tank-by-tank summation) value is significantly larger than the global inventory estimate. The measured inventory for manganese implies that the global inventory did not account for all of the KMnO_4 consumed. Also the potassium global inventory value does not include an allowance for potassium impurities in essential materials. Higher KMnO_4 usage, however, would account for only 15% of the potassium difference. Impurities in caustic used by Hanford have been suggested as a source of potassium. Although potassium is not listed as an impurity in industrial standards for caustic, potassium is present in sea water, a typical source of salt from which caustic is manufactured. Based on the K to Na ratio of sea water (0.036) and the inventory of sodium in the tanks, 1,900 MT of potassium would have been introduced if the salt used for caustic manufacture was not purified. However, during solar evaporation of sea water, nearly all of the Mg and Ca is separated from salt as a residual brine. Based on low initial concentration and high solubility of potassium, it is likely that this purification step would remove most of the potassium. Thus caustic is an unlikely source of the additional potassium found in the tank summation inventory.

Silicon. The global inventory estimate identifies substantially less silicon than has been measured in the tanks. Under-estimation of the amount of Al-Si bond material in aluminum clad

fuel has been suggested as a possible source of this discrepancy. However the observed difference between the best-basis inventory and the global inventory is substantially larger than can be reasonably attributed to this source.

Sodium. Comparison of the best-basis (tank-by-tank summation) value to the global inventory implies that the sodium best-basis inventory may be low. This discrepancy, if it exists, may be due to uncertainties in the interpretation of sample liquid fractions. Un-sampled tanks could be another contribution to this discrepancy. Estimates for TX Farm, which is essentially un-sampled, account for 17 percent of the best-basis inventory. Further evaluations of new sample data will be performed.

Sulfate. The global inventory estimate identifies substantially more sulfate than has been measured in the tanks. This discrepancy, may be due to (1) estimated inventories for TX Farm which is essentially unsampled, (2) errors in first generation saltcake templates, and (3) the presence of water insoluble sulfate forms. Many tanks have been identified where the inductively coupled plasma analysis of sulfur is significantly larger than the ion-chromatography result for sulfate indicating water insoluble sulfate forms. Individual best-basis inventory evaluations may have incorrectly assessed the ICP data. Data improvement plans include updating of waste type templates and the auditing of sulfate inventory calculations based on sample data, since the sulfate is an important glass volume driver for both LLW and HLW vitrification feed.

Strontium. The best-basis (tank-by-tank summation) value is significantly larger than the global inventory estimate. The difference may be due to un-accounted for strontium impurities in purchased chemicals and untreated water, or overly conservative interpretation of detection limit analytical results. Due to plans to recover ⁹⁰Sr for beneficial use in the mid 1960s, the strontium content of purchased chemicals became controlled. The first reference to strontium limits in the essential material manuals is January 1, 1964. Chemical impurities after January 1, 1964 appear to be an unlikely source of appreciable strontium. The strontium content of chemicals and water prior to January 1, 1964 is not known and may have been a significant source of strontium. However there is no evidence for this at this time.

Total Organic Carbon. The best-basis (tank-by-tank summation) value is significantly smaller than the global inventory estimate. The lower tank-by-tank summation value indicates that approximately half of the TOC sent to the tanks has degraded. This is a reasonable conclusion and the tank-by-tank summation is accepted as the best-basis inventory. Comparison of the best-basis inventory TOC values to the Organic Nitrate Safety Program TOC database is planned.

Uranium. The best-basis (tank-by-tank summation) value for uranium appears to be accurate based on comparison to the global estimate. The global estimate was established by material accountability records (1944-1971) and sample data (1972-1989).

Zirconium. The best-basis (tank-by-tank summation) value for zirconium is confirmed by the global inventory.

K2.2 RADIONUCLIDE ANALYTES

The global inventory estimates for radionuclides are derived from several sources. These sources include the DKPRO/SPLIT model, the DKPRO/HDW model, and material accountability records (see Table K-1). The details and differences of these sources are discussed in Section 6.0 and are summarized in Table 6.1.3. Best-basis inventory estimates for individual tanks are either derived from sample results or are estimated by the HDW model when sample data are not available. Many of the radionuclide inventories for 241-AZ-101 and -102 are based on a special ORIGEN2 run specific to these tanks.

Tritium. The global and model estimates for tritium are highly dependant on vapor/liquid partitioning assumptions that distribute ^3H between airborne releases, process condensate releases to cribs and evaporator bottoms sent to the tanks. The global inventory is obtained from the DKPRO/SPLIT model which attempts to account for losses of ^3H to separations process condensates and atmospheric emissions. The HDW model assumes that 100 percent of ^3H in fuel is routed to tank wastes. The best-basis inventory (tank-by-tank summation) includes some ^3H sample-based estimates for individual tanks, but nearly all values are estimated by the HDW model (Table K-1). Since most of the best-basis values are from the HDW model the best-basis inventory is expected to be conservative relative to the actual inventory.

Carbon-14. Most of the individual tank best-basis estimates and all of the global inventory are derived from HDW model results. The sample results imply that the process split factors assumed by the HDW model may have underestimated the ^{14}C airborne releases and losses to cribs. However, the sample values for AZ tanks are suspected to be low since only sludge analyses were performed.

Nickel-59 and Nickel-63. Nickel is a relatively insoluble species that is expected to fully partition to the tank waste. Beginning in 1959 the aluminum alloy fuel cladding contained 9,000 to 13,000 ppm nickel. Prior to 1959 the fuel cladding alloy contained no more than 100 ppm nickel. The HDW model assumes a constant 800 ppm, thus ^{59}Ni and ^{63}Ni activation products can be biased high for pre-1959 wastes and biased low for post 1959 wastes. The best-basis inventory is considered accurate although few sample results are available.

Cobalt-60. Cobalt is expected to fully partition to the tank waste. The ORIGEN2 estimates for activation product, ^{60}Co , assume a cobalt impurity level of 10 ppm in cladding and 0 ppm in core uranium. Since the actual levels of cobalt in uranium are unknown but probably not zero, it is predictable that the HDW model results are low. Thus, the best-basis tank sum is considered to be the more accurate estimate.

Selenium-79. Most of the best-basis (tank-by-tank summation) and all of the global inventory estimate for ^{79}Se is based on the HDW model. Curie values for ^{79}Se calculated by the HDW model are suspected of being high by about a factor of eight due to the use of an erroneous half-life value in the ORIGEN2 data library. Selenium is expected to be fully partitioned to the tank waste. Correction of the half-life error is planned.

Strontium-90 and Yttrium-90. The best-basis (tank-by-tank summation) value for ^{90}Sr is approximately 20 percent less than the ^{90}Sr value determined by the global analysis. This discrepancy, may be due to (1) estimated inventories for TX Farm which is essentially unsampled, (2) errors in first generation saltcake templates, or (3) the lack of representative ^{90}Sr analytical data. Frequently core samples have not been analyzed for ^{90}Sr or have only been partially analyzed. The lack of complete data may have resulted in the under-reporting of ^{90}Sr in tanks and templates by the best-basis inventory. Further evaluations of new sample data will be performed.

Zirconium-93, Niobium-93m, Cadmium-113m, Tin-126, and Samarium-151. The ^{93}Zr , $^{93\text{m}}\text{Nb}$, $^{113\text{m}}\text{Cd}$, ^{126}Sn , and ^{151}Sm values in the best-basis (tank-by-tank summation) inventory are almost entirely derived from the HDW Rev 4 model and are subject to the model limitations. The HDW model over-estimates the distribution of these isotopes to tanks 241-AZ-101 and 241-AZ-102 based on the special ORIGEN2 run for these tanks and under-estimates the inventory of these isotopes in the rest of the tanks. The tank-by-tank summation corrects the inventory of 241-AZ-101 and 241-AZ-102 but does not adjust the inventory of the rest of the tanks (i.e., HDW values are used). The result is that the best-basis inventory slightly under-estimates the ^{93}Zr , $^{93\text{m}}\text{Nb}$, $^{113\text{m}}\text{Cd}$, ^{126}Sn , and ^{151}Sm inventories.

Technetium-99. The best basis (tank-by-tank summation) value for ^{99}Tc agrees reasonably well with the process knowledge based (global) estimate. The best-basis value is likely the more valid since the global estimate does not account for known losses of Tc that occurred during fuel separation operations.

Ruthenium-106. Ninety-nine percent of the ^{106}Ru inventory is found in tanks 241-AZ-101 and 241-AZ-102. The best-basis (tank-by-tank summation) inventory estimates from ORIGEN2 indicate about 20 percent more ^{106}Ru than does the global (HDW model) for these tanks. Considering the relatively rapid decay of ^{106}Ru , a discrepancy of 20 percent is not surprising. An error of this size could be caused by (1) uncertainties in the exact date of sample counting, and/or (2) approximations in the models estimate of fuel discharge dates.

Antimony-125. Eighty-nine percent of the ^{125}Sb inventory is found in tanks 241-AZ-101 and 241-AZ-102. The best-basis (tank summation) inventory estimates from the special ORIGEN2 run for these tanks shows about 25 percent more ^{125}Sb than does the global (HDW model) for these tanks. The ^{125}Sb best-basis inventory of the rest of the tanks is nearly all from HDW model values.

Iodine-129. The best-basis (tank-by-tank summation) of ^{129}I is somewhat larger than the global estimate. The global (HDW model) estimate conservatively assumes that all of the ^{129}I in the fuel was routed to the waste tanks. Thus the reason for the higher tank sum is not inherently obvious. An audit of the eight sample-based and engineering-based tanks in the best-basis estimate, i.e. those tanks that collectively contain over 50 percent of the ^{129}I inventory is needed. The difference could be due to sample error, but is likely not due to underestimated model values. For example, the sample based inventory for tank 241-AZ-101 is 7.1 Ci, while the ORIGEN2 model value is 2.78 (assuming 100 percent of the ^{129}I is routed to tank 241-AZ-101). The sample

based value (7.1 Ci) is the average of two dissimilar core sample results (<0.14 Ci from Core 1, and 11.5 Ci from Core 2). If the Core 2 value is thrown out as being obviously too high, the remaining sample data (<0.14 Ci) give good agreement with the ORIGEN2 model value (0.10 Ci to 241-AZ-101) after applying a PUREX process split factor to account for 3.5 percent of the ^{129}I in fuel being routed to the high-level waste stream. In conclusion, both best-basis and global inventory values are likely to be significantly higher than actual values. Further evaluation is warranted.

Cesium-134. The ^{134}Cs values in the best-basis (tank-by-tank summation) inventory are primarily derived from the HDW Rev 4 model and are subject to the model limitations. The global ^{134}Cs inventory is calculated by the DKPRO/SPLIT model which calculates a slightly higher ^{134}Cs inventory than does the HDW model. The HDW model slightly under-estimates the distribution of ^{134}Cs to tanks 241-AZ-101 and 241-AZ-102. The tank-by-tank summation corrects the inventory of 241-AZ-101 and 241-AZ-102 but does not adjust the inventory of the rest of the tanks. The result is that the best-basis inventory slightly under-estimates the ^{134}Cs inventory.

Cesium-137 and Barium-137m. The best-basis (tank-by-tank summation) values for ^{137}Cs and $^{137\text{m}}\text{Ba}$ are considered to be confirmed by the close agreement with the global values.

Europium-152. The ^{152}Eu values in the best-basis (tank-by-tank summation) inventory are primarily derived from the HDW Rev 4 model and are subject to the model limitations. The HDW model over-estimates the inventory of ^{152}Eu to tanks 241-AZ-101 and 241-AZ-102 based on the special ORIGEN2 run for these tanks and under-estimates the ^{152}Eu inventory in the rest of the tanks. The tank-by-tank summation corrects the inventory of 241-AZ-101 and 241-AZ-102 but does not adjust the inventory of the rest of the tanks, i.e., HDW values are used. Tanks 241-AZ-101 and 241-AZ-102 contain 28 percent of the ^{152}Eu inventory. The result is that the best-basis inventory slightly under-estimates the ^{152}Eu inventory.

Europium-154. The best-basis (tank-by-tank summation) of ^{154}Eu is significantly larger than the global estimate. The global ^{154}Eu estimates for 241-AZ-101 and 241-AZ-102 from the HDW model are 30 percent lower than the best-basis ORIGEN2 estimates. Tanks 241-AZ-101 and 241-AZ-102 account for 40 percent of the best-basis inventory. The balance of the best-basis inventory, which includes sample values and engineering estimates is about 25 percent higher than the balance of the global HDW model estimate.

Europium-155. The best-basis (tank-by-tank summation) of ^{155}Eu is significantly larger than the global estimate. The ORIGEN2 estimates of ^{155}Eu in tanks 241-AZ-101 and 241-AZ-102 are about 90 percent higher than the respective global (HDW model) estimates. The best-basis inventory uses an ORIGEN2 ^{155}Eu value for 241-AZ-101 and a HDW model value for 241-AZ-102. The engineering estimate for 241-AZ-102 included a sample-based heel inventory of ^{155}Eu than was determined to be ludicrous. Most of the best-basis estimates for the remaining tanks (152) are from the HDW model with only 23 tanks based on sample data or engineering estimates. The best-basis inventory of the remaining tanks is about 23 percent higher than the balance of the global (HDW model) estimate.

Neptunium-237. The best-basis (tank-summation) value for ^{237}Np is about 30 percent higher than the global ^{237}Np estimate. This difference appears to be due to the ^{237}Np inventories in 241-AN-103 and 241-AN-105 which account for 30 percent of the best-basis total tank inventory. The best-basis values for these two tanks are the largest contributors to the tank-summation value and are several times larger than the values estimated by the HDW model. The best-basis values for 241-AN-103 and 241-AN-105 are based on "bounding value estimates" that are derived from analytical detection limit data. Further investigation of the basis for the best-basis ^{237}Np inventories of 241-AN-103 and 241-AN-105 is warranted.

Plutonium -238, -239, -240, -241, -and -242. The tank sum ^{239}Pu and ^{240}Pu inventories are significantly higher than those estimated by the global inventories (Table K-1). Examination of the analytical data shows a major source to be from the PUREX process cladding waste in 241-C-102, 241-C-103, 241-C-104, and 241-C-106. The other major sources are tanks 241-AY-101, 241-TX-118, and 241-SY-102. The plutonium in C-farm tanks comprises 33 percent of the tank sum inventory for ^{239}Pu and ^{240}Pu , and represents a significantly larger loss of Pu to the PUREX process cladding waste than assumed by the HDW model or global inventory estimates. The tank summation total Pu inventory accounts for approximately 940 kg which is in close agreement with Pu inventory estimates used in recent tank criticality safety considerations. The ^{238}Pu , ^{241}Pu , and ^{242}Pu isotope inventories are slightly less than predicted using isotopic ratios as predicted by the HDW model.

Americium-241 and -243, and Curium-242, -243, and -244. The tank summation inventories for this group of radionuclides are significantly higher (50 percent to 3-fold) than those estimated by the global model (DKPRO/HDW). Comparisons of sample based and model predicted ^{241}Am inventories for selected tanks suggests that sample results are biased high. This could be the reason for the tank summation inventory of ^{241}Am being 1.5 times greater than the global model prediction. A major reason for the discrepancy in sample versus model based inventory values for ^{243}Cm and ^{244}Cm is traced to sample results for tank 241-AW-105. Here the sample based inventory is essentially equivalent to the model based inventory for all 177 tanks. The large sample based inventory value for tank 241-AW-105 appears to be an anomaly since the other zircaloy cladding waste receiver tank (241-AW-103) contains 4000 fold less by sample determination. The possibility of analytical errors for both Am and Cm isotopes will be investigated.

Uranium-233, -234, -235, -236, and -238. Many of the uranium isotopic inventory estimates are based on the total uranium analysis of samples. The global basis uses accountability and sample data, with the result that global inventory values closely match tank summation values. These inventories are significantly lower than those estimated from total uranium by the HDW Rev. 4 model. The HDW model assumes that significant amounts of TBP (uranium recovery process) waste remain in the tanks while sample results and observations from Rodenhizer (1987) and MacCready (1957) confirm that recovery of uranium was more efficient.

Thorium-232. There are no analytical data for thorium-232. The best-basis tank summation thus reflects DKPRO/HDW inventory values which are about two-fold higher than those estimated by the DKPRO/SPLIT model. However, the DKPRO/SPLIT values are judged to be

the more realistic, being based on accountability measurements made during the thorium fuel processing campaigns in 1966 and 1970. A re-evaluation of the tank-summation basis will be made.

Radon-226, Actinium-227, Radon-228, Thorium-229, and Protactinium-231. The tank summation inventories for these isotopes (alpha decay chain daughters) are based essentially on the HDW model estimates, due to lack of any direct analytical data from samples. As expected, the tank-sum inventory is essentially identical to the global inventory as estimated by the DKPRO/HDW model. As such, the estimate for these daughter radionuclides is in error due to the lack of proper second-order decay functions in the HDW model. Future upgrades to the HDW model may delete these minor radionuclides from the near-term list of key nuclides.

K3.0 RECONCILIATION STRATEGY

The tank specific best-basis inventories continue to evolve as new sample data are made available. The new data will be reviewed and used to revise inventory estimates that are currently based on engineering assessment or the HDW model. The new data will also be used to update the tank groups used to empirically estimate tanks without sample data. Updating of wastes compositions for the tank groups is expected to be the major source of change to the tank summation inventories in the next year. However once the empirically derived waste compositions are updated this year (FY 1999), the tank summation values should be less volatile. At this point it will be appropriate to initiate further reconciliation of the tank summation and global values. This reconciliation is necessary to assure that a single standardized inventory is in use by the Tank Waste Remediation System.

K3.1 RECONCILIATION OF CHEMICAL INVENTORIES

The first step of reconciliation will be to determine whether or not the tank summation value is in agreement with the global value. In principle the tank summation value would be in agreement with the global value when the uncertainty limits of the two values overlapped. However the uncertainty limits on the best-basis inventories and the global inventories are quite broad. Thus to establish a single standard inventory engineering judgment must be applied. This can be accomplished by the use of a screening value negotiated with the end-users. For this discussion, a value of 15 percent was selected. Twelve of the 25 chemicals and 27 of the 46 radionuclides reported in the best-basis inventories fall within this limit. Best-basis inventories were not developed for 5 of the global chemicals for which the total inventory is small and the analytical data are limited.

For those analytes where the global and tank summation inventories agree within 15 percent, the tank summation value will be accepted as the best-basis inventory. Although coincidental agreement is a possibility, concurrence of the global and tank summation inventories will be re-checked in the future as new sample data are evaluated. Inventory data

will also be checked by comparing tank inventories (and/or tank concentrations) to each other to identify suspect data.

For those analytes where the tank summation value and the global value do not agree within 15 percent, the tank summation inventory will be accepted when there is an acknowledged poor basis for determination of the global estimate. The global values for Ca, Cl, CO₃, K, Mn, Ni, Pb, Si, Sr, TOC, and U, for example are in general more uncertain than the other analytes. These analytes are ones that were present as trace impurities or that are difficult to quantify based on the available process records

For those analytes where the tank summation value and the global value do not agree within 15 percent, the global inventory will be accepted when there is an acknowledged lack of sample data or the sample data consists largely of bounding values. In this case, that fraction of the global inventory not accounted for by sample data will be distributed to the remainder of the tanks using process knowledge or the HDW model distribution.

K3.2 RECONCILIATION OF RADIONUCLIDE INVENTORIES

For those radionuclides where the global and tank summation inventories agree within 15 percent, the sum of individual tank best-basis values will be accepted as the global inventory. This will be acknowledged in the appropriate section of this document. Note that agreement may occur due to a lack of analytical data and the dependence on the HDW model values.

When the tank summation and global inventories do not agree, the authenticity of the tank summation sample and engineering results will first be checked. Next to be checked are the assumptions and calculation methods of the HDW Rev. 4 model. If agreement is not obtained, reconciliation will be obtained by the following steps:

When the tank summation radionuclide inventories no longer agree within 15 percent of the global radionuclide inventory, inventory normalization is necessary. Inventory normalization will be obtained by the following steps: (1) subtracting the sum of the sample and engineering based values from the global value, and (2) redistributing the remaining inventory in accordance with ratios defined by the HDW model or by process knowledge.

If redistribution of the remaining inventory in accordance with the HDW model becomes suspect due to the small number of tanks for which only model values exist, the basis for global inventory will be re-examined, and the individual tank best-basis inventories will be re-examined in order of the largest values first. Reconciliation will be complete once the tank summation value is within 15 percent of the global value.

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