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Nuclear and Chemical Waste Programs

Remedial Action Program

ESTIMATES OF RELEASE RATES OF RADIONUCLIDES FROM BURIAL GROUNDS AT ORNL

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1. Introduction

An important aspect of environmental pathways and dose-assessment modeling for disposal of radioactive wastes at Oak Ridge National Laboratory (ORNL) involves obtaining estimates of source terms, i.e., rates of release of radionuclides from disposal facilities. Even crude estimates of release rates from disposal sites can be useful in planning for remedial actions.

Reliable (i.e., validated) models for predicting source terms for solid or liquid wastes placed in shallow trenches have not been developed, partly because the source term is usually determined by environmental conditions and waste disposal practices which are highly site-specific. However, efforts are underway at Brookhaven National Laboratory to develop general models for predicting source terms at shallow-land burial facilities (Sullivan and Kempf, 1987; Sullivan, 1988; Sullivan *et al.*, 1988). These models show promise for application to specific sites, such as those at ORNL, if the appropriate data on waste forms and on hydrologic and geochemical conditions are available.

In the absence of validated models, source terms can be estimated, in principle, on the basis of observations of facility performance. Estimation of the source term for a shallow-land disposal facility on the basis of field data would require (1) knowledge of the concentrations (or total inventories) of radionuclides in the disposal unit, (2) time-series data on concentrations of radionuclides in ground waters or surface waters that are believed to be recharged by water that has infiltrated through the disposal facility, and (3) data on the infiltration rates of water and the flow rates of the appropriate ground or surface waters. Measurement of the relevant time-series data over a period of at least a few years may be particularly important for estimation of the source term. It is difficult to specify *a priori* a sufficient length of time over which such data would be needed, because of such factors as the possibility of highly transient responses of the disposal system, particularly during a relatively short time period after disposal, and uncertainties in the time required for the releases to approach steady-state conditions. If a source term can be estimated from field observations, then an important source of data for validating models of facility performance is provided.

To the best of my knowledge, data do not presently exist for any waste disposal sites at ORNL which would be sufficient for reliable estimation of a source term. However, there are cases where data exist from which crude estimates of fractional release rates (i.e., the probability per unit time for release of a radionuclide) from some disposal sites into surface waters can be obtained. These estimates are highly uncertain because of several factors, including (1) the lack of reliable data on radionuclide inventories at disposal for many of the important burial grounds, particularly those for solid wastes, (2) uncertainties in apportioning observed radionuclide releases to surface waters among the different possible sources, and (3) the possibility that a substantial portion of the observed activity of some radionuclides in surface waters arises from ongoing operations at ORNL, rather than releases from waste disposal sites.

The following examples illustrate the potential importance of obtaining even crude estimates of fractional release rates from disposal sites. For ^{3}H, ^{90}Sr, and ^{137}Cs, which are important radionuclides with half-lives of a few decades or less, it would be useful to determine if the fractional release rate is substantially less than the rate of reduction in inventory due to radioactive decay. If this were the case, then predictions of dose to future inadvertent intruders could be based essentially on estimates of concentrations of these radionuclides in disposal facilities that take into account only the initial inventory and the reductions via decay. On the other hand, if the rate of removal of these radionuclides via transport processes were rapid compared with losses due to decay, but assuming that the doses to off-site individuals were still below acceptable limits, then the potential doses to future intruders would be greatly reduced and the need for cleanup of some disposal sites possibly obviated. For such important long-lived radionuclides as ^{99}Tc and isotopes of uranium, radioactive decay is not effective in reducing inventories over tens of thousands of years. Thus, predictions of dose to future intruders for these radionuclides could depend on the rate of removal from the disposal facility via transport processes, provided the removal rate were sufficiently rapid to significantly reduce the inventory in the facility during the time period over which active or passive institutional controls were assumed to be effective in preventing exposures of intruders. Finally, it is perhaps stating the obvious to note that estimates of release rates of radionuclides from disposal facilities are essential for predicting future doses to off-site individuals.

The following discussion considers some estimates of fractional release rates of radionuclides from particular waste disposal sites. Again, it must be borne in mind that much of the data used in these calculations involves assumptions that are quite speculative and, thus, may be highly uncertain. Therefore, the numerical results should not be taken literally but only as crude indicators of the possible behavior of radionuclides in the ORNL burial grounds. The analysis will focus on release rates of radionuclides from Solid Waste Storage Areas (SWSAs) 3-6 and from the pits and trenches used for disposal of liquid wastes.

2. Inventories of Radionuclides in SWSAs

A principal source of uncertainty in estimating fractional release rates of radionuclides from disposal facilities for solid wastes is the general lack of data on inventories of radionuclides at disposal. Only for SWSA 6 for the period since 1977 are radionuclide-specific inventories at disposal available (Boegly *et al.*, 1985). Disposals during prior operations at SWSA 6 could be estimated by extrapolation of the data since 1977, but such estimates must be regarded as uncertain because of the highly variable temporal pattern of disposals for particular radionuclides that apparently occurred (Davis and Solomon, 1987). For SWSAs 3-5, not only are radionuclide-specific data lacking, but reliable data on the total activity of all radionuclides at disposal also appear to be lacking. Furthermore, disposals at SWSAs 3-5 probably cannot be obtained with any reliability by extrapolation of the recent data for SWSA 6, because of the changing nature over the years of operations at ORNL that generated solid wastes (McNeese, 1987). Therefore, crude guesswork necessarily is involved in estimating radionuclide inventories in SWSAs 3-6.

Bates (1983) has estimated the total activity of radionuclides placed in SWSAs 3-5. These data, including the years during which each facility was operated, are given as follows:

SWSA 3 – 1946-51, 5×10^4 Ci;
SWSA 4 – 1951-59, 1×10^5 Ci;
SWSA 5 – 1959-73, 2×10^5 Ci.

Thus, the total disposals in SWSAs 3-5 are estimated to be about 4×10^5 Ci. The estimates for the individual SWSAs are consistent with the values recently compiled for the Remedial Action Program (Oak Ridge National Laboratory, 1987), presumably from the same data base. The ORNL report also has indicated that these estimates may be upper limits.

Detailed data have not been reported on the inventories of individual radionuclides in SWSAs 3-5. For purposes of estimating fractional release rates of radionuclides from these facilities, we make the following assumptions (J. R. Trabalka, private communication):

- one-third of the inventory at disposal was ^3H; and

- one-third of the inventory at disposal was ^{90}Sr plus ^{137}Cs, and the inventories of these two fission products were approximately the same.

The assumption of equal inventories of ^{90}Sr and ^{137}Cs is based on the approximate equality of their yields from thermal-neutron fission of ^{235}U (Hyde, 1964), the reported inventories in tank wastes at ORNL (Huang *et al.*, 1984), and the recent disposals in SWSA 6 discussed below.

From the foregoing assumptions, we obtain the following estimates of total inventories of ^{3}H, ^{90}Sr, and ^{137}Cs at disposal in SWSAs 3-5, which we further assume are upper limits:

^{3}H - $<1 \times 10^5$ Ci;
^{90}Sr - $<6 \times 10^4$ Ci;
^{137}Cs - $<6 \times 10^4$ Ci.

Again, the speculative nature of these estimates cannot be overemphasized. A previous report (U.S. Energy Research and Development Administration, 1976) noted that burials of ^{3}H through fiscal year 1967 may have been as high as 9×10^4 Ci, and that no significant amount of ^{3}H has been added since that year. This estimate for the total inventory of ^{3}H in the SWSAs agrees well with the estimate given above. However, this same report estimates that total disposals up to 1976 of radioactivity with long half-lives (presumably mostly ^{90}Sr and ^{137}Cs) were less than 10^4 Ci, which is considerably less than the estimate of about 10^5 Ci given above, and that annual disposals were less than 10^3 Ci. These estimates cannot be verified, but there is some evidence that they may underestimate, rather than overestimate, disposals of ^{90}Sr and ^{137}Cs, because recent disposals in SWSA 6 discussed below have averaged about 10^3 Ci per year and previous inventories of ^{90}Sr and ^{137}Cs at generation probably were considerably higher due to the nature of the waste generation processes (McNeese, 1987).

For purposes of this analysis, the previous estimates of ^{90}Sr and ^{137}Cs disposals (U.S. Energy Research and Development Administration, 1976), i.e., about 5×10^3 Ci for each isotope, are assumed to be lower limits. Therefore, the estimates of inventories for these two radionuclides may be uncertain by about an order of magnitude on the basis of the estimated upper limit we obtained above.

For SWSA 6, radionuclide-specific inventories at disposal for the years 1977-1984 are given by Boegly *et al.* (1985). The average yearly disposals during this time period for ^{3}H, ^{90}Sr, and ^{137}Cs were about 1,000 Ci, 400 Ci, and 700 Ci, respectively. Thus, from the assumed inventories listed above for the other burial grounds, total disposals of these radionuclides in SWSA 6 since 1969 may be much less than the disposals in SWSAs 3-5. Again, however, extrapolations of the data since 1977 to obtain estimates of prior disposals in SWSA 6, as well as the estimates of inventories in SWSAs 3-5, are uncertain.

3. Inventories of Radionuclides in Pits and Trenches

Spalding (1987) has reported data on inventories of radionuclides discharged as liquid wastes into the pits and trenches, the most important of which are Pits 2-4 and Trenches 5 and 7. On the basis of the data for Pits 2-4 and Trench 7, we assume that the ratio of ^{90}Sr to ^{137}Cs activity is 0.2. The estimated total activities at disposal in the pits and trenches then are as follows:

^{90}Sr - 1.4×10^5 Ci;
^{137}Cs - 6.2×10^5 Ci.

Olsen *et al.* (1986) have reported estimates of inventories for a large number of radionuclides in Trench 7, as well as measurements of radionuclide concentrations in Trench 7 ground water and in nearby soils and weathered bedrock. However, in the absence of time-series data on the concentrations in ground water, as well as data on infiltration rates of water through the trench and on ground-water flow rates, the data of Olsen *et al.* do not provide a basis for estimating a source term for Trench 7.

4. Estimates of Fractional Release Rates of Radionuclides

This section discusses some estimates of fractional release rates of radionuclides from various disposal facilities. These estimates are based on the assumptions regarding radionuclide inventories presented above and data on the activity of radionuclides in surface waters.

4.1 *Release Rates for Sr-90*

Bates (1983) has reported annual discharges of ^{90}Sr from SWSA 4 for the years 1963-1977 and from SWSA 5 for the years 1967-1977. For SWSA 4, the annual discharges are in the range 1-5 Ci, with a mean value of about 3 Ci; and for SWSA 5, the annual discharges are in the range 0.4-3 Ci, with a mean value of about 1 Ci.

On the basis of the assumptions regarding the upper limits for radionuclide inventories in SWSAs 4 and 5 discussed in Section 2, the following estimates of inventories of ^{90}Sr at disposal are obtained:

SWSA 4 – <2 x 10^4 Ci;
SWSA 5 – <3 x 10^4 Ci.

Therefore, the estimated lower bounds for the fractional release rates are as follows:

SWSA 4, 1963-1977 –
Release rate > (3 Ci/y)/(2 x 10^4 Ci) ≃ 2 x 10^{-4} y^{-1};

SWSA 5, 1967-1977 –
Release rate > (1 Ci/y)/(3 x 10^4 Ci) ≃ 3 x 10^{-5} y^{-1}.

Based on the assumptions regarding the data on inventories in the report of the U.S. Energy Research and Development Admininstration (1976) discussed in Section 2, the estimated upper bounds for the fractional release rates would be about an order of magnitude greater, i.e., less than about 2 x 10^{-3} y^{-1} for SWSA 4 and 3 x 10^{-4} y^{-1} for SWSA 5.

Estimates of annual releases of ^{90}Sr from SWSAs 4 and 5 also have been reported for the years 1979-1986 (Oakes *et al.*, 1987b), and the average values are 0.6 Ci and 0.5 Ci, respectively. Since these releases occurred a considerable time after disposal, we assume that the initial inventory in each burial ground has been depleted by about a factor of 2 due to radioactive decay. Thus, the estimated lower bounds for the fractional release rates for this time period are as follows:

SWSA 4, 1979-1986 –
Release rate > (0.6 Ci/y)/(0.5 x 2 x 10^4 Ci) ≃ 6 x 10^{-5} y^{-1};

SWSA 5, 1979-1986 –
Release rate > (0.5 Ci/y)/(0.5 x 3 x 10^4 Ci) ≃ 3 x 10^{-5} y^{-1}.

Again, the estimated upper bounds for the fractional release rates would be an order of magnitude greater.

Several tentative conclusions can be drawn from the estimates discussed above. First, the release rates of ^{90}Sr do not appear to vary greatly for the two burial grounds. Second, the release rates from either burial ground apparently have not varied greatly with time over the last 25 years. Third, the release rate from either burial ground appears to be much less than the rate of depletion due to radioactive decay, which is 0.024 y^{-1}, even on the basis of the upper bound estimates. Therefore, the inventory of ^{90}Sr in SWSAs 4 and 5 over time appears to be controlled primarily by radioactive decay, rather than removal by physical transport mechanisms.

Data given by Oakes *et al.* (1987b) also provide an upper limit for annual releases of ^{90}Sr from SWSA 3 for the period 1979-1986 of 0.6 Ci. This estimate is an upper limit, because releases from sources other than

SWSA 3 may be included. From the assumptions for the inventories of radionuclides in the SWSAs given previously, the estimated inventory of ^{90}Sr at disposal for SWSA 3 is less than 8,000 Ci but greater than 800 Ci. Thus, if we assume depletion of the initial inventory by about a factor of 3 due to radioactive decay, the estimated lower bound for the fractional release rate is as follows:

SWSA 3, 1979-1986 -
$$\text{Release rate} > (0.6 \text{ Ci/y})/(0.3 \times 8{,}000 \text{ Ci}) \approx 3 \times 10^{-4} \text{ y}^{-1}.$$

Again, the upper bound estimate would be an order of magnitude greater. These results are remarkably consistent with the previous estimates for SWSAs 4 and 5.

According to Sears (1987), the release of ^{90}Sr from SWSA 6 during 1985 was about 3 mCi, although it is uncertain if the monitoring frequency was sufficient to obtain an accurate estimate of the release. If we assume that the total inventory of ^{90}Sr at disposal for SWSA 6 is about twice the value reported by Boegly *et al.* (1985) for the period 1977-1984, i.e., about 6,000 Ci, and that radioactive decay can be neglected, then the fractional release rate is estimated as follows:

SWSA 6, 1985 -
$$\text{Release rate} = (0.003 \text{ Ci/y})/(6{,}000 \text{ Ci}) \approx 5 \times 10^{-7} \text{ y}^{-1}.$$

This is a very low release rate in comparison with the estimates for SWSAs 3-5 given above, and probably indicates (1) an improved waste isolation capability for SWSA 6 and/or (2) that ^{90}Sr released from SWSA 6 has not yet migrated to surface streams, i.e., that the time interval between waste burials and the measurements in surface streams is too short for a realistic picture of the source term for SWSA 6 to emerge.

Reported annual releases of ^{90}Sr from the pits and trenches for the years 1980-1986 (Oakes *et al.*, 1987b) average about 0.05 Ci. From the estimated inventory of ^{90}Sr at disposal in the pits and trenches given in Section 3, and assuming depletion of the inventory by about a factor of 2 due to radioactive decay, we estimate the following fractional release rate:

Pits and trenches, 1980-1986 -
$$\text{Release rate} = (0.05 \text{ Ci/y})/(0.5 \times 1.4 \times 10^{5} \text{ Ci}) \approx 7 \times 10^{-7} \text{ y}^{-1}.$$

This is a very low release rate when one considers that the wastes were in liquid form; i.e., the estimated release rate is at least two orders of magnitude less than the release rate from the solid waste burial grounds exclusive of SWSA 6. However, geochemical and hydrologic conditions in some of the pits and trenches have been controlled to inhibit migration of ^{90}Sr (Olsen *et al.*, 1986; Spalding, 1987).

A crude estimate of an upper limit for the release rate of ^{90}Sr from all burial sites can be obtained from published data on discharges from White Oak Dam summarized by Edwards (1986) and estimates of total burials at ORNL. Such an estimate is an upper limit, because it does not take into account that a substantial fraction of discharges from White Oak Dam probably have resulted from ongoing ORNL operations. From data discussed in Sections 2 and 3, we estimate that the total inventory of ^{90}Sr at disposal is about 2×10^5 Ci. Since most of the activity apparently was placed in the pits and trenches, this estimate is largely insensitive to the uncertainties in the estimated inventories in SWSAs 3-5 discussed in Section 2. About 1,200 Ci of ^{90}Sr have been discharged from White Oak Dam since 1949 (Edwards, 1986). Furthermore, only a small fraction of the releases to White Oak Lake appear to have remained in the sediments (Sherwood and Loar, 1987). Thus, the estimated upper bound for the fractional release of ^{90}Sr since 1949 is as follows:

All sites, 1949-1985 -

$$\text{Fractional release} < (1{,}200\ \text{Ci})/(2 \times 10^5\ \text{Ci}) \simeq 6 \times 10^{-3}.$$

This is a small fraction of the total inventory at disposal.

The annual discharge at White Oak Dam for the years 1965-1985 averaged about 4 Ci (Edwards, 1986). Thus, if we include a factor of 2 for the average depletion of the original inventory by radioactive decay, then an upper limit for the fractional release rate from all sites during the later years is estimated as follows:

All sites, 1965-1985 -

$$\text{Release rate} < (4\ \text{Ci/y})/(0.5 \times 2 \times 10^5\ \text{Ci}) \simeq 4 \times 10^{-5}\ \text{y}^{-1}.$$

This is a low release rate that again probably provides an upper limit for the estimated performance of the pits and trenches where most of the ^{90}Sr is assumed to have been placed.

From these crude analyses, it seems reasonable to conclude that ^{90}Sr placed in various disposal facilities at ORNL has been retained to a high degree; i.e., the releases beyond the site boundaries have only been a small fraction of the total inventory at disposal. Therefore, inventories of ^{90}Sr at ORNL in the future probably will be controlled primarily by radioactive decay, rather than transport from the burial sites.

4.2 *Release Rates for Cs-137*

Few data have been reported related to releases of ^{137}Cs, even though, as indicated in Sections 2 and 3, the total inventory of ^{137}Cs at disposal probably is considerably greater than the inventory of ^{90}Sr.

Again, most of the ^{137}Cs apparently was disposed of as liquid wastes in the pits and trenches.

From data summarized by Sears (1987), releases of ^{137}Cs from SWSAs 4 and 5 during 1985 may be estimated as 0.7 Ci and 0.005 Ci, respectively. From the assumptions regarding inventories discussed in Section 2, the estimated inventories of ^{137}Cs at disposal for SWSAs 4 and 5 are less than 2×10^4 Ci and 3×10^4 Ci, respectively. Thus, by assuming that the initial inventories have been reduced by a factor of 2 by radioactive decay, the estimated lower bounds for the fractional release rates are as follows:

SWSA 4, 1985 -

$$\text{Release rate} > (0.7\ \text{Ci/y})/(0.5 \times 2 \times 10^4\ \text{Ci}) \approx 7 \times 10^{-5}\ \text{y}^{-1};$$

SWSA 5, 1985 -

$$\text{Release rate} > (0.005\ \text{Ci/y})/(0.5 \times 3 \times 10^4\ \text{Ci}) \approx 3 \times 10^{-7}\ \text{y}^{-1}.$$

As with the estimates for ^{90}Sr in Section 4.1, the upper bounds for the fractional release rates would be about an order of magnitude greater. Comparison of the estimates for ^{90}Sr and ^{137}Cs provides agreement with the prevailing view that the mobility of ^{137}Cs is considerably less than the mobility of ^{90}Sr under normal conditions at ORNL; e.g., see Olsen *et al.* (1986).

Data summarized by Edwards (1986) indicate that a total of 700 Ci of ^{137}Cs has been discharged from White Oak Dam since 1949. Furthermore, a comparable amount of the releases to White Oak Lake apparently has been retained in the sediments (Sherwood and Loar, 1987). From the assumptions regarding inventories of radionuclides in SWSAs 3-5 and the pits and trenches discussed in Sections 2 and 3, we estimate an initial inventory of ^{137}Cs at all disposal sites of about 7×10^5 Ci, most of which occurs in the pits and trenches. As with ^{90}Sr, this estimate is largely insensitive to the uncertainties in estimated inventories in SWSAs 3-5 discussed in Section 2. Therefore, the estimated upper limit for the fractional release of ^{137}Cs since 1949 is as follows:

All sites, 1949-1985 -

$$\text{Fractional release} < (2 \times 700\ \text{Ci})/(7 \times 10^5\ \text{Ci}) \approx 2 \times 10^{-3}.$$

We regard this result as a considerable overestimate, because a substantial fraction of the discharges from White Oak Dam probably has resulted from ongoing operations at ORNL, rather than releases from burial sites. In addition, much of the total release occurred when White Oak Lake was maintained empty and lake sediments were scoured. Therefore, as expected, the fractional release of ^{137}Cs probably is considerably less than the corresponding estimate for ^{90}Sr in Section 4.1.

The annual discharge of ^{137}Cs from White Oak Dam since 1965 has averaged about 1 Ci (Edwards, 1986). Therefore, if we include a factor of 2 for depletion of the original inventory by radioactive decay, then an upper limit for the fractional release rate for all sites for this time period is estimated as follows:

All sites, 1965-1985 -

$$\text{Release rate} < (2 \times 1\ \text{Ci/y})/(0.5 \times 7 \times 10^5\ \text{Ci}) \approx 6 \times 10^{-6}\ y^{-1}.$$

This release rate is about an order of magnitude less than the corresponding estimate for ^{90}Sr given in Section 4.1, and again probably overestimates considerably the actual source term for the pits and trenches where most of the ^{137}Cs is assumed to have been placed.

Even though the data that are available for estimating source terms for ^{137}Cs are quite scanty, it nonetheless seems reasonable to conclude that ^{137}Cs placed in various disposal facilities at ORNL has been retained to a very high degree. Thus, as with ^{90}Sr, inventories of ^{137}Cs in the future likely will be controlled by radioactive decay, rather than transport from the burial sites.

4.3 *Release Rates for H-3*

Tritium is expected to be a highly mobile radionuclide in the environment. Data summarized by Edwards (1986) indicate a total discharge of ^{3}H from White Oak Dam for the period 1964-1985 of 1.6×10^5 Ci. On the other hand, in Section 2 we estimated an upper limit for the total inventory of ^{3}H at disposal of only about 1×10^5 Ci; i.e., the reported discharges from White Oak Dam apparently are greater than the estimated inventory at disposal, and this discrepancy becomes even more pronounced when radioactive decay for the earlier burials and the lack of data on discharges prior to 1964 are taken into account. These results suggest that releases of ^{3}H to White Oak Lake from ongoing operations at ORNL have been significant in comparison with releases from disposal sites or that the inventory at disposal has been greatly underestimated. Even though it is reasonable to conclude that the rate of removal of ^{3}H from ORNL by transport processes is important compared with depletion via radioactive decay, it is difficult on the basis of available data to quantify release rates from individual disposal sites.

4.4 *Release Rates for Transuranic Radionuclides*

On the basis of available data, only crude and highly uncertain estimates of releases rates for transuranic (TRU) radionuclides may be obtained.

Data summarized by Edwards (1986) indicate that about 5 Ci of TRU radionuclides have been discharged from White Oak Dam since 1949. The average yearly discharge for this time period is about 0.1 Ci, but yearly discharges for the period 1971-1985 are only about 0.04 Ci. In addition, about 1 Ci of TRU radionuclides are found in the top 15-cm layer of sediments in White Oak Lake (Sherwood and Loar, 1987); data on TRU radionuclides in deeper layers of sediments are not reported. These results suggest that most of the releases to the lake have been discharged from the dam, which is a somewhat surprising result.

Estimates of the inventory of TRU radionuclides that have been buried at ORNL appear to be highly uncertain. One report (U.S. Department of Energy, 1987) indicates that 270 Ci of alpha-emitting radionuclides have been buried. This inventory includes ^{233}U as well as TRU radionuclides, but the proportions are unknown; we also assume that it does not include liquid wastes placed in the pits and trenches.

It is difficult to estimate the inventory of TRU radionuclides placed in the pits and trenches. In particular, the data reported by Olsen *et al.* (1986) for ^{241}Am in Trench 7, which could be used in providing such an estimate, are erroneous (J. R. Trabalka and C. R. Olsen, private communication). Therefore, we obtain a crude estimate as follows.

We assume that the relative inventories of TRU radionuclides placed in the pits and trenches can be represented by the inventories in sludges in the Melton Valley Storage Tanks, since both materials were generated at about the same time and by the same processes (J. R. Trabalka, private communication). Data reported by Peretz *et al.* (1986) indicate that the total activity of TRU radionuclides in sludges in the storage tanks is about 3.5 times the activity of ^{239}Pu and ^{240}Pu. We further assume that the inventory of ^{239}Pu is significantly greater than that of ^{240}Pu. Then, based on the data for ^{239}Pu in the pits and trenches reported by Trabalka (1987), we estimate a total activity of TRU radionuclides in the pits and trenches of about 180 Ci. A similar but slightly lower estimate would be obtained using the inventories of ^{239}Pu in the pits and trenches reported by Nix *et al.* (1986).

If we assume 270 Ci of TRU radionuclides in solid wastes and 180 Ci in the pits and trenches, then the estimated total inventory is about 450 Ci. Therefore, the release rate for the period 1971-1985 may be estimated as follows:

All sites, 1971-1985 -

Release rate = $(0.04 \text{ Ci/y})/(450 \text{ Ci}) \simeq 1 \times 10^{-4} \text{ y}^{-1}$.

Again, this estimate is highly uncertain, but it may be reasonable to conclude that the release rate of TRU radionuclides from ORNL burial sites is very low, and that estimates of future doses to inadvertent intruders for these radionuclides can be based primarily on considerations of the initial inventories of TRU radionuclides at disposal, with appropriate corrections for radioactive decay to be included only when they are significant (e.g., for ^{241}Pu and ^{244}Cm).

4.5 *Release Rates for Uranium*

As in the case of TRU radionuclides, estimates of release rates of uranium from ORNL disposal sites are highly uncertain, due to the apparent paucity of data.

Data given by Oakes *et al.* (1987a) indicate that annual discharges of uranium from White Oak Dam for the period 1982-1986 averaged about 0.6 Ci. Olsen *et al.* (1986) estimate that about 3 Ci of uranium, mostly ^{233}U, were placed in Trench 7. If we assume that the activity of uranium in all pits and trenches is proportional to the total volume of wastes emplaced, and using the estimate of Spalding (1987) that the total volume of liquid in all pits and trenches is about a factor of 5 greater than the volume in Trench 7, then we estimate that about 15 Ci of uranium was placed in the pits and trenches. Boegly *et al.* (1985) report that about 440 Ci of uranium, mostly ^{233}U and ^{238}U, was placed in SWSA 6 during the period 1977-1984. Thus, if we again assume that disposals of uranium in SWSA 6 have been constant over the operating lifetime, which is a highly speculative assumption given the erratic pattern of disposals (Davis and Solomon, 1987), then the total disposals in SWSA 6 are estimated to be about 900 Ci. No information appears to be available on disposals of uranium in the other SWSAs.

Thus, we can account in a crude way for a total disposal of about 900 Ci of uranium. Then, the fractional release rate in recent years is estimated as follows:

All sites, 1982-1986 -

Release rate = $(0.6 \text{ Ci/y})/(900 \text{ Ci}) \simeq 7 \times 10^{-4} \text{ y}^{-1}$.

Again, this estimate is probably highly uncertain, due to uncertainties in the data on releases from White Oak Dam and the possibility of significant releases from ongoing operations at ORNL, as well as the lack of information on the inventories of uranium at various sites. In addition, in connection with the analysis for ^{90}Sr in Section 4.1, we discussed the possibility that releases from SWSA 6 to surface waters have not yet reached steady state, which further clouds the validity of the estimated release rate. If this result has any credibility, then it supports the suggestion that uranium from the pits and trenches is more mobile in the environment than ^{90}Sr (Olsen *et al.*, 1986). However, whether similar comparative behaviors would be observed in releases from the SWSAs is an open question. In any event, release rates of uranium may be sufficiently low that doses to future intruders can be based primarily on considerations of the inventories of uranium at disposal.

4.6 *Other Long-Lived Radionuclides*

Discharges from White Oak Dam for such other important long-lived radionuclides as ^{99}Tc and ^{232}Th also have been reported (Oakes *et al.*, 1987a), and the average yearly releases for the period 1982-1986 are about 4 Ci and 0.007 Ci, respectively. It is also noteworthy that releases of ^{99}Tc over this short time period varied over the range 0.02-17 Ci. Some information is also available on the inventories of these radionuclides in SWSA 6 (Boegly *et al.*, 1985); the inventories for the years 1977-1984 are about 40 Ci and 3 Ci, respectively. However, no information appears to be available on disposals of these radionuclides at other sites.

If we again assume that total disposals of radionuclides in SWSA 6 are twice the reported values for 1977-1984, we can estimate what are presumably crude upper limits for release rates of ^{99}Tc and ^{232}Th from all sites:

^{99}Tc, all sites, 1982-1986 -

$$\text{Release rate} < (4\ \text{Ci/y})/(80\ \text{Ci}) \simeq 5 \times 10^{-2}\ \text{y}^{-1};$$

^{232}Th, all sites, 1982-1986 -

$$\text{Release rate} < (0.007\ \text{Ci/y})/(6\ \text{Ci}) \simeq 1 \times 10^{-3}\ \text{y}^{-1}.$$

These results agree qualitatively with the notion that ^{99}Tc may be relatively mobile in the environment, whereas ^{232}Th may be relatively immobile. However, we would regard the results as so speculative as not to warrant any definitive conclusions.

5. Conclusions

In spite of the speculative nature of the analysis of source terms for ORNL burial grounds presented here, some conclusions may be drawn from the results. These are briefly summarized as follows.

- It is likely that both ^{90}Sr and ^{137}Cs, which have been disposed of in large quantities, have been retained to a high degree within the boundaries of ORNL.

- Tritium appears to be highly mobile in the environment, and past disposal practices probably have not provided significant retention of this radionuclide.

- Transuranic radionuclides probably have been retained to a high degree within the boundaries of ORNL, but the evidence for this is considerably less convincing than in the case of ^{90}Sr and ^{137}Cs because of the lesser information available.

- The available data for uranium, ^{99}Tc, and ^{232}Th do not appear sufficient to perform a credible analysis of release rates from ORNL burial grounds.

On the other hand, it seems clear that reliable data for estimating radionuclide source terms from ORNL disposal sites are not available. The development of reliable data on inventories of radionuclides at disposal sites clearly is of great importance. Such data, when combined with estimates of discharges from White Oak Dam, could provide estimates of release rates of radionuclides from all burial sites combined. Even such rudimentary data, if reliable, could provide useful information for validation studies on models for radionuclide mobilization and transport. Analyses of the source term for individual sites would require more detailed information on hydrologic conditions and radionuclide concentrations in water at those sites.

Collection of data for estimation of radionuclide source terms should be performed over an appreciable period of time, perhaps over a period of a few decades. Data over long time periods may be needed to provide reliable estimates of source terms because present release rates from disposal facilities may not provide reliable indicators of future performance.

Finally, it is important to note that even though some of the important radionuclides appear to have been retained to a high degree within the boundaries of ORNL, the data for estimating source terms often were obtained at locations far from disposal sites. Thus, it is not necessarily the case that the radionuclides have been retained within disposal facilities themselves. For example, the so-called bathtub effect has caused significant movement of radionuclides from burial trenches in SWSA 4 (Melroy *et al.*, 1986), and uptake by terrestrial vegetation could be an important vector for transport of some radionuclides (Williams *et al.*, 1987). Therefore, a proper coupling of models of radionuclide mobilization and environmental transport is required to obtain reliable estimates of releases beyond the boundaries of disposal facilities.

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