PROJECT TASK 3

Health Studies on
ROCKY FLATS

PHASE II:
HISTORICAL
PUBLIC
EXPOSURES

Colorado Department
of Public Health
and Environment

FINAL REPORT

Results of Screening Calculations to Assess the Relative Importance of Rocky Flats Uranium Releases

August 1999

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"Setting the standard in environmental health"

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Part of Task 3: Independent Analysis of Exposure, Dose, and Health Risk to Offsite Individuals

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Submitted to the Colorado Department of Public Health and Environment Disease Control and Environmental Epidemiology Division Rocky Flats Health Studies in Partial Fulfillment of Contract No. 100APPRCODE 391

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Part of Task 3: Independent Analysis of Exposure, Dose, and Health Risk to Offsite Individuals

August 1999

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Submitted to the Colorado Department of Public Health and Environment Disease Control and Environmental Epidemiology Division Rocky Flats Health Studies in Partial Fulfillment of Contract No. 100APRCODE 391

SUMMARY

In response to public concern, screening level estimates have been made of the consequences of releases of enriched and depleted uranium at Rocky Flats. Routine and accidental uranium releases from the Rocky Flats Plant were identified in Phase I of the Historical Public Exposures Studies on Rocky Flats. However, those releases have not been studied in detail during Phase II. The principal hazard from enriched uranium is due to radiation exposure. In contrast, the main concern about depleted uranium is its chemical toxicity.

Radiation doses from uranium released to the atmosphere have been compared with those from airborne releases of plutonium. Plutonium is the primary source of radiation exposure from Rocky Flats and the largest airborne releases have been studied in detail in Phase II. For chemical toxicity, guide values for concentrations of depleted uranium in air and water have been estimated using a cautious estimate of the toxicity threshold for uranium. That recently published threshold, 0.1 $\mu g\ U\ g^{-1}$ in the kidney, is 30 times lower than that traditionally used for workers.

None of the comparisons made suggests that uranium releases deserve much further attention. Releases of enriched uranium have been estimated to pose substantially less risk than the plutonium releases that have been the focus of Phase II. Radiological risks from depleted uranium are also small with respect to those from plutonium. The potential for toxic effects on the kidney because of intake of depleted uranium also does not appear to be large. Measured uranium concentrations in raw water are well below the derived guide value and treatment of drinking water befpre distribution could have reduced those levels. Very large releases of depleted uranium to the atmosphere in a short time under adverse meteorological conditions could have resulted in a transient elevation of uranium levels in kidneys of exposed persons. Such exposures are not likely to cause kidney damage.

Although the amount of material unaccounted for (MUF) is not a reliable guide to quantities of uranium released to air and water, it does provide a gross upper bound that may be useful for some types of calculations. The amount of MUF for enriched uranium at Rocky Flats, about 350 kg, was documented in the mid-1970s after enriched uranium operations were shut down and the facility was cleaned out. In the absence of similar documentation for depleted uranium, a speculative estimate of the MUF for depleted uranium was made. The amount is large, about 190,000 kg, and quite uncertain because of the lack of information. However, it is not unreasonable estimate considering the amount (about 200,000 kg) thought to have been shipped to Idaho for burial. Because methods for measuring the amounts of uranium in solid wastes were not reliable, it is quite conceivable that the amounts of Rocky Flats uranium identified as unaccounted for were buried.

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RESULTS OF SCREENING CALCULATIONS TO ASSESS THE RELATIVE IMPORTANCE OF ROCKY FLATS URANIUM RELEASES

1. INTRODUCTION

Members of the public have expressed concern that exposures to uranium released from the Rocky Flats Plant have not received adequate attention. Several types of planned and unplanned releases are the source of concern. Both enriched and depleted uranium were released to the atmosphere from stacks and vents that served various processing facilities. Open outdoor burning was used to oxidize, and stabilize against spontaneous combustion, pyrophoric metal chips of depleted uranium. A wooden pallet containing 60 kg of depleted uranium sheets was burned accidentally. Both intentional and accidental burning of uranium led to releases of some of the uranium to the atmosphere. Uranium was also released in liquid wastes to surface water streams that entered reservoirs from which drinking water was obtained. Uranium in solid wastes improperly disposed near or into surface water drainage areas was also susceptible to leaching into streams and subsequent transport to the reservoirs.

In Phase II of the dose reconstruction studies, releases of plutonium to the atmosphere have received the greatest attention, although exposures to chemicals and other radionuclides, such as tritium (^3H), have also been addressed. Evaluations of uranium releases were performed in Phase I, but uranium releases have not been a focus of dose reconstruction in Phase II. One way to assess the relative importance of the Rocky Flats uranium releases is to compare screening estimates of their consequences with those of the plutonium releases that were studied in detail.

Section 2 describes categories of uranium that span the range of enrichments that were released from Rocky Flats. For releases to the atmosphere, the radiotoxicity of uranium at these enrichment levels is compared with that of plutonium, the most important airborne radionuclide released from Rocky Flats. Section 3 contains the dose coefficients used to compare potential impacts of releases of different masses of enriched and depleted uranium with plutonium releases from the facility. Airborne uranium releases at Rocky Flats are discussed in Section 4. Section 4 also contains comparisons of the uranium releases with estimated plutonium releases from focused Phase II studies of routine operations and accidents.

The chemical toxicity of uranium, discussed in Section 5, is a concern primarily for depleted and natural uranium. Data on uranium in liquid wastes and measurements of reservoir concentrations are presented in Section 6. Comparisons are made with a proposed guide for uranium toxicity that is 30 times lower than the historical guide for workers. Section 7 discusses uranium accounting at Rocky Flats and includes estimates of the material unaccounted for (MUF) for both highly enriched and depleted uranium.

Section 8 summarizes the comparisons that have been made and conclusions that have been drawn from them. References are provided in Section 9.

2. COMPOSITION OF NATURAL, ENRICHED, AND DEPLETED URANIUM

Uranium found in nature is composed of three isotopes, all of which are radioactive. Each of the isotopes (^{238}U, ^{235}U, and ^{234}U) is an alpha-emitter and all are long-lived. The half-lives of ^{238}U and ^{235}U are longest, about 4.5 and 0.7 billion years, respectively. By comparison, the half-life of ^{234}U is rather short, only 244 thousand years. Because its half-life is shorter, ^{234}U has a higher specific activity than ^{238}U and ^{235}U. That is, 1 gram (g) of ^{234}U emits alpha particles at a much higher rate than 1 g of either of the other isotopes.

Table 1 shows the properties of three levels of uranium enrichment. The categories are defined by the fraction of ^{235}U, the primary fissionable isotope, present. Natural uranium contains 99.275% ^{238}U, 0.72% ^{235}U, and 0.0054% ^{234}U. Production of nuclear weapons required enriched uranium, that is, uranium whose ^{235}U content had been increased. Production of uranium enriched in ^{235}U simultaneously produced uranium whose ^{235}U content was reduced or depleted. Uranium containing more than 0.72% by weight is described as enriched, but very high enrichments were used for weapons production. Enrichment procedures were designed to rely on the difference in mass between ^{235}U and ^{238}U to separate the two isotopes. The enrichment process also increased the fraction of ^{234}U in enriched uranium and reduced the fraction of ^{234}U in depleted uranium. This is illustrated in Table 1, which shows the fractional contributions of each isotope to the total alpha activity. The specific activity of each uranium enrichment category is listed in the last column.

Table 1. Some Properties of Natural, Enriched, and Depleted Uranium

Type of uranium	Weight percent of ^{235}U	Contribution to total alpha activity			Specific activity ($\mu Ci\ \mu g^{-1}$)[a]
		^{234}U	^{235}U	^{238}U	
Natural	0.72	0.48	0.03	0.49	6.8×10^{-7}
Enriched	93	0.96	0.04	< 0.01	6.5×10^{-5}
Depleted	0.2	0.28	0.01	0.71	3.6×10^{-7}

[a] Specific activities ($\mu Ci\ \mu g^{-1}$) of the individual isotopes are 6.26×10^{-3} for ^{234}U, 2.17×10^{-6} for ^{235}U, and 3.33×10^{-7} for ^{238}U. The inverse of the specific activity is the mass per unit activity. Thus, 1 µCi of depleted uranium weighs $(1 / 3.6 \times 10^{-7}) = 2.8 \times 10^{6}$ µg or 2.8 g and 1 µCi of enriched uranium weighs 0.015 g.

The contributions of each isotope to radiation dose from the various categories of uranium depend upon the fraction of the total alpha-particle activity due to that isotope. Table 1 shows that for natural uranium, alpha-particle emissions come primarily from ^{234}U and ^{238}U, with only a small contribution from ^{235}U. For enriched uranium, nearly all the alpha-particle activity is due to ^{234}U. For depleted uranium, ^{238}U provides more than 70% of the total alpha activity.

3. RADIATION DOSE COEFFICIENTS FOR URANIUM AND PLUTONIUM

Because the uranium isotopes are primarily alpha-emitters, intakes of uranium by inhalation and ingestion are the most important pathways leading to human radiation doses. Table 2 contains radiation dose coefficients (doses per unit intake) for inhalation and

ingestion of the three isotopes of uranium. Also included in the table are the corresponding values for ^{239}Pu and ^{240}Pu, the primary alpha-particle emitters in weapons grade plutonium used at Rocky Flats. There is little difference in estimated lung doses among the isotopes of uranium and plutonium. The effective doses due to inhalation are larger for plutonium because it is retained in body tissues longer than uranium. Differences between uranium and plutonium are larger for ingestion exposures. However, ingestion doses per unit intake uranium or plutonium isotopes are much lower than those resulting from inhalation (compare columns 3 and 4 of Table 2).

Table 2. Doses per Unit Activity for Uranium and Plutonium Isotopes

Nuclide	Lung dose (rad) per μCi inhaled[a]	Effective dose (rem) per μCi inhaled[a]	Effective dose (rem) per μCi ingested[b]
^{234}U	14	35	0.18
^{235}U	13	31	0.17
^{238}U	12	30	0.17
^{239}Pu, ^{240}Pu	16	59	0.93

[a] Dose coefficients from ICRP (1995) for adults inhaling insoluble oxides with activity median aerodynamic diameters of 1 μm.

[b] Dose coefficients from ICRP (1996) for adults ingesting forms that may be more soluble because of modification in the environment.

Calculating doses per unit mass of natural, enriched, and depleted uranium requires information from Tables 1 and 2. The isotopic activity contributions and the specific activities listed in Table 1 were used together with the dose coefficients given in Table 2. The results, shown in Table 3, provide a basis for comparing the relative significance of releases of different masses of uranium and plutonium. As in Table 2, inhalation exposures to all materials are based on a reference activity median aerodynamic diameter (AMAD) for particles of 1 μm. The amount of each contaminant that reaches the deep lung varies with the AMAD of the aerosol inhaled and is independent of the contaminant. For that reason, ratios of lung doses (e.g., for enriched uranium compared to plutonium) for other particle sizes would be comparable to those in Table 3.

Table 3. Dose Coefficients per Unit Mass for Uranium and Plutonium

Material	Lung dose (rad) per μg inhaled	Effective dose (rem) per μg inhaled	Effective dose (rem) per μg ingested
Natural U	9.2×10^{-6}	2.2×10^{-5}	1.2×10^{-7}
Enriched U	9.3×10^{-4}	2.3×10^{-3}	1.2×10^{-5}
Depleted U	4.7×10^{-6}	1.1×10^{-5}	6.1×10^{-8}
WG Pu[a]	1.2	4.2	6.6×10^{-2}

[a] Weapons grade plutonium (WG Pu), which contains 93.8% ^{239}Pu and 5.8% ^{240}Pu, has a specific activity for alpha-particle emission of 0.072 μCi per μg.

Table 3 shows that enriched uranium poses a radiological hazard per unit mass that is about 100 times that from natural uranium and about 200 times that from depleted uranium. It can also be seen from Table 3 that inhalation of 1 μg of plutonium released from Rocky Flats would result in a dose of 1.2 rad to the lung. To receive the same dose from inhalation of enriched uranium would require inhalation of about 1300 μg (1.3 mg) of that material. Inhalation of about 250,000 μg (0.25 g) of depleted uranium would be needed to produce the same 1.2-rad dose to the lung. For intakes of such large quantities of uranium, chemical toxicity becomes a primary concern (see Section 5).

4. ROCKY FLATS RELEASES OF URANIUM TO THE ATMOSPHERE

Large masses of uranium were used at Rocky Flats and, as for plutonium, weighing was an important part of the inventory accounting procedure. Enriched uranium, like plutonium, was quite valuable and undoubtedly received more attention than depleted uranium, which was handled and released in larger quantities. For depleted uranium, there are two categories of routine releases: (1) effluents from stacks and vents that were measured routinely and (2) releases from open burning.

Estimates of the routine effluent releases of enriched and depleted uranium were made as part of Phase I. As part of that effort, independent estimates were made for some years as a check on reported values. Estimates of uncertainty in the release were based upon considerations of uncertainties in sampling flow rate, ventilation exhaust flow rate, analytical procedure, and in the identity of specific alpha-emitters. Previous estimates were found to be somewhat biased. A median bias factor of 1.3, with a geometric standard deviation (GSD) of 1.6 was applied to all previous estimates of uranium releases (ChemRisk 1994).

In Phase II, two other aspects of the effluent sampling program were examined. These were (a) the effect of estimating releases from a large duct with only one centrally located sampling point (that is, were samples collected in that manner representative?) and (b) the particular factor that had been used to account for self-absorption of alpha particles when samples were counted. This analysis led to further revision and increases in the estimates of routine releases of plutonium (Voillequé 1999a).

The estimates of bias that were obtained for the plutonium sampling results (Voillequé 1999a) are considered to be a first approximation of the additional bias correction that should be applied to routine uranium effluents in stacks and vents. For the purpose of the present comparisons, those corrections were applied to the results from Phase I to compute estimates of the highest annual and total routine releases of enriched and depleted uranium from Rocky Flats. The results are shown in Table 4. Routine airborne effluent releases of depleted uranium are estimated to be substantially larger than those for enriched uranium. The highest annual routine releases for both forms occurred during the mid-1950s.

The potential doses from these routine releases of uranium can be compared to plutonium releases using the information in Table 3. The lung dose of a release of 0.5 kg of enriched uranium, the 50th percentile value shown in Table 4, is comparable to that produced by the release of about 0.4 g of weapons grade plutonium. The comparison for depleted uranium is also based upon the 50th percentile value from Table 4 of 200 kg. The lung dose for that release is comparable to release of about 0.8 g of weapons grade plutonium.

Table 4. Percentiles of Estimated Distributions of Peak Annual and Total Routine Effluent Releases of Enriched and Depleted Uranium from Rocky Flats

	Peak annual release (kg)[a]			Estimated total release (kg)[a]		
	Percentile of distribution			Percentile of distribution		
Material type	5th	50th	95th	5th	50th	95th
Enriched uranium[b]	0.016	0.069	0.35	0.32	0.52	1.0
Depleted uranium[c]	5.6	24	120	120	200	380

[a] Based upon effluent measurements, revised to reflect identified biases and uncertainties that previously led to underestimation of releases.
[b] Year of peak release was 1956.
[c] Year of peak release was 1955.

The most appropriate plutonium release for comparison to these quantities is the approximately 1.8 g from routine releases to the atmosphere via stacks and vents (Voillequé 1999a). The comparison indicates that, based upon radiation dose, routine releases of plutonium were 4 to 5 times more important than routine releases of enriched uranium. On the same basis, routine releases of plutonium were about 2 times more important that routine releases of depleted uranium.

Accidental releases of plutonium, which were much higher than the routine releases, provide a basis for comparison with accidental releases of uranium. If all 60 kg of the depleted uranium sheets contained in the pallet that was burned were released to the air, which is unlikely, the lung dose would be comparable to that from a release of 0.2 g of weapons grade plutonium. The estimated release of plutonium from the 903 Area during the most important high wind day in January 1969 was about 12 g (Weber et al. 1999). Although both were effectively ground level releases, dispersion factors for the two releases are not likely to be equal. Nonetheless, the comparison indicates that, on the basis of radiation dose, it is likely that the accidental burning of depleted uranium is distinctly less important than the plutonium release from the 903 Area on just one of the five important high wind days.

Similar results are obtained when low probability release estimates (95^{th} percentile values) for uranium are compared with similar estimates for plutonium releases that have been studied in detail. Potential doses from routine and accidental releases of uranium are well below those from comparable plutonium release estimates.

5. CHEMICAL TOXICITY OF URANIUM

It is known that intakes of heavy metals, including uranium, can produce toxic effects in the body. For uranium, the organ of most concern is the kidney. Toxic effects may be most important for both natural and depleted uranium, whose specific activities (Table 1) and radiation dose coefficients per unit mass (Table 3) are small. For enriched uranium, radiation dose rather than chemical toxicity is the primary concern (see Table 3).

A detailed review of the chemical toxicity of uranium is given in Appendix R of our report dealing with the Fernald dose reconstruction project (Killough et al. 1998). That review is summarized briefly here. A concentration of 3 μg of uranium per gram of kidney tissue (3 μg U g^{-1}) has been used as a threshold level for workers in uranium facilities for many years. Those workers, whose kidney burdens were probably consistently lower than the guide value, have not exhibited effects. The BEIR IV report (NAS/NRC 1988) indicates that kidney damage appears to be definite at a concentration of 3 μg U g^{-1} and may occur at 1 μg U g^{-1} in some animal species. Recent papers have suggested that it may be more prudent to apply a safety factor of 10 to this lower level and consider a guide value of 0.1 μg U g^{-1} for members of the public.

While it is not uniformly agreed that effects will be produced at levels near the proposed guide value, a graded approach based on probability of effects was adopted for evaluation of kidney toxicity in the Fernald study. The interpretation of the probability of effects is as follows. Effects are unlikely for concentrations in the range 0.1–0.2 μg U g^{-1}. Possibly mild effects on the kidney occur at about 0.5 μg U g^{-1}. More severe effects begin at concentrations of ~1 μg U g^{-1} and become quite definite at ~3 μg U g^{-1}.

The guide value that limits uranium concentration in the kidney can be translated into a concentration limit for uranium in drinking water. The necessary metabolic data and water intake rates for three ages are provided by Killough et al. (1998). The ages considered are infants, 10-year old children, and adults. There are three components of uranium retention by the kidney. They are characterized by retention half-times of approximately 8–10 d, 100–240 d, and 2000–3000 d, respectively. In the ranges shown, the longer retention half-times are for infants. While the latter two components increase quite slowly, the first component of the kidney burden approaches an equilibrium uranium content after about one month of constant intake of uranium contaminated drinking water.

Because the kidney of the infant is small and because uptake of uranium from the GI tract by infants is considered to be greater than for older ages, drinking water intake of uranium by infants is the most limiting case. To avoid a concentration of 0.1 μg U g^{-1} in the infant kidney, the concentration of uranium in drinking water must be less than 200 μg L^{-1}. For depleted uranium, the corresponding activity concentration is about 70 pCi L^{-1}.

For intakes by inhalation that continue for period of 1–2 years, the limiting exposure is to an active adult. For short (~1-month) exposure periods, an infant's exposure is limiting but the derived air concentration guide is about 4 times larger than for the extended exposure of the active adult. Exposure of an adult near the site boundary is also much more probable. To avoid a concentration of 0.1 μg U g^{-1} in the exposed adult's kidney, the concentration of uranium in air at the point of exposure must be less than 20 μg U m^{-3}.

The main uranium processing facilities are on the south side of the developed area at the Rocky Flats site. Average annual dispersion factors for ground level releases (DOE 1980) from those facilities are estimated to be 3–5 x 10^{-7} s m^{-3} for areas of likely exposure. The expected average air concentration due to a depleted uranium release of 120 kg in a year (95^{th} percentile value for 1955 from Table 4) is about 2 x 10^{-3} μg U m^{-3}. That concentration is about 10,000 times less than the derived guide value for air.

It is reasonable to consider short-term exposures of infants that were passing by the facility. For short-term releases the dispersion factors are higher. For points of likely exposure, values in the range 2–4 x 10^{-5} s m^{-3} are estimated for stability Class E and a wind speed of 3 m s^{-1} (DOE 1980). For a day with a persistent wind direction and those conditions,

a release rate of about 170 kg d^{-1} of depleted uranium would produce air concentrations above the guide (80 μg U m^{-3}) derived for infant exposures lasting a month. However, such a short exposure would cause only a transient elevation of uranium concentration in the kidneys of infants who were exposed. This type of transient concentration increase is less likely to cause damage than an extended exposure that maintains the kidney concentration above the guide value of 0.1 μg U g^{-1}.

6. ROCKY FLATS RELEASES OF URANIUM TO SURFACE WATERS

Rocky Flats operations released uranium to surface water in both the Walnut Creek and Woman Creek drainages. These streams flow into Great Western Reservoir and Standley Lake, respectively, both of which served as sources of drinking water. Measurements of the effluent concentrations and of radioactivity in the reservoirs that these streams enter have been described in Section VI of the Phase II Task 4 report (Rope et al. 1999). Historic measurements of stream flow rates are also presented by Rope et al. (1999).

Early effluent measurements were of gross alpha radioactivity and there was no differentiation between the uranium and plutonium fractions from plant effluents. In later years, specific radiochemical analyses indicated that about 37% of the alpha activity in liquid effluents was plutonium and about 63% was uranium (Rope et al. 1999).

The highest monthly average concentrations of gross alpha activity in water discharged to Walnut Creek were between 200 and 300 pCi L^{-1} in 1954. Peak daily values as high as about 500 pCi L^{-1} were measured in effluent from Building 74. Effluent concentrations in later years were generally lowered by the installation of settling ponds in both parts of the Walnut Creek drainage. However, operational problems in 1960 and 1962 caused liquid waste releases that were nearly as high as those that occurred in 1954. When work was performed on the settling ponds in 1972–73, two weekly gross alpha concentrations in the range 600–800 pCi L^{-1} were measured by the Colorado Department of Health. Monthly average concentrations comparable to those in 1954 were also measured during this period.

On the basis of flow measurements made by the US Geological Survey, liquid effluents from Rocky Flats are estimated to contribute between 2.5 and 4.7% of the water that enters the Great Western Reservoir (Rope et al. 1999). By neglecting any settling of material in the stream bed and in the reservoir sediment and by assuming rapid mixing or reservoir waters, one can estimate a peak concentration of uranium in Great Western Reservoir of 20–40 pCi L^{-1}. This estimate also assumes that all of the gross alpha activity during the period of highest concentrations was uranium. The highest monthly average value to which an infant may have been exposed during that period is estimated, using the same assumptions, to be 8–15 pCi L^{-1}. These water concentrations are less than 70 pCi L^{-1}, the concentration needed to produce uranium concentrations as high as 0.1 μg U g^{-1} in an infant's kidneys.

The water used to backwash the filter in the Rocky Flats water treatment system was discharged to Woman Creek before 1975. Improper disposal of solid wastes could also lead to uranium contamination in that stream. Woman Creek flows into Standley Lake, which has also been used as a drinking water supply. Measurements of radioactivity concentrations in Woman Creek are not discussed in the Task 4 report. However, measurements of radioactivity in Standley Lake were performed regularly and the data are presented by Rope et al. (1999). Annual average concentrations of total long-lived alpha activity were generally

in the range 1–3 pCi L^{-1} and were always below 6 pCi L^{-1} during the years 1951–1971. The highest annual average concentrations (5–6 pCi L^{-1}) in both Standley Lake and Great Western Reservoir both occurred during 1966. These measurements include the contributions of naturally occurring uranium and thorium and their alpha-emitting progeny, such as ^{226}Ra. The highest monthly average concentration during that year could not have exceeded 50–60 pCi L^{-1} because concentrations during the other months would have been 1–2 pCi L^{-1}.

Consumption of drinking water that contains 6 pCi L^{-1} of depleted uranium throughout the year would produce a committed dose to an adult of about 0.5 mrem. Estimated doses to children are somewhat higher, 0.7–0.9 mrem. Children are smaller, but they also drink less water; therefore, the difference in dose between adults and children is not as large as one might expect from considerations of body size alone. For comparison, the dose to an adult who consumed ^{3}H that was released from Rocky Flats and appeared in drinking water from Great Western Reservoir in 1973 was estimated to be 0.3 mrem (Rope et al. 1999). The estimated dose to a child who regularly consumed that water is about 0.5 mrem. The peak annual average doses from uranium in drinking water (in 1966) are seen to be 1.4–1.8 times those from tritium during the year (1973) of highest releases of that radionuclide from Rocky Flats.

7. URANIUM ACCOUNTING AT ROCKY FLATS

In the mid-1970s there was a review of the amounts of material unaccounted for (MUF) for critical materials, uranium and plutonium, used at Rocky Flats. Similar reviews were conducted at other uranium and plutonium production facilities around the country. The report, with deletions, for enriched uranium at Rocky Flats (Young 1976) is available. By the time the review was performed, work on production of weapons using enriched uranium had been concentrated in Oak Ridge, Tennessee. The Rocky Flats enriched uranium production work stopped in 1965 and the processing facility was then cleaned out.

Young (1976) reported that the amount of MUF for highly enriched uranium at Rocky Flats was about 347 kg over the entire period, fiscal years (FY) 1954–1976. At the end of FY 1964, the MUF amount was about 414 kg. Approximately 67 kg of enriched uranium were recovered during cleanup after the facility had been shut down (Young 1976). Young concluded that inadequate measurement methods for wastes and residues were probably the main factor that caused imbalance in the accounting of enriched uranium. Nondestructive assay capabilities, such as barrel counting, only became available after the facility was shut down (Young 1976). For comparison, the MUF amount for plutonium at Rocky Flats in 1976 was 975 kg (Young 1977), about two-thirds of which was unaccounted for by the end of 1964 (Zodtner and Rogers 1964).

Through the end of 1970, it was estimated that about 130 kg of ^{235}U had been shipped to Idaho in solid waste (Lee 1971). Based upon experience with plutonium waste, discussed in the report on the 1957 fire (Voillequé 1999b), the true amount could easily have been 2–3 times greater.

It has been shown (NAS 1990; Voillequé 1999b) that when the quantities released from a facility are much smaller than the throughput of the plant, mass balance accounting has no value for determining releases by difference. That is, the MUF amount of 347 kg is not a reliable guide to the amount of enriched uranium released to air and water at Rocky Flats.

However, the MUF amount does provide a gross upper bound that may be useful for some types of calculations.

No comparable analysis of MUF for depleted uranium at Rocky Flats has been located. Information on inventories of depleted uranium during the years 1952–1976 has been found, suggesting that such an analysis was at least considered at the same time analyses were performed for enriched uranium and plutonium. However, only changes in the order of magnitude of the inventory are indicated. Large shipments of depleted uranium were received after April 1954 and total monthly inventories of tens of thousands of kilograms were reported from June 1954 through March 1955. At that time, the inventory increased to hundreds of thousands of kilograms. Except for the period from July 1955 until September 1956, monthly inventories of depleted uranium at Rocky Flats were hundreds of thousands of kilograms until late in 1969. From November 1969 through the end of 1976, the inventory was tens of thousands of kilograms, except for 7 months in 1971–1972 when it was again hundreds of thousands of kilograms.

To estimate the MUF for depleted uranium at Rocky Flats, it was necessary to make an assumption about the relationship between throughput and inventory. Operating experience at the Fernald, Ohio, uranium processing plant was used as a guide. During peak production years there, the throughput, as measured by shipments, was about double the inventory of natural and low enriched uranium. We further assume that, on the average, an inventory of "tens of thousands" of kilograms can be assigned the value 50,000 kg. Assuming that the maximum inventory was about 600,000 kg, we interpret "hundreds of thousands" of kilograms to be approximately 300,000 kg. Using these estimates, we can estimate the depleted uranium throughput at Rocky Flats. There were 170 months during the period when the inventory was hundreds of thousands of kilograms. An estimate of the throughput during most of the time between 1955 and 1970 is about 600,000 kg per month.

At Fernald, the overall (1952–1976) MUF as a fraction of receipts of natural uranium was 0.17%. The overall fractional MUF for low enriched uranium at Fernald was lower, 0.03%, which seems to reflect greater care of that material. If the fractional MUF for natural uranium at Fernald is a reasonable guide, the monthly MUF for depleted uranium at Rocky Flats can be estimated to be about 1000 kg during periods of peak inventory. During the 114 months when the inventory was tens of thousands of kilograms, the same analysis leads to an estimate of about 170 kg for the monthly MUF. An estimate of the total MUF for depleted uranium at Rocky Flats is about 190,000 kg.

Lee (1971) estimated that about 200,000 kg of depleted uranium had been shipped to Idaho for burial. Given the inadequacy of measurements of radionuclides in solid wastes that were shipped, this estimate of the MUF amount for depleted uranium is not inconsistent with the estimated amount shipped for burial. Historic plutonium burial estimates made at Rocky Flats are now thought to be 2–3 times lower than the amount actually in the ground (Voillequé 1999b). Similar underestimates for uranium are not inconceivable. The enriched and depleted uranium identified as unaccounted for may well be in the Burial Ground at what is now called the Idaho National Engineering and Environmental Laboratory.

The cautions given above about interpreting formal estimates of MUF as indications of releases to air and water apply even more strongly to the even more uncertain estimate of the MUF for depleted uranium at Rocky Flats. While the estimated MUF amount is not a credible basis for estimating the amounts of uranium that were actually released in airborne and liquid effluents, it may serve other purposes.

8. CONCLUSIONS

These calculations have examined the expected consequences of releases of enriched and depleted uranium at Rocky Flats. The principal hazard from enriched uranium is due to radiation exposure. In contrast, exposure to depleted uranium poses a greater chemical toxicity hazard. For radiological risks, we used comparisons with plutonium, the primary source of radiation exposure from Rocky Flats. For chemical toxicity, we estimated guide values for concentrations in water and air using a cautious estimate of the chemical toxicity threshold for uranium in the human kidney. That recently proposed threshold, 0.1 $\mu g\ U\ g^{-1}$, is 30 times lower than that traditionally used for worker protection.

We compared releases of enriched uranium to the atmosphere to airborne releases of plutonium that have been studied in detail in Phase II. Both radiological and chemical toxicity comparisons were made for releases of depleted uranium to the atmosphere. Releases of depleted uranium to streams that feed reservoirs used for drinking water were compared with the concentration guide value derived to keep kidney concentrations below the 0.1 $\mu g\ U\ g^{-1}$ threshold.

None of the comparisons made suggests that uranium releases deserve much further attention. Releases of enriched uranium have been estimated to pose substantially less risk than the plutonium releases that have been the focus of Phase II. Radiological risks from depleted uranium are also small with respect to those from plutonium. The potential for toxic effects on the kidney from intakes of depleted uranium also does not appear to be substantial. Measured uranium concentrations in raw water are well below the derived guide value and treatment of drinking water before distribution could have reduced those levels. Very large releases of depleted uranium to the atmosphere in a short time under adverse meteorological conditions could have resulted in a transient elevation of uranium levels in kidneys of exposed persons. Such exposures are not likely to cause kidney damage.

The accountability data indicate that nearly 350 kg of enriched uranium was unaccounted for after closure and cleanup of the processing facility at Rocky Flats. As is the case for plutonium, whose MUF was larger, the failure to make reliable measurements of the amounts of material in solid wastes is likely responsible for most of the MUF. We made a speculative, but not unreasonable, estimate of the MUF for depleted uranium. The amount is large, 190,000 kg, and quite uncertain because of the lack of information. Likely reasons for the depleted uranium MUF were a large throughput; a lower level of concern for that less valuable material; and inadequate measurements of solid wastes, which were disposed onsite and shipped to Idaho for burial. Because methods for measuring the amounts of uranium in solid wastes were not reliable, it is quite conceivable that the amounts of Rocky Flats uranium identified as unaccounted for were buried.

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