SOURCE TERM EVALUATION FOR PERFORMANCE ASSESSMENT OF LLW DISPOSAL

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ABSTRACT

Information compiled on the low-level radioactive waste disposed at the three currently operating commercial disposal sites during the period 1987-1989 have been reviewed and processed in order to determine the total activity distribution in terms of waste stream, waste classification and waste form. The data from waste disposed during 1989 at one of the sites (Richland, WA) were more detailed than the data available during other years and at other sites, and thus were amenable to a more in-depth treatment. This included determination of the distribution of activity for each radionuclide by waste form, and thus enabled these data to be evaluated in terms of the specific needs for improved modeling of releases from waste packages.

INTRODUCTION

Assessment of the performance of low-level radioactive waste (LLW) disposal facilities depends, among other things, on the availability of a radionuclide inventory (source term) evaluation methodology which can be applied to specific waste streams. Methodologies currently in use tend to treat this source term in a general manner and take little account of the characteristics of the original waste streams or of the forms in which the wastes are being disposed. These two factors, in practice, are often related, in that the characteristics of the waste stream tend to dictate the form in which the waste is finally disposed. The final form is the most important in determining the release mechanisms and rates of the individual isotopes. Thus the development of a methodology which incorporates information on the characteristics of the waste streams and, particularly, the waste forms will lead to an enhancement of the current ability to assess LLW disposal facility performance.

This paper describes the results of a preliminary assessment to determine where the emphases should be placed in constructing a source term evaluation methodology which takes into consideration waste characteristics (for example, the chemical composition, the disposal form of the waste and the container system). It also identifies areas where lack of detailed information severely compromises the development and implementation of the methodology. The assessment itself is based primarily on information compiled by Roles for the U.S. Nuclear Regulatory Commission (NRC) (1) from shipping manifests which accompanied LLW disposed at the three currently operating commercial disposal sites (Barnwell, SC, Beatty, NV, and Richland, WA) during the period 1987 through 1989. Roles provides essentially a compilation showing the volume, activity and radionuclide distributions of this waste, which itself is variously defined in terms of the original waste stream, the waste form immediately prior to disposal and the waste classification. Although some evaluation of the data is presented, the report was prepared primarily as a data source for use by others. In the present paper we have attempted to evaluate those data in terms of the specific needs of the source term modeler, with emphasis being placed on the activity of the wastes and on the distribution of radionuclides in the different sorbent and solidification media.

DATA EVALUATION

Data Limitations

A major problem encountered in evaluating the data available (and which was highlighted by Roles) is that, currently, there is no common shipping manifest information system. The manifests which accompanied the waste shipments described in (1) were developed by the existing disposal site operators, Chem-Nuclear Systems, Inc., at Barnwell and U.S. Ecology, Inc., at Beatty and Richland. Although both types of manifest contain all the basic information currently required for regulatory purposes, they are not interchangeable, each having its own unique reporting characteristics. Thus, for example, Barnwell stores the information as summarized across entire shipments whereas Beatty and Richland record individual container information. The latter two sites also have a more detailed breakdown on the waste streams. These differences result in more detailed information being available on the waste delivered to the U.S. Ecology sites compared with that received at the Chem-Nuclear site. However, as will be shown later, Barnwell received far more waste than the other two sites combined for the three year period under study. Thus some of the more detailed analyses presented in the present paper are, of necessity, probably not representative of the nationwide data.

With respect to the lack of a common manifest system, it should be noted that efforts are now being made at the NRC to develop a uniform manifest (2). Implementation of such a manifest reporting system will certainly greatly simplify and improve the evaluation of wastes disposed in the future. At the same time, it is recognized that this may not be a simple task as the sited states and compacts are likely to have their own individual requirements.

It must also be pointed out that the data contained in the Roles compilation (1) is taken to be exact and no attempt has been made to determine the actual accuracy. That some imprecision exists is not in question but will not be discussed here. For further information on this matter, reference should be made to (1,3).

General Distribution of Waste Activity

The amount of LLW disposed at the three sites during the period 1987-1989 is shown in Table I, from which it can be seen that Barnwell received more waste, in terms of both

TABLE I

Volume and Activity of Waste Disposed at Commercial Sites during the Period 1987-1989

Facility	Volume (m^3)	Activity (MCi)
Barnwell	84680	1.166
Beatty	15340	0.062
Richland	38730	0.178

volume and activity, than the other two sites combined. In addition, site-to-site variations were noted in the activity distribution among the different waste streams and waste classifications, even when the information was averaged over the three year period (Tables II and III). Indeed, although it will not be discussed here, there were also variations from year to year at individual sites.

TABLE II

Activity Breakdown by Major Waste Streams Disposed at Commercial Sites During the Period 1987-1989

Facility	Waste Stream	% of Total Activity at Site
Barnwell	Equipment, components	79.8
	Resins	12.4
	Solid noncombustibles	4.7
	Others	3.1
Beatty	Dry solids	81.4
	Non-cartridge filter media	4.1
	Solidified resins	3.5
	Evaporator bottoms	3.0
	Solidified liquids	2.9
	Others	5.1
Richland	Dry solids	45.9
	Solidified liquids	24.0
	Dewatered resins	13.3
	Activated reactor hardware	12.4

At Barnwell, about 80% of the activity in 1987-1989 came from the disposal of activated equipment and components, practically all of which was designated Class C waste, a category which comprised over 83% of the total activity at the site. By contrast, both at Beatty and at Richland, Class B wastes contributed most of the activity. The major waste stream with *regard to activity at each U.S. Ecology* site was that termed "dry solid," though the actual percentages varied with the site (over 81% of the activity at Beatty came from "dry solid" but at Richland the portion was only 46%). More detailed breakdown of the information summarized above can be found in (1,3,4).

Distribution of Activity by Waste Form

Activity-based analyses of the form in which the waste has been disposed were performed only on the Beatty and Richland data, the information available in this area from Barnwell being limited to waste volumes. However, it is possible to construct an approximate distribution at Barnwell based on the information provided on the waste stream characteristics (3).

TABLE III

Activity Breakdown by Classifications of Waste Disposed at Commercial Sites During the Period 1987-1989

	Percent of Total Activity at Each Site		
Class	Barnwell	Beatty	Richland
AU (A, unstabilized)	1.20	10.81	8.47
AS (A, stabilized)	3.96	3.84	0.004
A (total)	5.08	14.65	8.47
BS (B, stabilized)	11.19	73.94	63.28
CS (C, stabilized)	83.73	11.41	28.25

For convenience, the numerous waste form categories found in the U.S. Ecology manifests were grouped into four simplified subcategories: cement-based; sorbents; none required or noted; and other. The "cement-based" group includes waste solidified in media such as "Structural Concrete," "Concrete (2500 psi)" and several commercial cement-based solidification products. "Sorbents" covers such commercial products as "Floor Dri," "Hi Dri," "Celaton," etc., most of which are silica- or silicate-based. The category "none required or noted" indicates that the original manifests either stated that the waste did not require any sorbent or solidification media or that none was noted in the appropriate space. "Other" media include bitumen and gypsum.

About 75% of the activity disposed at Beatty was in cement-based waste forms, in sharp contrast to Richland where less than 28% of the activity was in such waste forms (Table IV). At Richland, about 70% of the activity was contained in wastes for which mixing with sorbents or solidification media was not required or not mentioned.

Although no such direct breakdown can be provided for the Barnwell site, it should be noted that approximately 80% of the activity disposed there is contained in a waste stream described as "Equipment, Components." Typically, such a waste stream is unlikely to require solidification before disposal or the addition of sorbent media to reduce the amount of free liquid available. Thus it can be inferred that less than

TABLE IV

Activity Breakdown by Waste Treatment Media for Waste Disposed at Beatty and Richland Sites During the Period 1987-1989

Media	Percent of Total Activity at Site	
	Beatty	Richland
Cement-based	75.2	27.7
Sorbents	0.6	1.5
None required or noted	10.7	70.2
Other	13.5	0.6

20% of the activity received at this site received treatment with either a solidification or sorbent agent.

Distribution of Individual Radionuclides

The radionuclide inventories at the three sites were analyzed in terms of those isotopes which are relatively long-lived (half-life greater than about 30 years) and those which are relatively short-lived (half-life less than about 30 years). Over the three year period evaluated, the relatively short-lived isotopes together comprised over 95% of the total activity (Table V). H-3 contributed over 50% at both Beatty and Richland but was only a very minor portion of the activity at Barnwell. The major activity contributors at Barnwell were Fe-55 and Co-60. The long-lived isotopes constituted less than 5% of the disposed activity at each site, and more than half of this activity was attributable to Ni-63 (Table VI).

Only the data available on wastes disposed at Richland during 1989 contained sufficient detail to allow processing to determine which isotopes were contained in which waste forms and to what extent they were present in those waste forms. For the purposes of source term modeling, rather than simply rank the isotopes in terms of activity, these data were ordered using a relationship which takes into account the activity of the isotope, its half-life and its radiotoxicity properties:

$$(\text{Activity}) \times (0.5)^n = \text{ALI} \qquad \text{(Eq. 1)}$$

where ALI is the annual limit for intake by ingestion, the latest values of which can be found in (5), and n is the number of half-lives necessary to reduce the activity to the ALI value.

The value of n can be calculated by inserting the applicable Activity and ALI values in Eq. 1. Multiplication of n by the half-life of the isotope then gives the equivalent time it will take for the inventory to decay to a level which is equal to the annual limit for intake by ingestion of that particular isotope. That is, a determination can be made of which isotopes in the inventory will remain of concern (in terms of the allowable intake by ingestion) for the greatest lengths of time. However, it should be recognized that this represents a very simplified approach and does not attempt to take into account such factors as the mobility of the various radionuclides within the waste and after release to the surrounding environment. It also gives equal weight to the half-life of the isotope and to its ALI value. Thus extreme values of either property can give rise to inordinate importance being attached isotopes whose contribution to the overall activity may be very small.

TABLE V

Activity Breakdown by Short Half-life Radionuclides for Waste Disposed at Commercial Sites During the Period 1987-1989

Barnwell		Beatty		Richland	
Isotope	Percent	Isotope	Percent	Isotope	Percent
Fe-55	46.34	H-3	56.19	H-3	62.56
Co-60	32.59	Co-60	23.46	Fe-55	11.64
Mn-54	4.69	Cs-137	9.10	Co-60	9.26
H-3	2.78	Fe-55	6.72	Cs-137	4.80
Co-58	2.48	Mn-54	1.34	Sr-90	3.51
All	95.97	All	99.02	All	98.42

TABLE VI

Activity Breakdown by Long Half-life Radionuclides for Waste Disposed at Commercial Sites During the Period 1987-1989

Barnwell		Beatty		Richland	
Isotope	Percent	Isotope	Percent	Isotope	Percent
Ni-63	3.78	Ni-63	0.582	Ni-63	1.28
U-238*	0.105	U-238*	0.220	C-14	0.228
Th-232#	0.096	C-14	0.092	U-238*	0.036
C-14	0.029	Ra-226	0.065	Th-232#	0.022
Ni-59	0.016	Am-241	0.007	Ni-59	0.010
All	4.03	All	0.98	All	1.58

Notes: U-238* - includes natural uranium and depleted uranium
Th-232# - includes natural thorium.

The manifest data indicate that nearly 200 different radionuclides were disposed at the Richland site during 1989. Evaluation of these data using Eq. 1 led to a ranking which was used to eliminate many of these isotopes from further consideration because of very short half-life and/or very small radiotoxicity effect. The top-ranked of the remaining isotopes (that is, those thought to be most significant from the aspect of source modeling) are shown in Tables VII (short-lived isotopes) and VIII (long-lived isotopes). A complete ranking of all the isotopes is contained in (4). Tables VII and VIII reflect some personal judgement exercised in view of the limitations described above. It also needs to be emphasized that the distribution of radionuclides disposed at Richland during 1989 is not typical of the nationwide distribution or, for that matter, even the average distribution at that site over the period 1987-1989. These tabulations are not exclusionary and it is not intended to suggest that only those isotopes identified in Tables VII and VIII should be incorporated in a source term model.

Tables VII and VIII also indicate the activity distribution between the four simplified waste form categories in which each of these isotopes are disposed. Again, it must be pointed out that the distribution characteristics of the waste disposed at Richland in 1989 was significantly different from the average distribution over the three year period 1987-1989. For example, although cement-based media contained 27.7% of the activity disposed over those three years (Table IV), the equivalent in 1989 alone was only 3.5% ("none required or noted" comprised over 95% during that year).

Of the short-lived isotopes, by far the most significant isotopes appear to be Cs-137 and Sr-90 when account is taken of half-life and radiotoxicity in addition to activity level. It should be noted that this would not have been the case if attention had only been directed to their contribution to the activity of the total inventory (such a ranking would have placed H-3, Fe-55 and Co-60 ahead of them). For both Cs-137 and Sr-90, most of the activity was in Class B or C waste and has been disposed in a form which required no sorbent or solidification media or for which no such media were noted on the manifest. The principal waste streams involved were dewatered resins and compacted dry active wastes.

The same type of analysis when applied to the long-lived isotopes identified Th-232 as the most important for source term modeling, followed by U-238 (in terms of activity alone, Ni-63 would have been ranked ahead of both isotopes). The largest amounts of Th-232 (including natural thorium) were found in dry solid waste (unstabilized Class A) and were associated with sorbent media. By contrast, only a small portion of U-238 activity was associated with sorbent while more than a half of it fell into the "none required" category and over a quarter was disposed of in cement-based media.

TABLE VII

Distribution of Short Half-life Radionuclides in Waste Forms Disposed at Richland in 1989

	Waste Treatment Media			
Isotope	Cement	Sorbent	None Required	Others
Cs-137	2.0	0.0	94.0	4.0
Sr-90	0.0	0.0	99.99	0.0
H-3	4.0	0.4	95.0	0.6
Co-60	3.3	0.5	95.8	0.4
Fe-55	1.6	0.9	96.7	0.8
Cs-134	8.1	0.1	88.0	3.8

TABLE VIII

Distribution of Long Half-life Radionuclides in Waste Forms Disposed at Richland in 1989

	Waste Treatment Media			
Isotope	Cement	Sorbent	None Required	Others
Th-232	5.0	80.5	14.5	0.0
U-238	27.3	6.1	63.9	2.5
I-129	0.4	15.0	80.6	4.0
Tc-99	0.0	3.9	94.7	1.4
Pu-239	2.6	3.5	90.5	1.6
C-14	43.3	5.9	49.9	0.9

DISCUSSION

Waste Containment

That a substantial portion of the activity disposed during the period 1987-1989 can be categorized as not requiring any sorbent or solidification media or had none identified is of great concern from the source term modeling aspect. This implies that there is very little information on the actual forms in which this very large fraction (over three quarters) of the total activity was disposed. Some of this activity is traceable to activated hardware and components which may possibly have been disposed in their original condition. However, most, if not all, of the waste would have been placed in containers before disposal. The containers, in turn, constitute the primary barriers isolating the waste from the environment. A source term model needs to incorporate information of the effectiveness of such barriers; that is, the manner and rate of degradation of the containers must be known. The shipping manifests currently in use provide little information of relevance in this area. The uniform manifest being developed at the NRC (2) proposes to require more information on the containers and on their material(s) of manufacture, and should help considerably to rectify the problem.

Availability of Radionuclide Release Data

The waste streams that typically do not require treatment with sorbents or solidification media include equipment and components, dewatered resins, dry solids, etc. Although, as mentioned in the previous section, these waste streams make up a very large portion of the total activity disposed, very little information exists on the release of the various radionuclides from any of them. In some instances, it may be possible to make reasonable estimates based on general information. For example, the equipment and components are often made of stainless steel for which a considerable corrosion rate database exists. Other waste streams, however, such as the dry

solids, are very poorly defined and the modeler is left to his/her own devices in estimating (and justifying) release rates, which, perforce, must be conservative in nature.

Radionuclide Release from Cement-Based Media

It has been estimated that perhaps 16% of the activity disposed nationwide during 1987-1989 was solidified in a cement-based media (3). The problem of radionuclide release from such media has been addressed by extensive test programs over the years and a considerable database now exists focused primarily on Cs, Sr and Co. Unfortunately, as can be seen in Table VII, only a very small percentage of the activity contributed by these elements is to be found in cement-based waste forms. In addition, the data which do exist tend to exhibit a high degree of variability. On the other hand, long-lived isotopes such as C-14 and U-238 have significant fractions of their activity tied up in cement-based waste forms. However, release rate data for such isotopes are in very short supply.

Radionuclide Release from Sorbent Media

It is apparent from Table IV that only a very small portion of the total disposed activity at each of the two sites is associated with sorbent media. This is somewhat fortunate in that there is essentially no information in the open literature on the ability of the different commercial sorbents to retain radionuclides in a disposal site environment. Very little of the activity disposed at Richland in 1989 was derived from short-lived isotopes in wastes treated with sorbents and for none of these isotopes did the portion of activity in sorbent media exceed 1%. On other hand, one of the long-lived isotopes, Th-232, has most of its activity (about 80%) associated with sorbent media, while other long-lived isotopes such as I-129, U-238 and C-14 also had significant portions of their activity in sorbents. Thus the lack of information (for example, on solution-solid partition coefficients) assumes much more importance with respect to these long-lived isotopes and severely restricts the ability to model their release from the sorbent media into the disposal site environment.

CONCLUSIONS

The data available on the activity of LLW disposed at the three currently operating commercial disposal sites during the period 1987-1989 have been evaluated in order to determine the distribution in terms of waste stream, waste classification and waste form. The data from waste disposed during 1989 at one of the sites (Richland, WA) were amenable to a more in-depth treatment, including determination of the distribution for each radionuclide by waste form. These data have been reviewed in terms of the specific needs for improved modeling of releases from waste packages.

Based on the evaluation of available data, the following improvements are needed to facilitate performance assessment:

1. There is a need for the development of a uniform manifest system which includes, among other items, more information on containers and a requirement for reporting the distribution of radionuclides within each container.
2. Better characterization of the waste form is required. Currently, a very large proportion of the waste is being disposed in forms described simply as "sorbent or solidification media not required." The physical and chemical characteristics of these waste forms must be better defined in order that appropriate release rate data can be identified.
3. Although long-lived isotopes such as Th-232 and U-238 comprise only a small fraction of the total disposed activity, they are often found in sorbents or cement-based media in much higher proportions than are the short-lived isotopes. There is a distinct lack of relevant release data on these isotopes and this impedes the development of an effective source term model.

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