Conf-911114-4

BNL-NUREG-46766

EVALUATION OF THE DATA AVAILABLE FOR ESTIMATING RELEASE RATES FROM COMMERCIAL LOW-LEVEL WASTE PACKAGES'

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BNL-NUREG--46766

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ABSTRACT

Information on the inventory, waste stream, waste form and containers used in LLW disposal has been obtained primarily from information compiled for the Nuclear Regulatory Commission (NRC) from shipping manifests which accompanied wastes disposed at the three currently operating commercial disposal sites (Barnwell, SC, Beatty, NV, and Richland, WA) during the period of 1987 - 1989. These data have been reviewed in order to determine the total activity distribution by waste class, waste stream and wasteform. The 1989 Richland shipping manifest data have been evaluated in more detail, including information on a radionuclide-specific basis. This Richland 1989 data have been compiled into a database that contains the waste stream, waste form, classification, half-life, annual limit for intake and activity for each radionuclide. Data from the other disposal sites and other years were insufficient for this grouping. This database has been compiled in terms of the distribution of activity for each radionuclide by waste stream and wasteform. This review evaluates these data in terms of the specific needs for improved modeling of releases from waste packages.

INTRODUCTION

Performance assessment of low-level waste (LLW) disposal facilities depends, among other things, on the ability to predict the rate of radionuclide release from the waste packages. For releases through the liquid pathway, modeling release rates requires knowledge of the length of time that the containers prevent water from contacting the waste form and on the waste form release characteristics which depend on the chemical form of the waste. Currently, most performance assessment methodologies treat release from the waste package in a general manner and take little account of the actual waste container/wasteform characteristics. To improve upon this, accurate data on the inventory, chemical form (i.e., waste stream and waste form), and container that comprise the waste packages are needed. This review evaluates the available inventory disposal data in terms of the specific needs for improved modeling of releases.

This work sponsored under the auspices of the Nuclear Regulatory Commission, Office of Nuclear Materials Safety and Safeguards.



Information on the inventory, waste stream, waste form and containers used in LLW disposal has been obtained primarily from information compiled for the Nuclear Regulatory Commission (NRC) from shipping manifests which accompanied wastes disposed at the three currently operating commercial disposal sites (Barnwell, SC, Beatty, NV, and Richland, WA) during the period of 1987 - 1989.¹ This Nk⁺ report provides a compilation showing the volume, activity, and radionuclide distributions of these wastes, and although some evaluation of the data is presented, the report was prepared primarily as a data source for use by others.

The 1989 Richland shipping manifest data¹ have been evaluated in more detail and compiled into a database that contains the waste stream, waste form, classification, half-life, annual limit for intake² and activity for each radionuclide. Data from the other disposal sites and other years were insufficiently detailed to permit such a grouping. This database has been evaluated in terms of the distribution of activity for each radionuclide by waste stream, wasteform and waste container.

In addition to reviewing existing manifest data, two other areas have been examined in terms of data sufficiency. These are: a) accuracy of the manifest inventory data for key radionuclides such as I-129 and Tc-99; and b) the availability of release rate parameters pertaining to cement solidification agents, sorbent materials, and key waste streams that frequently do not require a solidification/sorbent media.

In the next section of this paper, an overview of our findings concerning the distribution of activity within LLW will be presented. This will begin in a general fashion and consider the distribution of the total activity by each of the following: waste class, waste stream, wasteform, and waste container. A radionuclide specific breakdown by waste class and wasteform follows. The findings are reviewed in terms of performance assessment modeling needs. Finally, we present our conclusions.

DATA EVALUATION

After low-level waste has been generated it may be treated to, among other things, improve handling capabilities, remove free liquids, or provide radiation shielding. Some of the more widely used treatment options include stabilization in accordance with the stability criteria in the NRC's technical position on wasteforms,³ solidification in an agent that is not required to have those stability properties, use of sorbents to remove free liquids, compaction and dewatering.

After treatment, the waste is placed in a container. For Class B and Class C wastes where the wasteform does not meet the NRC's stability criteria, a high integrity container (HIC) must be used. For other wastes, carbon steel drums or liners are typically used.

In reviewing the data from the three sites it became clear that the sites do not have a common manifest information system and they store data in different formats. The differences are discussed in detail elsewhere.^{1,4} For this discussion, the two important differences that arise are:

Barnwell (operated by Chem-Nuclear Systems, Inc.) stores manifest information as summarized across entire shipments whereas Beatty and Richland (both of which are operated by U.S. Ecology, Inc.) record individual container information.

Waste streams are described differently, depending on the manifest specified by the disposal site operators. The U.S. Ecology sites have a more detailed breakdown on waste streams.

The net result of these differences is that considerably more detailed information is available on the waste delivered to the U.S. Ecology sites as compared to the Chem-Nuclear site. However, the Barnwell site received over 60% of the volume and 82% of the activity for the three year period under study. Consequently, some of the analysis discussed later on in this paper which are based only on the information at the Richland site in 1989 may not necessarily be representative of the nationwide data.

In reviewing the U.S. Ecology manifest forms it is noted that the wasteform is described by a numerical key to indicate if the waste stream is treated with a stabilization or sorbent media. Treatment of the waste is not typically required for several waste streams including equipment components, dewatered resins, dry active waste, and dry solids. In this case, the key indicates "none required" for the treatment option.

When treatment is used, ten different stabilization agents and twenty-three different sorbents were identified at the U.S. Ecology sites. To simplify the analyses, we have consolidated all of the cement-based stabilization agents into a single category which we call "cement." Similarly, the sorbents are all grouped into a category called "sorbents." Everything else is placed in the category "none required." For this analysis, the "cement" category does not include bitumen, gypsum cement, or vinyl ester styrene wasteforms. These three types of wasteform contributed much less than 1% of the total activity at the U.S. Ecology sites and have been included in the "none required" category.

Distribution of Activity and Volume by Waste Class

For the period of 1987 - 1989 the nationwide percentage of activity disposed of as Class C wastes was 73%. Class B wastes contained approximately 21% of the activity. Class A wastes contained the remaining 6%. Class A wastes were further categorized by whether the wasteform met the stabilization criteria in the NRC's technical position. 2.6% was classified as A-unstable and 3.4% was A-stable.

During this period, over 82% of the activity was disposed at Barnwell. Therefore, the Barnwell distribution is similar to the nationwide distribution. At the Richland and Beatty sites, the distribution was different. The majority of the activity was found in Class B wastes. Class A wastes were primarily A-unstable and accounted for 15% of the activity at Beatty and 8% at the Richland site.

In contrast, the majority of the volume (>96%) of the wastes are in Class A wastes. The volume of Class B wastes was 2.3% of the nationwide inventory and Class C wastes were contained in 0.75% of the volume.

Table 1Distribution of Activity by Waste Class at the Disposal sites for the years1987 - 1989 (in Percent)							
	Barnwell	Beatty	Richland	Total			
A-Unstable A-Stable A-Total B C Total	0.99 3.26 4.25 9.2 68.8 82.2	0.5 0.18 0.68 3.4 0.53 4.6	1.12 0.0005 1.12 8.3 3.72 13.2	2.6 3.4 6.05 20.9 73.1 100.0			

A detailed breakdown of the volume and activity by waste class has been presented.¹ The distribution by activity for each site for the three year period is also presented in Table 1.

Distribution of Activity by Waste Stream

One point one million curies were disposed at the Barnwell site from 1987 - 1989 in twelve different waste streams. Of these, only five contributed more than 1% of the activity. By far the most significant contribution (over 80%) to activity during this period came from waste described as "Equipment Components." Practically all of this waste stream was judged to be Class C. "Resin" wastes contributed over 12% of the total activity, but this activity was spread evenly over the three classifications, A, B, and C. Activity (principally Class B) of "Solid Non-Combustibles" comprised 4.7% of the total while another 1.4% was categorized as "Combustible plus Noncombustible" (again mostly Class B). "Filter media" contained about 1% of the activity. The other 7 waste streams contributed less than 1% of the total activity.

Sixty-two thousand curies were disposed at Beatty during the period 1987 - 1989 in twenty-one different waste streams. Over 81% of the activity was contained in waste described as "Dry Solid," 4% from "Non-Cartridge Filter Media," and 3.5% from "Solidified Resins." "Solidified Liquids" and "Evaporator Bottoms" each contributed about 3% of the activity but, for the latter, this was accumulated in just one year (1989). Other waste streams that comprised at least 1% of the activity were "Gas," "Activated Reactor Hardware" (1989 only), and "Compacted Dry Active Waste." Most of the "Dry Solid" was categorized as Class B; however, 10% of it qualified as Class C. All the "Activated Reactor Hardware" was Class C, as was about a third of the "Compacted Dry Active Waste" and "Solidified Liquids."

One hundred seventy eight thousand curies which came from twenty-two different waste streams were disposed at Richland over the same period. The major contributors to the activity were described as "Dry Solid" (45.9%), "Solidified Liquids" (24%), "Dewatered

Resins" (13.2%) and "Activated Reactor Hardware" (12.4%). The largest proportion of the combined activity of these four waste streams was Class B, but one of the waste streams ("Activated Reactor Hardware") was almost 100% Class C. "Filter Media," "Solidified Resins," and "Compacted Dry Active Waste" each contained between 1 and 2% of the total activity at the Richland site.

Normalizing the waste stream inventory information from each site to the nationwide inventory and combining activities from the three sites when appropriate (e.g., "Equipment Components" and "Activated Reactor Hardware") results in 67.5% of the activity in "Equipment Components," 12.3% in resins (Dewatered or Solidified), 9.8% in "Dry Solid," 3.9% in "Solid Non-Combustibles," 3.3% in "Solidified Liquids," and approximately 1% in each of "Filter Media," and "Combustibles and Non-Combustibles." This distribution by waste stream at the three sites is presented in Table 2. A detailed breakdown of the activity in each waste class, waste stream, and site can be found in Reference 4.

	Barnwell	Beatty	Richland	Total
Equip. Comp ¹	65.8	0.08	1.6	67.5
Resins ²	10.2	0.2	1.9	12.3
Dry Solids	_3	3.7	6.1	9.8
Solid Non-Comb	3.9	-	-	3.9
Solidified Liq	-	0.1	3.2	3.3
Filter Media	0.8	0.2	0.2	1.2
Comb + Non-Comb	1.1	-		1.1
Other				0.9
Total				100.0

Reactor Hardware" waste streams.

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² Includes dewatered and solidified resins.

³ A dash indicates that this waste stream identifier was not used at this site.

Distribution of Activity by Wasteform

Only limited information is available on the wasteforms disposed at the Barnwell site. That which is presented treats the waste in terms of volume disposed and not activity. A review of the data indicates that the volume percentage of wastes solidified in cement at Barnwell decreased from 14% in 1987 to 7% in 1989. "Solidified Liquids," "Resins," "Filter Media," and "Solid Non-combustibles" were the waste streams that had the most volume encased in cement. "Solidified Liquids" accounted for approximately 1/2 and "Resins" accounted for 1/3 of the volume in cement.

Approximately 80% of the activity disposed at Barnwell is in the form of "Equipment Components" and this waste stream does not typically require solidification. Therefore, less than 20% of the activity is solidified at Barnwell. Further, only 20% of the volume of "Resins" which contain 12.4% of the activity were solidified in cement. It is likely that less than 10% of the activity is disposed in cement-based material at Barnwell.

At the U.S. Ecology Sites information is available on the solidification and sorbents used to treat the wastes. Over 75% of the activity at Beatty was solidified in cement-based material during the period 1987 - 1989. 24% of the waste activity was in the category "none required" and 0.6% of the activity was treated with sorbents. At the Richland site, almost 71% of the material was contained in "none required," 27.7% was in "cement," and 1.5% was in "sorbent."

Assuming that 10% of the activity is stabilized in cement at the Barnwell site gives a nationwide average of 16% of the activity in cement. Approximately 1% of the activity is treated with a sorbent and the remaining 83% is in the category "none required."

Distribution of Activity by Waste Container

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The Barnwell site records information by waste shipment and not container. Therefore, there is no information on the activity within each container.

At the U.S. Ecology sites the inventory is listed by container. However, the only information available on the container is the type (drums, boxes, cask liners, or others) and its volume. The container material and the thickness of the container wall are not listed. However, the manifest did specify if a HIC was used, but did not tell which type of HIC.

The number of containers received at the U.S. Ecology sites during 1987 - 1989 was 143,223. Over 75% of these had a volume of 7.5 ft³, i.e., the volume of a 55 gallon drum. The range of container sizes was 0.02 - 1450 ft³.

Although not reported in Reference 1, theoretically the manifest data could be used to determine the activity of each radionuclide within each container at the U.S. Ecology sites. The usefulness of this information is limited by the lack of knowledge on the container materials and thickness.

DISTRIBUTION OF INDIVIDUAL RADIONUCLIDES

During 1987 - 1989 almost 300 radionuclides were identified as being disposed of at the three disposal sites. Many of these have half-lives less than 1 year or have inventories less than 1 millicurie. In these cases, it is unlikely that the radionuclides will cause a significant dose to the general public. To prioritize the need to examine each of the 300 different radionuclides a screening criterion is needed. Such a criterion has been proposed.⁴ This criterion takes the disposal inventory, multiplies it by a half life dependent factor, and divides the result by the maximum annual limit for intake recommended for radiation workers.² The exact expression is discussed in Reference 4. For this paper, we will divide the radionuclides into two classes according to their half-life. The dividing point for the classes will be the half-life of Cs-137. Radionuclides with a half-life less than or equal to that of Cs-137 will form one class. The reason for this distinction is that the shorter half-life class will undergo substantial decay during the 300 year period required for structural stability of the wasteform/container system. Further, as we will show, the distribution of radionuclides with a long half-life is markedly different than for those with a short half-life.

In this analysis, the screening criterion was applied to the Richland, 1989 data as this was the only data that contained a detailed breakdown of the inventory by waste stream and wasteform. The following radionuclides were determined to have the highest ranking for the short half-life class: Sr-90, Cs-137, Co-60, Cs-134, Fe-55, and H-3. These radionuclides contain more than 94% of the activity. It is interesting to note that tritium had over 60% of the inventory at Richland during 1989, yet it ranks sixth on the list due to its higher maximum annual limit for intake. For the long half-life class, Th-232, U-238, Ra-226, C-14, Am-241, Pu-239, I-129, and Tc-99 were identified as radionuclides with a potential for a large impact on performance assessment. The activity of all of these nuclides comprise 0.5% of the inventory. Ni-63 which contains the bulk of the long half-life group activity, approximately 2% of the total inventory at this site, did not make the list because of its relatively high annual limit for intake.

Distribution of Radionuclides by Waste Class

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In 1989 at Richland, 6.6% of the activity was class A-unstable waste. For the short half-life group, this distribution was approximately followed. The percentage amount of 5 of the 6 radionuclides identified earlier ranged between 2 and 8%. There was no Class A Sr-90. In contrast, for the long half-life group, most of the activity is Class A-unstable. 100% of the Th-232 and U-238 are in this category. 74% of the I-129, 62% of the C-14, 43% of the Ra-226, and 38% of the Tc-99 are also Class A-unstable.

Distribution of Radionuclides by Wasteform

As previously discussed, a review of the data indicated three major classes of wasteform: "cement," "sorbent," and "none required." The distribution by wasteform follows.

At the Richland disposal site in 1989, only 3% of the activity was solidified in cement. This contrasts markedly with the previous two years when over 50% of the activity was contained in cement. For the short half-life group, with the exception of Cs-134 which had 8% solidified in cement, the others had less than 4% in cement. The distribution of the long half-life group was markedly different. Over 60% of the Ra-226, 40% of the C-14 and Am-241, and 27% of the U-238 are in cement.

The amount of wastes treated with sorbent amounted to 1.6% of the activity in 1989 at Richland. Essentially none of the short half-life radionuclides considered received this treatment. Fe-55 had 0.9% in sorbents and all of the rest had less than 0.5% treated with sorbents. The opposite was true for the long half-life group. Over 80% of the Th-232 was

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treated with sorbents, as were 15% of the I-129, 6% of the U-238 and C-14, and 4% of the Tc-99 and Am-241.

The amount of activity in "none required" wasteforms was greater than 95% during 1989 at the Richland site. For the short half-life group, Cs-134 had 92% of its activity in this category while all others had more than 95%. Most of the long half-life group also had the majority of their wastes in the "none required" category. However, most were far less than 95% and only 14% of the Th-232 was in this category.

The distribution of these fourteen radionuclides by waste class and wasteform is presented in Table 3. For each nuclide, the final three columns total 100%. For this data set, wastes that are not Class A-unstable are either Class B or Class C.

Table 3Distribution of Radionuclides disposed at Richland in 1989 by Class and Wasteform (in Percent)								
Long half-life group								
	A-Unstable	Cement	Sorbent	None				
C-14 Tc-99 I-129 Ra-226 Th-232 U-238 Pu-239 Am-241	62.4 37.6 73.8 42.8 100 100 12 32.8	43.3 0.0 0.4 62.4 5.0 27.3 2.6 41.3	5.9 3.9 15 3.6 80.5 6.1 3.5 4.6	50.8 96.1 84.6 33.9 14.5 66.6 93.9 54.1				
Short half-life group								
	A-Unstable	Cement	Sorbent	None				
H-3 Fe-55 Co-60 Sr-90 Cs-134 Cs-137	4.0 7.3 7.8 0.0 4.7 2.0	4.0 1.6 3.3 0.0 8.1 2.0	$\begin{array}{c} 0.4 \\ 0.9 \\ 0.5 \\ 0.0 \\ 0.1 \\ 0.0 \end{array}$	95.6 97.5 96.2 100 91.8 98.0				

DISCUSSION

Availability of Release Rate Data

The fact that 83% of the commercial waste disposed of in the U.S.A. for the threeyear period is in the form "none required" indicates that information is needed on the release characteristics of the waste streams. The waste streams that typically do not require treatment include "Equipment Components," "Dewatered Resins," "Dry Solids," "Solid Non-Combustibles," and "Combustibles and Non-Combustibles." Each of these waste streams contribute more than 1% of the nationwide inventory.

Unfortunately, there is very little information on the release rate from any of the above waste streams. Therefore, the modeler is left to guess this key parameter. In some cases, the release rates are expected to be quite low and their value may be estimated from other systems. For example, "Equipment Components," which contain over 67% of the nationwide activity, are often made of stainless steel which is known to corrode slowly in soil systems. Similarly, the release rate of stainless steels in concrete vaults characterized by high pH may be estimated by rebar corrosion data. However, there will be a high degree of uncertainty in applying corrosion data taken from one system to another system because of all of the uncontrolled variables that may influence corrosion. Further, the possibility of preferential release of constituents has not been examined.

Other waste streams, e.g., "Dry Solids," "Solid Non-Combustibles," and "Combustibles and Non-Combustibles," are poorly defined at this time. These waste stream descriptions are inadequate for modeling purposes because they do not define the physical and chemical form of the waste. Therefore, the modeler is left with the need to conservatively estimate their release rates. These waste streams contain approximately 15% of the nationwide inventory.

For waste streams treated with sorbents, once again there is little information on release rates from these wasteforms. Many sorbents are essentially inert fillers, e.g., diatomaceous earth and silicates, and will have only a minor effect on the release of radionuclides. However, this question has not been experimentally addressed. Although only 1% of the total inventory is treated with sorbents, knowledge of release rates may be important because many of the long-lived radionuclides are treated with sorbents. For example, 80% of the Th-232 disposed at Richland in 1989 was treated with a sorbent and was Class A-unstable.

For waste streams solidified in cement, approximately 16% of the nationwide activity, there have been a large number of leaching experiments conducted. This experimental database is heavily focused on Cs, Sr, and Co. Unfortunately, as shown previously, only a small fraction of their inventory is solidified in cement. Furthermore, performance assessment calculations indicate that these nuclides generally do not significantly contribute to the dose to man. The long-lived nuclides which often contribute to the dose to man and were contained in significant fractions (at Richland in 1989) in cement include C-14, Ra-226, U-238, and Am-241. However, there is very little information on the release rates of these elements from cement waste forms.

Even though there is a substantial database for leaching of several radionuclides from cement, there may be a high degree of uncertainty in the release rate parameters. Typically, diffusion is believed to be the rate controlling process from cements. The measured apparent diffusion coefficients for a single nuclide in different cement formulations may vary by several orders of magnitude depending on the cement aggregate, water-to-cement ratio, the

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presence of chelating agents or other factors.⁵ Therefore, care must be exercised if the modeler uses diffusion coefficient values taken from the literature on a system that differs from the one under study.

Inventory

The inventory data presented in this report are from the shipping manifests received at each of the three disposal sites. These data have been taken to be exact and no attempt has been made to determine the actual accuracy.

In practice, it is likely that the activity level of the radionuclides that are major contributors to total activity (e.g., Cs-137, Sr-90, Co-60, H-3, etc.) are measured with a reasonable degree of accuracy. These radionuclides are usually detected directly and occur at substantial levels. However, the activity of minor radionuclides such as Tc-99 and I-129 have a large degree of uncertainty in their measurement. The activity level of these radionuclides are often near or at detection limits and are often estimated by scaling ratios with other radionuclides.

These scaling ratios for Tc-99 and I-129 from power plant ion exchange resin wastes are believed to be at least 2 - 4 orders of magnitude too large.⁶ Therefore, their estimated inventory in these wastes are 2-4 orders of magnitude larger than their actual inventory. During the period 1987 - 1989 over 95% of the I-129 and 66% of the Tc-99 came from power plant wastes. Much of this activity was contained on resins.

I-129 and Tc-99 are important contributors to the dose to man for many release scenarios from LLW disposal sites because of their high mobility and long half-lives. In many scenarios, I-129 is the leading contributor to the dose to man. If their inventories are incorrectly overestimated by a few orders of magnitude, as believed for power plant wastes, the predicted dose will be overestimated by a similar amount. For this reason it is crucial to obtain accurate measurements of the activity levels for these isotopes from all sources.

Container Lifetime

As a minimum, the container thickness and material, e.g., carbon steel, stainless steel, HDPE, etc., must be known before an estimate on container life time can be made. This information is not available from the shipping manifests. Therefore, estimates on container lifetime are at best educated guesses.

Distribution of Activity by Waste Container

In general, a HIC is expected to prevent radionuclide release for a longer time period than a carbon steel drum. Therefore, when modeling releases, it would be appropriate to know the distribution of radionuclides by container. The activity in a particular container is reported on the U.S. Ecology manifests, however, it is not available in Reference 1. To portion the radionuclides into containers with the available information, the modeler must make assumptions for everything. For example, a) Class A wastes could be assumed to be in carbon steel containers; b) Class B and C wastes with a waste treatment option of "none required" could be assumed to be stored in HIC's in order to meet the structural stability requirements in 10 CFR 61; and c) Class B and C wastes with stabilized wasteforms could be assumed to be placed in carbon steel liners, etc. However, the usefulness of these assumptions is limited by lack of information pertinent to estimating container lifetime.

CONCLUSIONS

The available radionuclide data for 1987 - 1989 inventory received at the three commercial disposal sites have been evaluated in terms of the distribution by waste stream, wasteform, and waste container. These data were reviewed in an attempt to determine data needs for performance assessment modeling.

The distribution of activity by waste stream and wasteform differ dramatically from site to site and year to year. For this reason, whenever possible, the three-year average for all three sites was used in determining the distribution of the radionuclides. This was not possible for the detailed breakdown of each radionuclide by wasteform and waste stream. In this case, only the Richland 1989 data were sufficiently detailed.

Based on our review the following improvements to the database are needed to improve performance assessment:

- a) There is a need for an improved shipping manifest. The manifest should include more information on the container, e.g., the container material and thickness. The information provided is insufficient for estimating the time that the container prevents moisture from contacting the wasteform. Also, the distribution of radionuclides within each container should be available. Currently, any attempt to determine the distribution of radionuclides within a class of containers (e.g., HIC, carbon steel drum, etc.) requires significant assumptions. This information could be obtained through better record keeping on the shipping manifest.
- b) Better characterization of the wasteform is required. Over 83% of the activity of wastes in this country are disposed in "none required" wasteforms. When treatment of the wastes are not required, once the container is breached, the untreated waste becomes the barrier to release. There is little information on the release parameters appropriate for untreated wastes, e.g., "Equipment Components," "Dewatered Resins," "Dry Solids," and "Combustibles and Non-Combustibles."

c) Better characterization of the inventory is needed for minor constituents with long half-lives and the potential to cause relatively high predicted

doses to the general public, e.g., I-129 and Tc-99. These radionuclides are difficult to measure directly and are often conservatively estimated using scaling factors.

Two other important observations are:

d) The distribution of long half-life radionuclides is markedly different than for short half-life radionuclides. Based on the Richland 1989 data, the long half-life radionuclides are primarily in Class A wastes and are often found in sorbents or cements in higher proportions than average. Short half-life radionuclides tend to be Class B and C, typically are not treated with sorbents.

e) Resources should be focused on obtaining relevant release information from the wastes containing radionuclides that provide the largest risk to the public. These nuclides are typically long-lived and have a distribution within the wastes as discussed in d).

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