Section 6
Discussion and Conclusions

6.0 Discussion and Conclusions

Based on the review of the documents located, the RL Team concludes the following:

6.1 Recycled Uranium Shipped and Received

6.1.1 Operating History

Uranium recovery from irradiated fuel began at Hanford in 1952 using the REDOX process located in the 202-S Plant. Shortly thereafter, the U-Plant also began supplying uranyl nitrate hexahydrate (UNH) containing uranium recovered from the waste tanks. In 1956, the PUREX Plant came on line and also recovered uranium, as UNH, from irradiated fuel for calcination and recycle. The UNH product was piped from REDOX Plant and U-Plant and trucked from PUREX to the UO_3 Plant. At the UO_3 Plant the UNH was converted by calcination to UO_3 powder. The powder was sampled and packaged into either drums or specially designed "T-Hoppers" for shipment.

6.1.2 Recycled Uranium Specifications

Hanford received recycled uranium metal billets for reactor fuel rod manufacture starting about mid-1952; however, the recycled uranium used to produce these billets had been processed through the gaseous diffusion plants (GDPs) and was reported to contain approximately 30 parts plutonium per trillion parts uranium. After about 1963, as a result of a process change at Fernald involving blending, their metal billet plutonium specification rose to a not to exceed 10 parts plutonium per billion parts of uranium level. Hanford did not routinely perform a radionuclide analysis on the incoming billets, but relied on the shipper.

6.1.3 Recycled Uranium Shipments and Receipts

In all, Hanford shipped approximately **109,792** metric tons (MTU) of recovered (recycled) uranium. Of this, **74,491** MTU were shipped (as UO_3) to the Paducah GDP, **4,404** MTU were shipped to the K-25 GDP and Y-12 Plant, and **25,251** MTU were shipped to Fernald. Metal turnings and scrap produced during fuel rod manufacture were returned to Fernald for recovery into new fuel rod billets. Lesser quantities of recycled uranium were sent to Harshaw Chemical Co. and Mallinckrodt Chemical Works for further refining to remove non-radioactive contaminants. Additional recovered uranium, in minor quantities, was sent to over 100 other destinations to support various DOE missions.

Hanford received and processed approximately **109,144 MT** of recycled uranium, with approximately 85% (92,767 MT) being received from Paducah, Fernald and Oak Ridge. Uranium metal received for fuel fabrication before 1952 was made from natural uranium and is out of scope for this report. With the exception of the material remaining at Hanford, the majority of this material was used for fuel, irradiated in the Hanford

reactors, processed in the separations facilities to recover the plutonium and uranium, and the recovered uranium shipped offsite for use or recycle.

6.1.4 Current Inventory

Excluding uranium in solid and liquid waste and releases to the cribs, Hanford currently has approximately **4,006** MTU remaining in various forms including metal received for fabricating fuel, unused fuel, irradiated fuel, unirradiated uranium in mixed oxide fuel at the Fast Flux Test Facility and at the Plutonium Finishing Plant, and recovered uranium as UO_3 stored in T-Hoppers awaiting final disposition. The recycled uranium in the irradiated fuel (2,137 MTU) is outside the scope of this study.

~958 MTU is mixed with fission products and other chemical wastes in high-level waste storage tanks. **~1,054** MTU is buried solid waste, and **~162** MTU was released to the environment through various cribs, ponds, and ditches.

6.1.5 Shipper/Receiver Differences

During and after the May 17, 2000 Uranium Mass Balance Project workshop at Oak Ridge, TN, the RL team worked with other site representatives to compare shipping and receiving quantities of recycled uranium. For the Hanford shipments to Paducah, the percent variance between 1952 through March 30, 1999, was approximately 0.02% of the approximate 74,491 MT total. Percentage variances for shipments from Hanford to Fernald were approximately 0.7%. Percentage variances for Hanford receipts from Fernald were approximately 0.2%. Comparison between Hanford and Oak Ridge for shipments and receipts to the Oak Ridge aggregate of K-25 and the Y-12 will not be completed until the Y-12 transactions are prepared. The percentage variance for Hanford shipments to Savannah River (DuPont) were approximately 0.3%.

6.1.6 Inventory Difference

Hanford uranium shipments, receipts, and material in storage, and waste records indicate a small material difference of about **0.5** wt% of the uranium received remains unexplained based upon the reviewed records. The calculation of this material difference includes an estimate that **~140** MTU was consumed during reactor operations and the generation of plutonium. The material difference resides largely in the uncertainties associated with quantities of uranium in liquid and solid wastes in the waste tanks, and in the estimate of uranium fissioned and transmuted to operate the reactors and generate plutonium.

Section 6
Discussion and Conclusions

6.2 Typical Impurities and Isotopic Composition

6.2.1 Plutonium

Hanford participated in development of the recycled UO_3 product specification starting in about 1950. By 1952, before commencement of uranium recovery operations, a preliminary specification requirement of less than 10 parts Pu per billion parts uranium was established. This limit was firmly established in 1953 and remained in place for the entire UO_3 production period. The bulk of Hanford's shipments of recycled UO_3 powder to the GDPs contained only trace amounts (<10 ppb) of Pu. Based on the limited amount of historical documentation located, it still appears the preponderance of Hanford recycled UO_3 powder contained $\leq$5 ppb. Six shipments were identified in Hanford documents as containing about 12, 13, 16, 19, 22, and 30 ppb average, respectively; however, records indicate receiver sites were typically notified of these out-of-specification conditions and accepted these materials prior to their shipment from Hanford. These shipments represented **~193** MTU containing ~3.4 g Pu. Information from Oak Ridge indicates that their site received four shipments from Hanford for which their analyses indicated Pu concentrations of 13, 17, 17, and 28 ppb. These shipments are in addition to the shipment containing 22 ppb mentioned above. Hanford analyses on these four shipments indicate the Pu concentrations were all <10 ppb. Available documentation indicates that Hanford and Oak Ridge both acknowledged this discrepancy. This is further discussed in Section 6.4.3. These four shipments totaled **~123** MTU and contained ~2.3 gPu. The limited analytical data located is insufficient for RL to determine the total quantity of plutonium shipped to the various GDPs; however, it appears that approximately **110** to **550** grams of plutonium were shipped with the **109,792** MTU, based upon an assumed average plutonium concentration range of one to five ppb. The mean of this range of Pu concentrations is 330 grams of Pu.

Hanford did not routinely analyze the incoming uranium metal for radioactive constituents, but relied on the shipper's guidelines for the metal product. It appears, based on the information available, that metal received prior to 1963 had been processed through the Paducah GDP and contained on the order of 10 parts or less plutonium per trillion parts uranium. Metal that was received in 1963 and beyond had been directly blended at Fernald and contained <10 ppb Pu.

6.2.2 Neptunium and Technetium

Hanford did not routinely analyze the UO_3 product for neptunium or technetium before 1980, as there were no related product specification requirements. For Hanford recycled UO_3 powder, technetium and neptunium limits were not considered required. Until 1967, Hanford analyzed the recycled UO_3 powder for total beta and gamma activity and conformed to the required specification levels of less than 100% of the beta activity of aged natural uranium and less than 300% of the gamma activity of aged natural uranium, respectively. In 1967, the beta and gamma measurements were dropped in favor of specific isotopic measurements (^{95}Zr/Nb, ^{103}Ru, and ^{106}RuRh) as

discussed in Section 4.3.2. Information provided by Fernald indicates that metal produced from recycled uranium contained ^{99}Tc ranging from 3 - 12 ppm and ^{237}Np with an upper bound of 500 ppb.

No uranium processing activities at Hanford have been identified that would increase the plutonium component of the recycled uranium after it was separated in the reprocessing plants. In the course of this study, no documentation has been found that reports concentration of constituents in areas accessed during maintenance activities. However, time has not been available to perform a detailed assessment of all uranium processing steps utilized at Hanford to assure that impurities in uranium were never concentrated. The UO_3 Plant calciners, which converted the recovered UNH to UO_3 powder did operate at elevated temperatures in an oxidizing environment. It is reasonable to believe that any volatile fission products which may have been present in the recycled uranium could have been released through the off-gas system, plated out on equipment surfaces, accumulated in off-gas scrubber solutions, or have been released to the environment. Documentation has been found to indicate that some of the ruthenium volatilized during UNH calcination, with decontamination factors (DF) ranging from <1 to 6 (see Appendix G, Section G.5). No operations in the fuel fabrication processes have been identified which would be expected to have further concentrated the constituents, other than the burning of metal fines to uranium trioxide.

6.3 Activity Assessment and Occupational Potential Exposure

The DOE-HQ mass balance project is a Department-wide effort to review each site that was involved with recycled uranium to provide an estimate of specific activities involving recycled uranium, and to develop a preliminary estimate of the approximate number of employees whose work subjected them to potential exposure from the constituents in the recycled uranium. The estimate of occupational potential exposure (OPE) is based on guidance developed during a workshop meeting held at the ORNL in May, 2000. This OPE criteria is listed at the bottom of Table 6-1. The recycled uranium activity and OPE assessment for Hanford is provided below.

The operations, maintenance and waste handling operations of the facilities described in Section 2 contributed to some personnel exposure and environmental releases. However, distinguishing any such personnel exposures to trace quantities of transuranics and fission products in recycled uranium from those associated with other Hanford operations which involved the handling and processing of significant quantities of irradiated fuel, high-level waste, and plutonium, would be very difficult. Assessment of personnel uranium exposure is further complicated by the practice of transferring personnel between facilities to meet ongoing work needs. The facilities at Hanford that had the highest potential for uranium exposure were the fuel fabrication facilities where large amounts of uranium metal and scrap were handled, and the UO_3 Plant, which handled large quantities of dry UO_3 powder. Other facilities involved with the handling of separated recycled uranium (as UNH) and of any waste from recycled uranium processing also had the potential for contributing to some exposure.

Table 6-1 Assessment of Activities at Hanford Where Workers Were Most Likely to Contact Recycled Uranium

Location	Interval (s)	Processing Steps/Work Performed	Constituents of Interest[1]	Constituent Levels[1]	Airborne Potential[2]	Exposure Duration[3]	OPE[4]
U Plant	1952-1958	Reference – Hanford Mass Balance Report Figure 2-4, beginning at "Uranium Stripping column.					
		• Concentrator operation	Typically Pu = 1-5 ppb Np = 20-500 ppb Tc = 3-12 ppm	1	0	3	0
		• Acid handling		1	1	2	2
		• Sampling		1	1	2	2
		• Maintenance in uranium-handling areas		1	1	2	2
		• All assays and waste handling and disposition		1	1	2	2
		• UNH transfer to UO_3 Plant (pumping)		1	2	1	2
		• UNH transport to 321 Bldg. (truck)		1	1	1	1
		• Recycle and rework off-spec material.		1	1	1	1
REDOX	1952-1967	Reference-Hanford Mass Balance Report Figure 2-9. Beginning at "U-Stripping column.					
		• Concentrator operation	Typically Pu = 1-5 ppb Np = 20-500 ppb Tc = 3-12 ppm	1	0	3	0
		• Acid handling		1	1	2	2
		• Sampling		1	1	1	1
		• Maintenance in uranium-handling areas		1	1	2	2
		• All assays and waste handling and disposition		1	1	2	2
		• UNH transfer to UO_3 Plant (pumping)		1	1	2	2
		• Recycle and rework off-spec material.		1	1	1	1
		• Acid Receipt and handling.		1	1	1	1
PUREX	1952-1972 1982-1990	Reference-Hanford Mass Balance Report Figure 2-10 beginning at final uranium cycle					
		• Extraction column operation	Typically Pu = 1-5 ppb Np = 20-500 ppb Tc = 3-12 ppm	1	0	3	0
		• Concentrator operation		1	0	3	0
		• Sampling and assay		1	1	1	1
		• Maintenance in uranium-handling areas		1	1	2	2
		• Waste handling and disposition		1	1	2	2
		• UNH transport (truck) to UO_3 Plant		1	2	2	4
		• Recycle and rework off-spec material		1	1	1	1
		• Receipt and off-load of reclaimed nitric acid		1	1	2	2
UO_3 Plant	1952-1972 1984-1994 (Inter-mittent)	Reference-Hanford Mass Balance Report Figures 2-5 and 2-6. All processing and handling activities including:					
		• UNH receipt (truck and pipeline)	Typically Pu = 1-5 ppb Np = 20-500 ppb Tc = 3-12 ppm	1	2	2	4
		• Concentration		1	1	3	3
		• Calcination (batch & continuous)		1	3	3	9
		• Product handling, size reduction, & loadout		1	3	3	9
		• Product storage and shipping		1	1	3	3
		• Acid recovery, storage, shipping (rail)		1	1	2	2
		• Sampling and assay		1	2	2	4
		• Waste collection, handling, disposition		1	2	2	4
		• Recycle, blending of off-spec materials		1	2	1	2
		• All equipment and facility repairs		1	3	2	6
		• Development of continuous calciners		1	3	3	9

Table 6-1 Assessment of Activities at Hanford Where Workers Were Most Likely to Contact Recycled Uranium (Cont.)

Location	Interval (s)	Processing Steps/Work Performed	Constituents of Interest	Constituent Levels[1]	Airborne Potential[2]	Exposure Duration[3]	OPE[4]
Fuel Fab 313-314 Building	1952-1971	Reference-Hanford Mass Balance Report Figure 2-13.					
		• Metal melting, pickling, and acid etch	Typically* Pu = 0.01- 6 ppb Np = 3 - 10 ppb Tc = 0.01 - 6 ppm	0	3	3	0
		• Foundry and metal forming		0	3	3	0
		• Cladding application		0	2	3	0
		• Scrap		0	3	2	0
		• Waste handling and disposal		0	1	2	0
Fuel Fab (N Fuel) 333 Building	**1963-1987**	**Reference-Hanford Mass Balance Report Figure 2-14.**					
		• Metal pickling, and acid etch	Typically* Pu = 0.01- 6 ppb Np = 3 - 10 ppb Tc = 0.01 - 6 ppm	0	3	2	0
		• Metal forming, machining, and welding		0	3	3	0
		• Cladding application		0	2	3	0
		• Scrap		0	3	2	0
		• Waste handling and disposal		0	1	2	0
Solar Pond 183-H Basin	**1975-1985**	• Solar Pond receipt of waste from 300 Area; evaporation of those liquids		0	1	1	0
		• Removal of residues and interment at Environmental Restoration Disposal Facility.		0	2	2	0

Numbers of people at "No Significant Exposure Potential"		Numbers of people at "Moderate Occupational Exposure Potential"	
300 Area AL-CLAD Fuel	350	**U-Plant**	12
300 Area ZR-Clad Fuel	300	**REDOX**	36
183-H Solar Basin	20	**PUREX**	64
		UO_3 Plant	344
	670		**456**

(See Figure 6-1 for staffing estimate assumptions)

Criteria for Occupational Potential Exposure (Potential exposure criteria developed during Uranium Mass Balance Workshop, 5/2000)

Factors

1) Constituent Level – Typical constituent concentrations in material being handled
- 0 – Sum of constituents clearly below de minimis levels (clearly less than 10% added dose)
- 1 – Sum of constituents likely to cause up to 20% total dose
- 2 – Sum of constituents likely to cause more than 20% but less than 50% total dose
- 3 – Sum of constituents likely to cause 50% >more of total dose

2) Airborne Potential – Likelihood to be airborne or Potential concentration in air
- 0 – No likelihood of being airborne
- 1 – Low airborne potential
- 2 – Moderate airborne potential
- 3 – *High airborne potential*

*See Section 4.8.4

3) Exposure Duration – Time of worker exposure on job
- 1 – 50 hours / year or less
- 2 – More than 50 hrs / year but less than 500 hours
- 3 – 500 or more hours per year

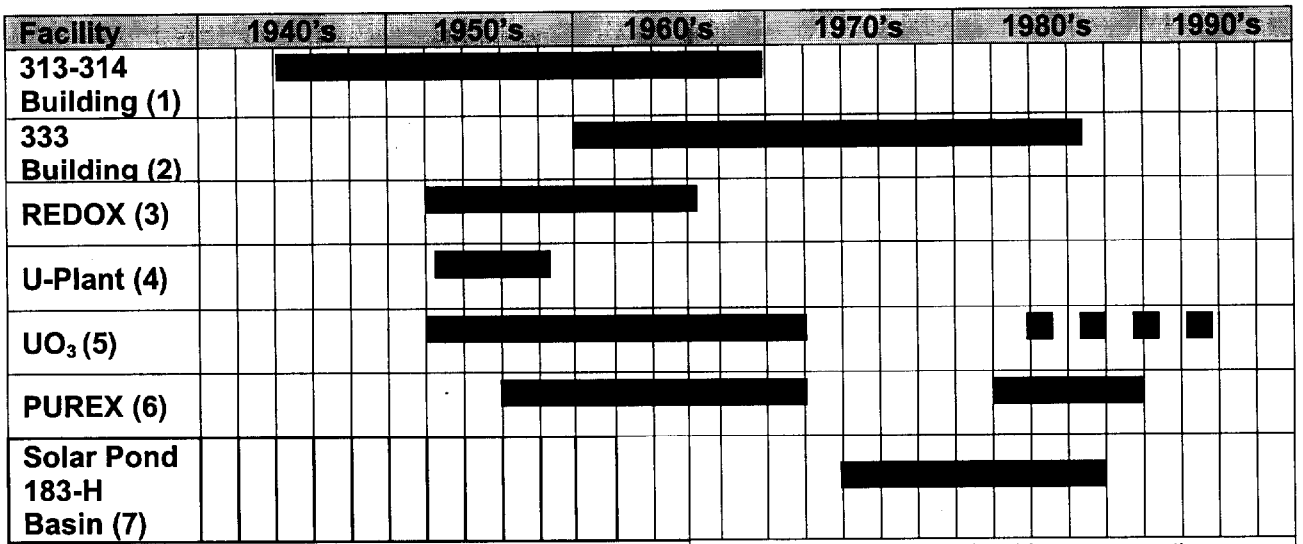
4) Occupational Potential Exposure (OPE) =
Constituent Level X Airborne Potential X Exposure Duration
- 0 – No significant Occupational Exposure Potential
- 1 – Low Occupational Exposure Potential
- 2 – 2-9 = Moderate Occupational Exposure Potential
- 3 - >10 = High Occupational Exposure Potential

Section 6
Discussion and Conclusions

Time was insufficient to complete an in-depth activity and potential worker exposure assessment. However, a cursory estimate, utilizing assumptions of direct labor and maintenance staffing at the facilities of interest, was completed to provide an overview of recycled uranium work activities in these facilities and an OPE estimate.
The facilities that were identified to have involved the handling of recycled uranium included: (1) the 313-314 Building complex in the 300 Area which was primarily involved with the fabrication of aluminum-clad reactor fuels, (2) the 333 Building complex in the 300 Area which was involved in the fabrication of zirconium-clad reactor fuels, (3) the REDOX Chemical Separations Plant in the 200 West area where reactor fuels were dissolved for plutonium and uranium recovery (including transfer of the UNH to the UO_3 Plant for calcination), (4) the U-Plant in the 200 West area where pre-1952 tank wastes were processed for uranium recovery, (5) the UO_3 Plant in the 200 West area where uranium recovered as UNH was received, concentrated, calcined, and packaged for shipment and recycle, (6) the PUREX Plant in the 200 East area where irradiated fuels were dissolved for separation of plutonium and uranium (with the UNH being shipped by truck to the UO_3 Plant for calcination), and (7) the 183-H solar basin in the 100-H area that was used for a ten-year interval to evaporate dilute liquid wastes generated at the 300 area fuel fabrication plants.

The interval of operation for each of these facilities is illustrated in Figure 6-1, along with an estimate of probable direct labor staffing support. The staffing calculations were based upon estimates of the number of operators and craft personnel per shift, the number of shifts per week, and an estimate of the average length of time that an employee would work at the facility. This information was used to estimate the total number of people who worked at each facility during its operating lifetime. These staffing totals, by facility, were then used as inputs to Table 6-1 to allow an estimate of the number of people likely to have been exposed to various levels of constituents of recycled uranium. Below is a typical staffing estimate calculation for Figure 6-1:

> As an example, consider the 313-314 Building complex which had a PQ operations shift schedule and operated for 25 Years. It is estimated that the following operational information would apply to these facilities:
>
> 25 Operators/Shift; 10 Crafts/Shift; and an average Employee Time at Facility of ~5 years.
>
> Thus, the number of workers on any given shift that may have been in contact with uranium would be 25 fuel fabrication operators plus 10 maintenance craftsmen for a total of 35. The "PQ" shift arrangement provided for Monday through Friday coverage of the 8AM to 4PM shift and the 4PM to Midnight shift. If the average worker remained at the job for 5 years then,
>
> $$(35 \text{ workers/shift}) \times (2 \text{ shifts/day}) \times \frac{\text{25 years facility operation}}{\text{5 years at that facility for a typical worker}}$$
>
> = 350 involved employees at that facility over the lifetime of the facility.



1) **313-314 Building** PQ shift 25 years operation
25 operators/shift
10 crafts/shift
average time at facility - 5 years
(25 + 10) x 2 x 25/5 = 350 personnel

2) **333 Building** PQ shift – 25 years operation
20 operators/shift
10 crafts/shift
average time at facility – 5 years
(20 + 10) x 2 x 25/5 = 300 personnel

3) **REDOX ABCD shift** – 15 years operation
Assume 3 employees/shift worked with recycled uranium
average time at facility – 5 years
3 x 4 x 15/5 = 36 personnel

4) **U Plant** ABCD shift – 4 years operation
Assume 3 employees/shift worked with recycled uranium
average time at facility – 4 years
3x 4 x 4/4 = 12 personnel

5) **UO_3 Plant ABCD shift** – 20 years operation
12 operators/shift
8 crafts/shift
average time at facility - 5 years
(12 + 8) x 4 x 20/5 = 320 personnel
Calciner Development ABCD shift – 1 year operation
4 techs/shift
2 crafts/shift
average time at facility – 1 year
(4 + 2) x 4 = 24 personnel

6) **PUREX ABCD shift** – 20 years operation
Assume 4 employees/shift worked with recycled uranium
average time at facility - 5 years
4 x 4 x 20/5 = 64 personnel

7) **183-H Basin** ABCD shift – 10 years operation
Assume 1 employee/shift worked this task
average time at facility – 2 years
1 x 4 x 10/2 – 20 personnel

Note: Shift definitions = ABCD is three 8 hour shifts/day, 7 days/week
PQ is two 8 hour shifts/day, 5 days/week

Figure 6-1 Estimate of Personnel Having Work Potentially Involving Exposure to Recycled Uranium at Specific Facilities

Specific recycled uranium activities where worker contact with recycled uranium may have occurred were identified from plant description and process flow information which has been provided in Section 2 of this report. Estimated ranges of constituent concentrations in the recycled uranium were identified for each facility, based upon analytical information provided in Section 4. Criteria for estimating the OPE for each activity were then used to assess the exposure potential for each activity. The summary assessments for each activity in the facilities were then applied to the staffing estimates

shown in Figure 6-1 to arrive at an estimate of the number of personnel who could have had some level of OPE. All of this information is presented in Table 6-1.

The data summarized in Table 6-1 indicates that a total of about **1,126** people are estimated to have worked in areas directly involving recycle uranium during the operational time of the Hanford Site. Using the given OPE assessment criteria, of the **~1,126** staff personnel, it is estimated that **~670** had "no significant exposure potential" while **~456** had "moderate occupational exposure potential." It should be noted that this is only a rough estimate based upon limited data, engineering judgements and assumptions, and the application of some broad general OPE criteria which were established specifically for the mass balance project.

Since 1946, Hanford has had formal personnel monitoring programs in place which are designed to identify uptake of radioactive materials by personnel. Any employee who was assigned to a work location where contact with radioactive materials was judged to be possible was required to participate in the bioassay program. Each decision/request for bioassay evaluation on an individual or group of people represented a rather conservative contemporary judgement for protection of employees from radioactivity. Not all of those who were placed on the uranium bioassay program were expected to have direct contact with the radioactive material. It is therefore reasonable to suggest that the number of employees sampled on the uranium-specific bioassay program represents a conservative and upper bounding estimate of the number of individual workers potentially exposed to recycled uranium at Hanford.

Site records show that more than 50,000 employees were hired at Hanford over the operating period of the installation. In contrast, since the sampling program was established in 1946, only ~4,200 Hanford employees have been subjected to uranium-specific bioassay sampling and evaluation. A preliminary review of the records of that program was conducted. Those records (which included estimates for the constituents of interest) identified no significant doses associated with the recycled uranium for any of the Hanford Uranium Bioassay program participants. Due to the eligibility criteria applied, it is considered highly improbable that any substantial number of un-assayed site employees could have had any significant uranium uptake. For this study on recycled uranium traffic at Hanford, the very brief and somewhat empirical staffing and timing models for the facilities that handled large quantities of recycled uranium suggests that ~1,126 of the 4,200 employees did in fact perform substantial duty in the facilities that included opportunities for rather close contact with recycled uranium. Of that number, perhaps ~456 individuals had "moderate occupation exposure potential" as assessed and defined in the criteria that was given.

For a more specific analysis of worker contact with recycled uranium at Hanford than that contained in this report, an examination of Hanford's uranium bioassay records, a more detailed review of plant-by-plant operations, abnormal events, maintenance, and facility upgrades, overlaid by staffing models and production intervals would be required. Corroboration of such an analysis by examination of various records and

interviews of retired employees would be necessary, in order to validate the OPE estimates.

6.4 Data Validation

6.4.1 Recycled Uranium Shipped from Hanford

External validation of Hanford shipment and receipt quantities of recycled uranium were performed, to the extent possible, during and after the May 17, 2000 workshop previously described in Section 6.1.5. For those sites identified as Major Tier 1 sites (Paducah, Fernald, and Oak Ridge) in which comparison was possible, the percentage variances between Hanford and other site data was below 1% (0.02-0.7%) for transaction categories detailed in Section 6.1.5.

6.4.2 Impurities Shipped with UO_3 from Hanford

As shown in Section 4, fairly complete analytical data for Pu contained in shipments of UO_3 have been located for the years from 1963 to 1988. Data on shipments prior to 1963 are limited for the constituents of concern including Pu, ^{237}Np, or ^{99}Tc. Hanford data for the ^{237}Np and ^{99}Tc were not available on a routine basis until 1984. In an attempt to reconstruct the quantity of constituents that were present in Hanford produced UO_3 , source documents from other sites were used to make the estimates, i.e. Smith 1984 and Ritter K/ETO-30 and draft U Mass Balance reports of receipts from the Major Tier 1 receiver sites. The Smith and Ritter documents report the result of special studies performed at Paducah using Hanford UO_3 as a starting material. Analyses were completed at these sites on hundreds of samples between 1959 to 1973 for Pu, ^{237}Np, and ^{99}Tc.

Engineering estimates of the quantities of constituents (Pu, Np, and Tc) which were present in recycled uranium received, shipped, contained in waste, released to the environment, or contained in the current recycled uranium inventory at Hanford have been made. The detailed results of these estimates are provided in Appendix I of this report. Analytical data on the concentration of Np and Tc in Hanford recycled uranium is minimal, since there were no specification requirements for these elements. Reasonable analytical data has been identified which indicates that recycled uranium shipped from Hanford typically contained Pu in the range of 1 to 5 ppb, and limited analytical data indicate Np ranged from 20 to 500 ppb, and Tc ranged from 3 to 12 ppm.

Since it is not possible to provide a complete historical assessment of the constituent levels in all recycled uranium Hanford, rough estimates of annual quantities of constituents in recycled uranium shipped each year were developed by assuming that the constituent levels during the years when analytical data is not available were the same as those for the years when analytical data is available. The annual estimates provided were based upon the low, mean, and high values for the constituent ranges mentioned above. Summaries of the total amount of constituents sent to Paducah,

Fernald, and Oak Ridge sites are shown in Figures 6-2, 6-3, and 6-4. The **~110,000** MT of recycled uranium shipped from Hanford is estimated to have contained from **~110** to **~550** g Pu, **~2.2** to **~28** Kg Np, and **~330** to **~1,318** Kg of Tc. In general, these estimated constituent quantities appear to be consistent with data from sites receiving Hanford recycled uranium.

6.4.3 Discussion of Pu Data Differences Between K-25 and Hanford

In the K-25 Uranium Mass Balance Preliminary Site Report, analytical data are presented that indicate the Pu concentrations of UO_3 reported by K-25 between January and April 1953 are significantly outside the 10 ppb specification threshold. Hanford and Oak Ridge agree that data generated before and after the January to April 1953 time period are generally well within the 10 ppb threshold. Analysis of source documents are not available from Hanford for shipment composites or lots during this time period. However, monthly and weekly reports for this time period do not indicate that UO_3 shipments to Oak Ridge or Harshaw were made of out-of-specification UO_3 material in the Pu constituent. A reference [Schmidt 1953] recognizes that there was some discrepancy in the plutonium analytical results between Oak Ridge and Hanford. This reference also indicates other constituent data are in agreement. Communications were ongoing at that time to resolve the differences, specifically on shipments 18, 19, and 20. This reference states that Hanford reported less than 10 ppb and Oak Ridge reported concentrations in the range of 15 to 20 ppb on those three shipments. Both laboratories reported 22 ppb on the shipment composite number 43.

In 1952, Hanford was analyzing each lot of material but reduced the analytical workload in November 1952 by only analyzing the carload composite for Pu. The K-25 report apparently includes measurements of each lot. A lot represented eight drums, each drum contained about **300** Kg of UO_3, and a car shipment included 11 or 12 lots (about **31-32** MTU) but ranged from 5 to 12 lots. Hanford transfer records show **411** MTU of UO_3 and metal scrap were shipped to Oak Ridge during the January to June 1953 time period. On the graphic in the K-25 draft report, approximately 45 –50 data points are shown during the January to April 1953 time frame. Assuming each point represents the results of one lot reported, this indicates four shipments were made and would have contained about **120** to **150** MTU. This is consistent with the weekly and monthly reports for the time period. This represents about three percent of the uranium sent to Oak Ridge. During the same time period Hanford shipped approximately four times more material to Harshaw Chemical Co. than to Oak Ridge. The Harshaw product was shipped on to Oak Ridge after processing to remove non-radionuclide constituents. The Oak Ridge report indicates the Harshaw material met the Pu specification.

The reference, cited above, indicates that Oak Ridge was using an analytical procedure that was not included in the authorized procedure manual of that time. They added an aluminum nitrate "salting agent" that could enhance the extraction of Pu into the organic extractant. Hanford was using the procedure in the manual and this could account for the discrepancy in the constituent concentration. It is evident that the Hanford results

Plutonium Contained In 110,000 MT Recycled Uranium From 1952- March 1999 As Trace Impurity, Typically 1 - 5 ppb

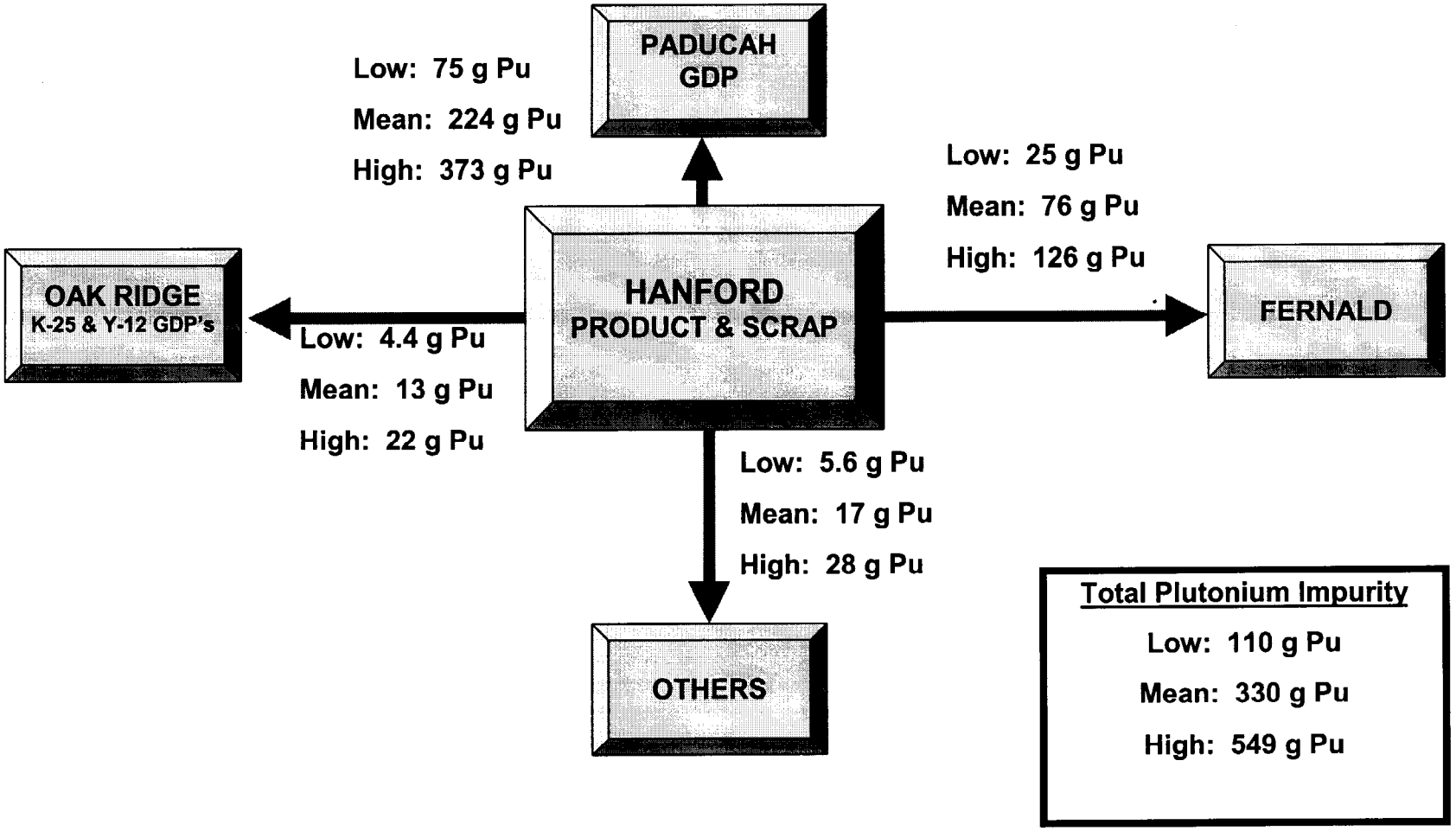


Figure 6-2 Plutonium Contained in Recycled Uranium Shipped From Hanford

Neptunium Contained In 110,000 MT Recycled Uranium From 1952- March 1999 As Trace Impurity, Typically 20 - 500 ppb

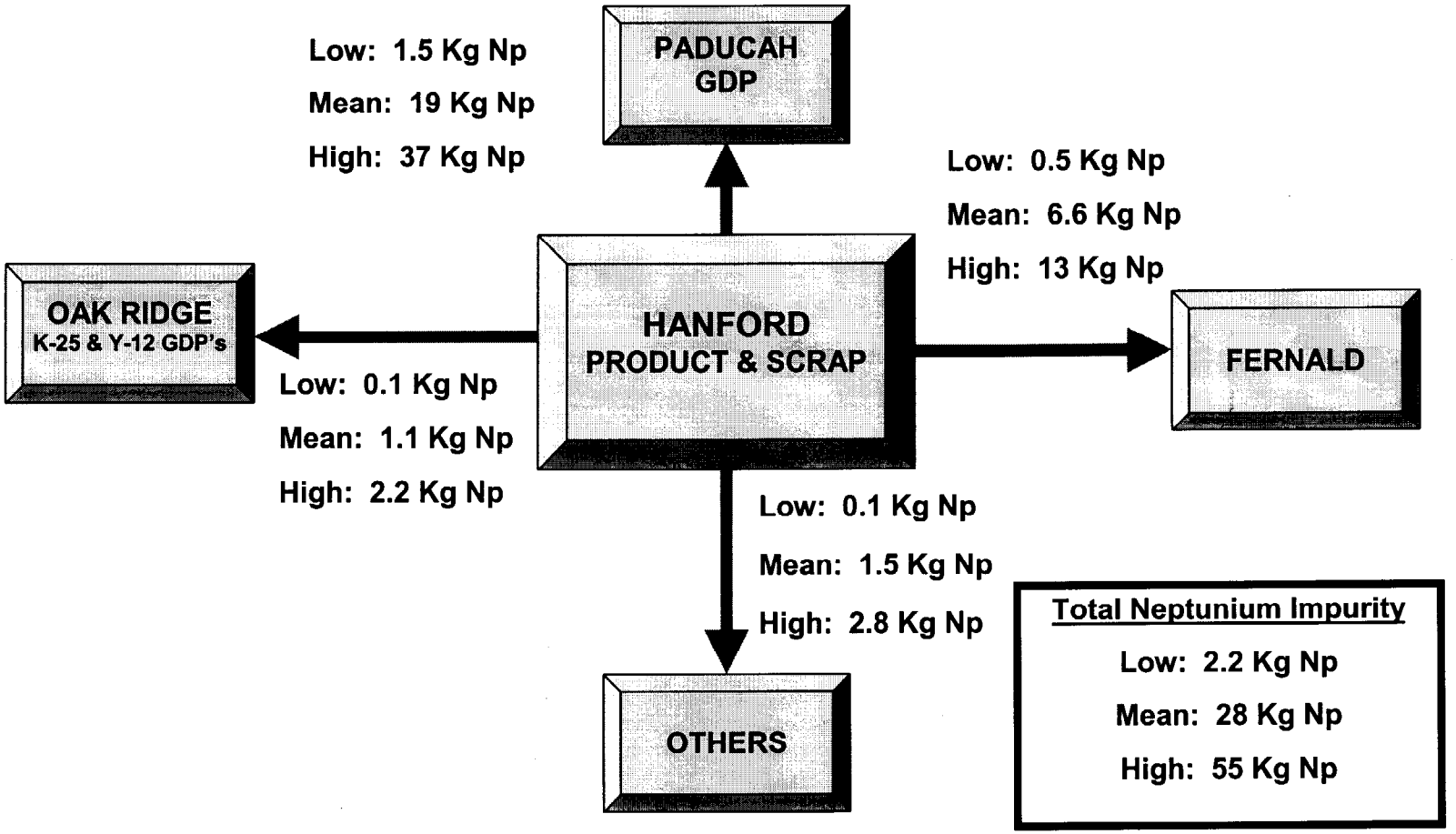


Figure 6-3 Neptunium Contained in Recycled Uranium Shipped From Hanford

Technetium Contained In 110,000 MT Recycled Uranium From 1952- March 1999 As Trace Impurity, Typically 3 - 12 ppm

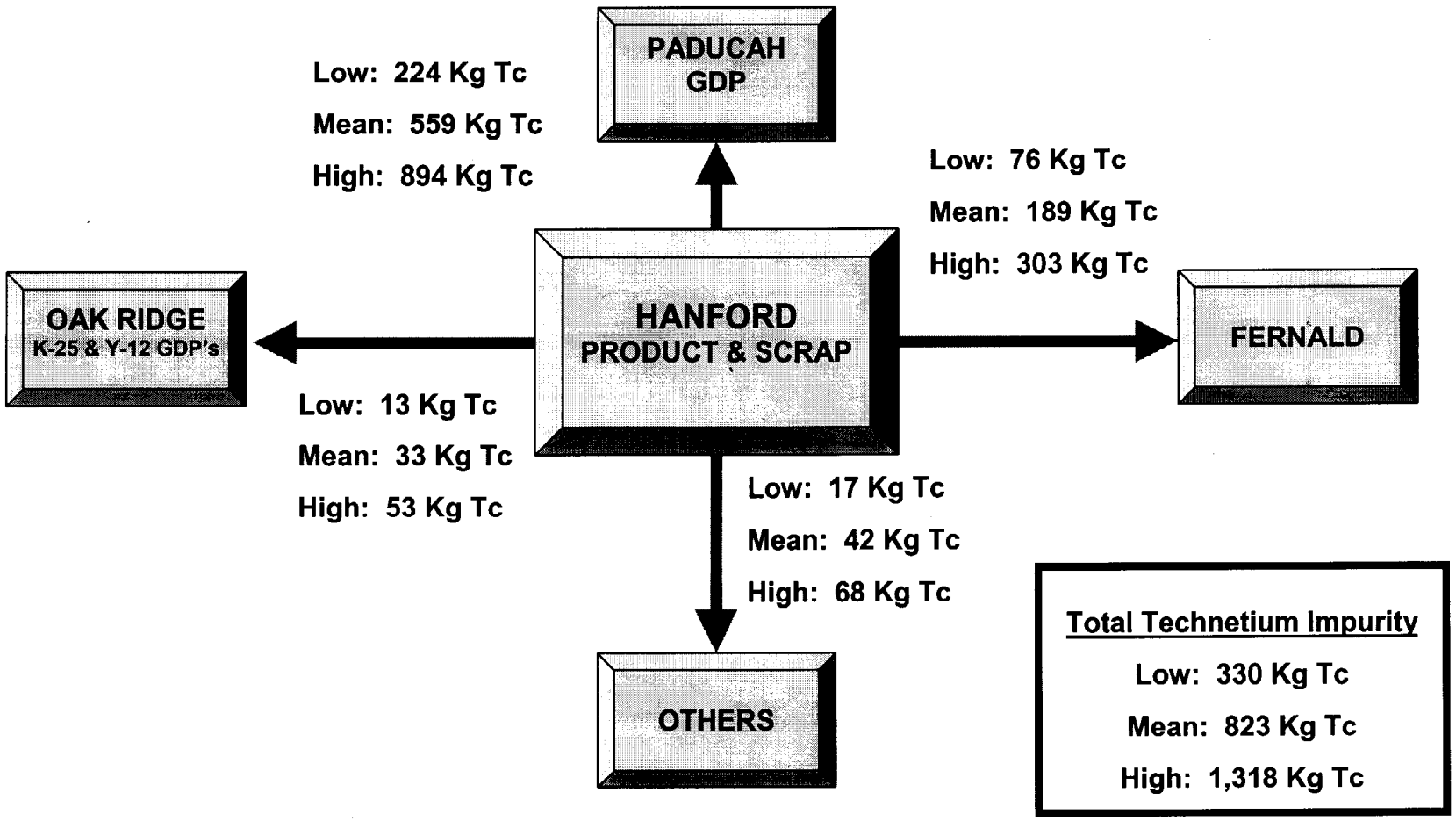


Figure 6-4 Technetium Contained in Recycled Uranium Shipped From Hanford

are biased low compared to the K-25 results. It is interesting to note that during the same time period that Oak Ridge reported receiving high Pu concentrations from Hanford, they also reported receiving slightly higher Pu levels from Harshaw than in preceding or succeeding periods. It seems that there would be a delay of a month or more in the receipt at Oak Ridge of Hanford produced UO_3 while processing was performed at Harshaw. The potential of the Harshaw process to reduce the Pu concentration in UO_3 product from Hanford is not known.

If Hanford data (less than 10 ppb for all shipments except shipment number 43 which Hanford reported as being 22 ppb) are used, about 4.5 grams of Pu would have been included with the UO_3 shipped during the January – April 1953 time period. If Oak Ridge's example data are extrapolated to the material shipped during this period and the average is 22 ppb, the Pu included with the UO_3 is 9 grams. This represents a difference of 4.5 grams of Pu in approximately **120** to **150** MTU of uranium.

In 1948 and again in 1951 the concentration of Pu allowable in recycled UO_3 was established to limit the increase in potential hazard to personnel to no more than 10 percent of the hazard present from handling aged natural uranium in equilibrium with its decay daughters [Gamertsfelder 1948 and 1951]. The threshold established for Pu was 10 ppb and included a large conservatism factor. In 1985, a task force re-evaluated the 10 ppb limit against the then current air concentration guides and concluded that the 10 ppb value represented 3.5 percent of the concentration guide [DOE/OR-859 1985]. Therefore, with respect to personnel hazards from Pu impurities in recycled uranium, even the Hanford shipments which were above the specification of 10 ppb were well within the concentration guidance as established both in 1951 and again when the guidance was re-evaluated in 1985.

Section 6
Discussion and Conclusions

References:

DOE/OR-859 1985
DOE/OR-859, *The Report of the Joint Task Force on Uranium Recycle Materials Processing,* Task Force Members Dick Egli et. al., September 1985

Gamertsfelder 1948
HW-10515 by C.C. Gamertsfelder to C.N. Gross, *Plutonium and Fission Product Contamination in Reclaimed Uranium,* July 19, 1948

Gamertsfelder 1951
Letter HW-20628 (HAN-36668) from C.C. Gamertsfelder to R.H. Beaton, *Recent Changes Affecting the Specifications Concerning Activity Levels in Reclaimed Uranium,* March 27, 1951

Hauff 1952
Letter T. W. Hauff to Donald G. Sturges, *General Analytical Control Program I,* November 5, 1952

Smith 1953
HW-27990, *Summary of Oak Ridge Discussions Relative to Hanford Uranium Trioxide Specification,* R. E. Smith, May 5, 1953

Smith 1984
Report KY/L-1239, R. F. Smith, *Historical Impact of Reactor Tails on the Paducah Cascade, Paducah Gaseous Diffusion Plant*, March 1984

Schmidt 1953
Letter HW-27314, H. R. Schmidt to L. M. Knights, *Determination of Pu in* UO_3 , March 5, 1953