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Carbon-14 in Low-Level Waste

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REPORT SUMMARY

This report describes EPRI's collective efforts to understand and model the behavior of long-lived radionuclide Carbon-14 (^{14}C) in low-level waste (LLW) disposal facilities.

Background
NRC's analysis on low-level waste performance assessments (PA) identified ^{14}C as a potentially major contributor of dose to the public. In that study, projections showed the majority of ^{14}C in a LLW disposal site were from nuclear power stations. This report lays the groundwork for refining ^{14}C dose projections from new disposal facilities.

Objectives
To understand the workings of existing PA models; to identify opportunities for model enhancement; to provide a technical basis for refining ^{14}C dose projections; to characterize ^{14}C in materials typically disposed of in low-level facilities; to define the release mechanisms that may occur in a disposal facility to mobilize ^{14}C; and, to describe or develop models of the mechanisms that allow for the identification of the most sensitive input parameters.

Approach
The project team undertook an inventory assessment of key ^{14}C -bearing waste forms projected for disposal in LLW facilities. The team integrated data on historical waste forms and used them to characterize potential content and chemical distribution of ^{14}C in future wastes. The characterizations were used in performance models to predict performance impacts of ^{14}C due to various system parameters. Team members developed a new model for low-level waste facilities that assessed the impact of ^{14}C release via postulated airborne pathways. Finally, they developed an improved approach to the overall performance assessment and used it to illustrate the relative changes in a hypothetical performance assessment resulting from this study.

Results
The study achieved its objectives by

Improving the description of ^{14}C inventory in waste disposal facilities

Developing mechanistic models of ^{14}C release from key waste types identified in the inventory assessment

Assessing the role of gas-phase release of ^{14}C and its potential for reducing groundwater dose

Defining improvements in the modeling approaches in complete performance assessment analyses, including sensitivity analyses.

EPRI Perspective

Longer lived mobile nuclides such as ^{14}C are important to performance assessment analyses. The better we understand the quantities and chemical makeup of these radionuclides, the better we will be able to project their migration through the environment. Based on the latest NRC, EPA, and EPRI research, EPRI has taken a systematic approach to identifying PA areas where modifying some of the estimates will refine the dose projections from ^{14}C disposal. To provide a level of comfort in their outcomes, the PA codes are purposely conservative. EPRI recognizes this and has investigated only those areas where there is technical support for modifying the models. The body of the report presents results of this study in a clear, concise manner. Details of the work's technical basis are contained in the appendices and the interim report.

Revisions resulting from this work were incorporated into an alpha version of a new research performance assessment code with capabilities of running all necessary sub-models as a systems analysis tool. This tool allows an investigator to enter essential data once and get the annual dose as the final product. Though not in a user-friendly format, this code could be made user friendly. It could then provide new site developers with a powerful tool for screening proposed sites and conducting iterative site analyses as the siting progresses.

Related reports dealing with ^{14}C are *Characterization of Carbon-14 Generated by the Nuclear Power Industry* (TR-105715), *Soil to Plant Transfer Of Carbon-14 for Environmental Assessment of Radioactive Waste Repositories* (NP-6946), and the interim report for this project, *Technical Issues and Low-Level Waste Performance Assessment:* ^{14}C (TR-107995).

TR-107957

Keywords

Low-level waste (LLW)
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ABSTRACT

This report summarizes EPRI’s latest efforts to understand and model the behavior of the long-lived radionuclide ^{14}C in low-level waste (LLW) disposal facilities, and provides a technical basis for refining ^{14}C dose projections from proposed facilities.

The research is aimed at reducing the uncertainty and conservatism in existing data and models to allow more realistic ^{14}C dose projections. The results of these efforts are presented in terms of a hypothetical performance assessment to illustrate the potential improvements afforded by this research.

This Final Report provides detailed information and impacts of these findings relative to the issues identified:

- Improving the description of ^{14}C inventory in waste disposal facilities;
- Developing mechanistic models of the release of ^{14}C from the key waste forms identified in the inventory assessment;
- Assessing the role of gas-phase release of ^{14}C and its potential for reducing groundwater dose; and
- Improvements in the modeling approaches in complete performance assessment analyses, including sensitivity analyses.

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BACKGROUND

Chapter Summary

Siting of low-level radioactive waste (LLW) disposal facilities is of keen interest to the nuclear power industry. Communities that accept a radioactive waste disposal facility need assurance that existing regulatory public health limits will be met and their sites will not become environmental hazards for future generations.

This chapter summarizes some of the basic performance assessment information as applicable to a long-lived isotope of carbon (Carbon-14 or ^{14}C).

1.1 Introduction

The performance requirements for low-level radioactive waste disposal facilities are given in 10 CFR Part 61which states:

"Concentrations of radioactive material which may be released to the general environment in groundwater, surface water, air, soil, plants and animals must not result in an annual dose exceeding an equivalent of 25 millirems to the whole body, 75 millirems to the thyroid, and 25 millirems to any other organ of any member of the public. Reasonable efforts should be made to maintain releases of radioactivity in effluents to the general environment as low as reasonably achievable."

To achieve these or other more stringent performance regulations that may be imposed by host states, the disposal facility may limit the amount of any radionuclide allowed into the facility. To understand how this might occur it is important to recognize that the total allowable inventory of any radionuclide is determined by conservative performance assessment. If a particular radionuclide is predicted to be a potential performance issue when it is included in a waste inventory, special restrictions could be placed upon waste types that contain the particular radionuclide. The restrictions would limit the total amount of a particular radionuclide in the inventory, based on performance assessment modeling, to meet the 25 millirem dose limit.

In most cases, the majority of ^{14}C in a disposal site will be from nuclear power stations. Due to the results of conceptual performance assessment analyses, longer-lived, mobile nuclides such as ^{14}C can be identified as potential contributors to off-site radiological impacts. Better understanding of the quantities and chemical makeup of these radionuclides will allow improved projection of their migration through the environment and dose contributions. The success of such efforts may assist in licensing related decisions with performance assessment.

1.2 Importance of Performance Assessment (PA) Models to the Siting Effort

Proposed radioactive waste disposal facilities are modeled to predict the potential radiation dose to humans from these facilities. Some performance assessment models used in estimating radiation doses from hypothetical LLW disposal facilities have predicted that ^{14}C will be a major contributor of radiation dose to humans [NRC, 1997]. These projected doses are due, in part, to the long half life of ^{14}C (5,730 years), the typically mobile nature of carbon in the environment (in anionic form), and its presence in LLW produced by various generators including nuclear power plants, academic institutions, medical facilities, and biomedical research.

1.3 Carbon-14 Basics

Carbon-14 is a naturally occurring radionuclide, produced by cosmic radiation interactions in the upper atmosphere. ^{14}C decays to non radioactive nitrogen-14 (^{14}N). The predicted dose rate to humans from atmospherically produced ^{14}C is about 1.25 millirem per year [NCRP, 1985]. ^{14}C is also produced synthetically from nuclear power plants, nuclear weapons testing, and research facilities. The quantities of ^{14}C produced by human activities is small compared to naturally occurring ^{14}C.

^{14}C appears in a wide variety of compounds used in hospitals, universities, biomedical research institutes, and nuclear medicine procedures. For the purpose of tracing organic compounds, ^{14}C is used in the pharmaceutical and organic chemical industries thus enabling the mechanisms of reactions and biological processes to be determined. LLW resulting from these procedures include liquid scintillation vials, absorbed aqueous and organic liquids, calibration standards, biological wastes and trash.

^{14}C is present in LLW generated at nuclear power plants. Low-level waste containing ^{14}C come from activated metals (usually retained within the component). Another source of ^{14}C are activation products of the primary coolant water, where LLW results from contact with the coolant water contaminantes, which includes ion exchange resins, filter sludge, cartridge filters, trash, and, historically, solidified concentrated liquids.

Ingestion of ^{14}C occurs by eating food and breathing air contaminated with ^{14}C. The metabolism and kinetics of ^{14}C follow those of ordinary carbon in the human body. A fraction of the ^{14}C introduced into the body is retained as protein, fat, carbohydrate and other cellular components until equilibrium is reached. The decay of ingested ^{14}C results in the release of an electron (beta decay with and average energy of 0.045 MeV and a peak energy of 0.156 MeV) that deposits its energy in local tissues. The internal dose is the key concern and thus the determination of the ingestion and retention of ^{14}C in the body is a key component of performance assessments.

Although on a per-curie basis, ^{14}C results in approximately an order of magnitude lower dose than ^{60}Co or ^{137}Cs, its long half-life, biomobility, potential for bioaccumulation, and incorporation into tissue make it a potentially significant contributor to dose over the life of a low-level waste facility.

2

STATUS OF C-14 PERFORMANCE ASSESSMENT PRIOR TO THIS WORK

Chapter Summary

This research was motivated by the question: “Could the existing state-of-the-art performance assessment models be improved by:

- integrating a mechanistic source-term code into an overall systems code,
- providing an inventory distribution database, assessing the need and impact of a gaseous release model,
- developing a specific model for ion exchange **[IX]** resins, and other specific waste forms, and, developing near-field transport models to provide a distinction in geochemistry between the waste form and the near-field, such that different values of K_d and/or solubility could be assigned to a radionuclide for different parts of the calculation.”?

A simplified means of illustrating the results of this work is through a hypothetical performance assessment. An accepted state-of-the-art model was used as the benchmark and the effects of the work presented in this report are shown relative to this prior benchmark. Figure 2-1 illustrates the benchmark. Since the effects of the improvements are often observed at different time scales, the graphs presented in the following chapters may change in terms of the scaling, but the data that represents the relative prior performance assessment calculation is consistent with Figure 2-1 throughout.

The intention of this presentation of the research is to show the potential changes that may be realized in PA if the approaches are adopted as part of the PA. The magnitude of the results will change based upon specific PA conditions, and thus only qualitative benefits are highlighted.

A few features of Figure 2-1 are worth noting. The early release period (0-500 yrs) the facility is assumed to be intact and thus the infiltration rate is low, and thus the releases are low. At 500 years, the facility is assumed to catastrophically fail and larger amounts of water are assumed to access the facility. Because a conservative approach to the release of radionuclides is assumed in this hypothetical model, a large spike in the relative release is predicted to occur soon after the facility failure at 500 years. After the high release period, the facility is depleted of all of the easily accessible radionuclides (Class A). In the longer term the relative releases are dominated by the slow release from wastes contained in high-integrity containers (Class B and C wastes).

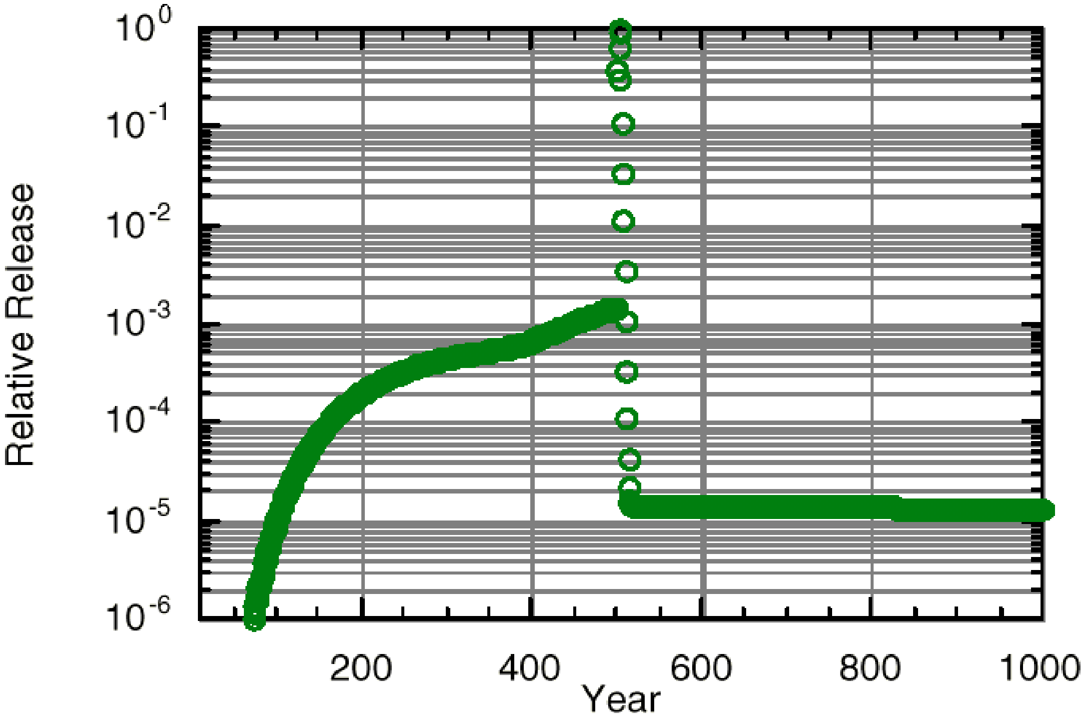

Figure 2-1
Relative Performance Assessment Results

2.1 Performance Assessment Model Review

Appendix A provides a detailed review of the existing performance assessment models. The improvements to the performance assessment models that are specific to the issue of ^{14}C are mainly in the area of the source term. Source term is the phrase that is used to refer to the prediction of the release of radionuclides from the engineered boundaries of a low level waste disposal facility. The source-term analyses fit into the overall performance assessment, as depicted in Figure 2-2.

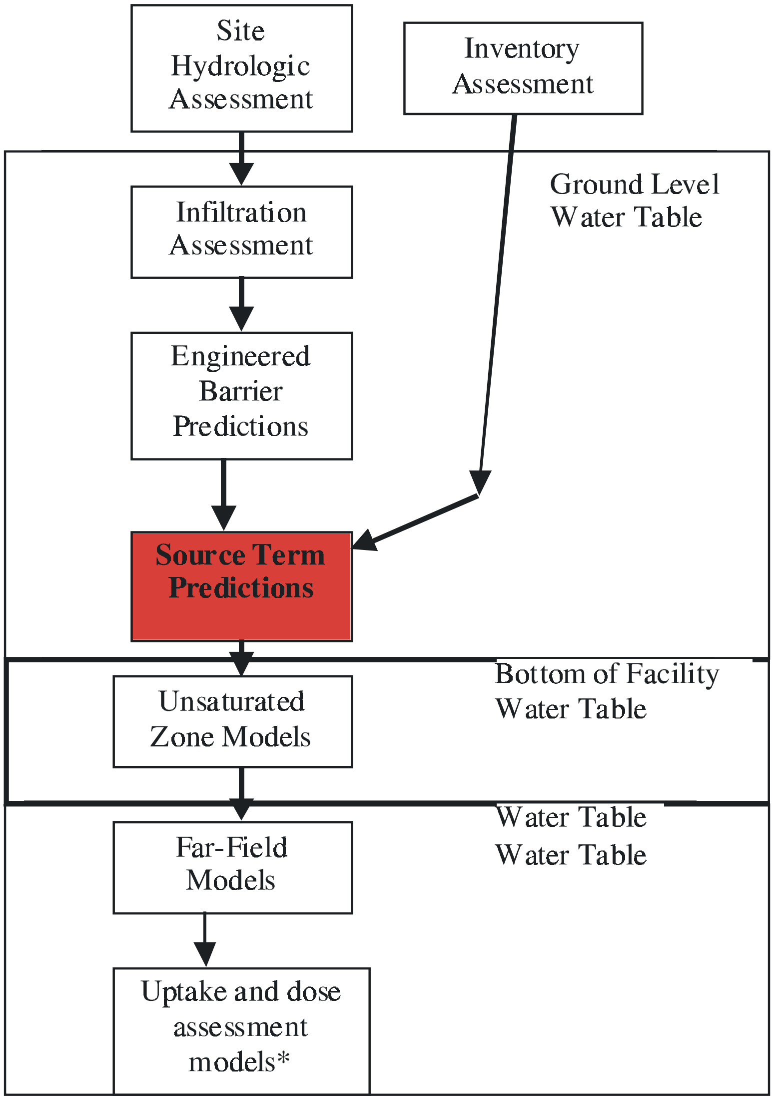

Figure 2-2
Components of a Performance Assessment

Predicted results of source-term releases directly dictate the subsequent analysis of geosphere and biosphere transport in performance assessment. Source-term analysis is typically very conservatively estimated in an overall performance assessment.

Uncertainties are a necessary part of any modeling. In the models discussed in this report, uncertainties may arise from the compositions of the wastes (input data, see Chapter 3), modeling approaches or assumptions, and uncertainties in the parameters involved in the models used. The importance of these uncertainties on an overall assessment are discussed in Chapter 6.

* Each model box generally requires the compilation of several sub-models to form an integrated "model". In addition, each major modeling area there are several models, these are discussed in Appendix A.

2.2 Conclusion of PA Review

Many uncertainties in performance assessment can be eliminated by careful selection and characterization of a candidate site. NRC's BTP on performance assessment[28] stresses that model uncertainty should be addressed in all performance assessments and recommends the use of comparing multiple alternative models, where use of unreasonable modeling assumptions must be avoided.

This leaves opportunities and challenges. In this report we have attempted to develop improved or enhanced models, for some of the processes which appear to impact performance objectives. The models reviewed in this study can be important tools for various analyses in performance assessment. Selection and use of these models should be exercised based on the identified needs of a site.

Groundwater models involved in describing the transport of radionuclides in the environment and the resulting doses have been the subject of research for many years and are in general well developed. A new breakthrough in performance assessment modeling by the researchers was judged to be unlikely through the enhancement of these models. Adequate site data collection and integration still remain key to good results in this area.

The modeling of certain aspects of the engineered site and the wastes contained within are very specific to LLW. This allowed us to enhance the existing models on gaseous release of radionuclides from an engineered facility; describing radionuclide release from IX resins; degradation of cemented waste and subsequent increase in radionuclide release.

With the increasing trend of using advanced technologies in LLW disposal, more reliance on backfilling the disposal units with cement is expected. When the disposal units are backfilled with cement, many of the processes in the near field are affected. Existing models and their boundary conditions for the release of radionuclides from various waste forms need to be evaluated in the presence of cement backfill, as this may improve radionuclide retention within a LLW facility.

Chapter 3 and Appendix B provide a re-assessment of the inventory, and an electronic database is provided as part of this report. Chapter 4 and Appendix C are improved mechanistic models of the processes occurring within a LLW facility to degrade the wastes containing ^{14}C and the subsequent release to ground water. Chapter 5 gives and overview of improved mechanistic models occurring within a LLW facility to degrade the wastes containing ^{14}C and the subsequent release to the gas within the facility which was covered in detail in the interim report (Yim, 1997). Chapter 6, Feature Integration, goes beyond the source term to discuss work done to improve the overall performance assessment. Chapter 7 provides our conclusions.

3

INVENTORY AND DISTRIBUTION OF ^{14}C IN LLW

Chapter Summary

An accurate description of ^{14}C inventory in different waste streams, waste forms, and disposal containers is the starting point of generating a source term analysis in performance assessment. The distribution and chemical nature of ^{14}C in the various waste forms and materials is a key factor in the prediction of the length of time that the waste containers prevent groundwater contact with the waste and subsequent release to the environment. This chapter summarizes the source of ^{14}C in nuclear power plants, evaluates the distribution among various potential waste forms, including a mass balance, summarizes key chemical factors of the inventory distribution, and presents the impact on a hypothetical performance assessment (PA). The key summary of the impact of the work described in this chapter and in Appendix A is shown in Figure 3-1. Improved estimates of the inventory and distribution within the inventory are shown to have the potential to change the nature of the release such that lower peaks may be observed. The key reason for this difference is the redistribution of ^{14}C into higher classification wastes (i.e., class B and C vs. class A). The engineered barriers for the higher classes have better integrity and rather than a large predicted release at 500 years, the releases are lower, but extended over a longer period of time.

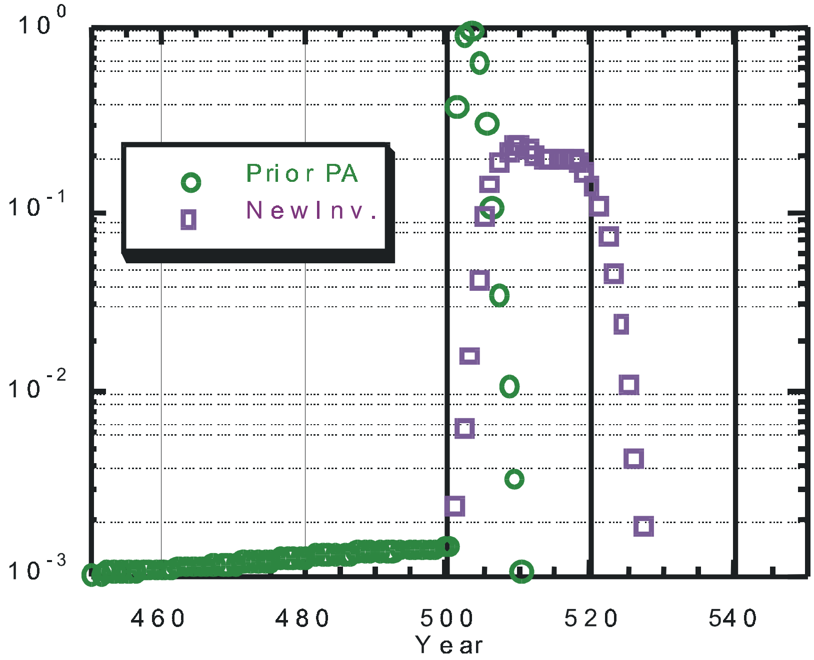

Figure 3-1
Effect on PA Results with Revised Inventory Estimates

3.1 Sources of ^{14}C in Nuclear Power Plants

Sources of ^{14}C relevant to LLW are core structural materials and reactor coolant activation. Although ^{14}C is produced in reactor fuels, this is destined for high-level waste disposal.

3.1.1 Core Structural Materials

Carbon-14 production from core structural materials is due to the presence of nitrogen, the predominant isotope, ^{14}N, reacts by capturing a neutron and emitting a proton to become ^{14}C (^{14}N(n,p)^{14}C). Nitrogen is typically found at significant levels in stainless steel support hardware, zircaloy cladding, and nickel alloys used as fuel and fuel bundle hardware. A production rate of ^{14}C in core structural materials was estimated at approximately 20 Ci/Gw(e)-yr [Bonka, 1974].

3.1.2 Reactor Coolant

Carbon-14 is also produced in reactor coolant from reactions with oxygen, nitrogen, and carbon, in decreasing importance:

- oxygen atoms in the water molecules ^{17}O(n,α)^{14}C;
- nitrogen dissolved in the water ^{14}N(n,p)^{14}C; and
- carbon dioxide organic compounds in the water ^{13}C(n,g)^{14}C.

Predicted production rates of ^{14}C from these reactions are listed in Table 3-1 [EPRI, 1995]. These predicted values are based on the following assumptions:

- An average flux in a typical LWR is 4.8×10^{13} neutrons/cm^2/sec;
- A 1000 MW(e) PWR has a mass of coolant water exposed to this flux of about 13,400 Kg; and
- an equivalent BWR has a mass of about 33,0000 Kg.

Table 3-1
Predicted Rates of C-14 Production in LWRs [EPRI, 1995]

Reaction	*PWR*	*BWR*
^{17}O(n,α)^{14}C	6.0 Ci/GW(e)-yr	14.5 Ci/GW(e)-yr
^{14}N(n,p)^{14}C	1.2 to 5.0 Ci/GW(e)-yr[1]	<1 Ci/GW(e)-yr[2]
^{13}C(n,γ)^{14}C	0.011 Ci/GW(e)-yr[3] per ppm of CO_2	0.027 Ci/GW(e)-yr[3] per ppm of CO_2

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[1] based on 10 to 40 ppm of dissolved nitrogen gas with 0.12 Ci/GW(e)-yr/ppm of N-14

[2] based on 10 to 40 ppm of dissolved nitrogen gas with 0.29 Ci/GW(e)-yr/ppm of N-14

[3] Not a significant source of C-14 production

From these sources, a portion of ^{14}C becomes part of the waste stream and is sent to LLW facilities for disposal. Waste streams containing ^{14}C include:

- ion-exchange resins,
- irradiated hardware,
- evaporator bottoms,
- filter sludge,
- cartridge filters, and
- trash.

Annual average quantities of ^{14}C shipped in low-level solid radioactive waste for plants excluding the irradiated hardware, on a reactor year basis is 1.9 and 1.23 Ci/GW(e)-yr for PWRs and BWRs, respectively [EPRI, 1995]. These values are used in Section 3.3 to verify an overall mass balance.

3.2 Distribution of ^{14}C in LLW From All Generators

3.2.1 Waste Form Types

As currently practiced, LLWs are disposed of in a wide variety of materials and forms, including metals, resins, filters, mixed trash, liquids solidified in cement, or stabilized with sorbent. The type of waste form has a considerable impact on the ability to predict the retention of radionuclides in the waste once they are in contact with ground water. The understanding of the chemical and physical attachment of ^{14}C to waste forms is variable, depending upon the particular waste form. Historically, very little or no credit has been taken for potential benefits of the waste form in preventing its release in performance assessment.

In encapsulating waste forms, e.g., cement, the physical and chemical distribution of ^{14}C is better defined and credit can be taken for limited release to the environment. For example, if wastes are solidified in cement, the chemical environment surrounding the waste will be altered to a high pH. The high pH will result in reduced microbial activity and low solubility of typical ^{14}C compounds in solution. Both of these facts can be utilized in a performance assessment, which results in lower releases to the environment and reduced environmental impact.

The value of this type of understanding of waste form inventories is becoming increasingly recognized, not only for ^{14}C, but for other radionuclides as well. Pioneering studies have been completed by U.S. NRC [U.S. NRC, 1981a], EPA [U.S. EPA, 1988], and EPRI [EPRI, 1988, 1989] and improvements in the rigor and detail of the reporting of waste type and inventories being delivered to LLW facilities have resulted from these efforts. Examination of these historical works for the purposes of the studies that were being undertaken for ^{14}C by EPRI indicated that additional analyses of the inventories were necessary to generate more detailed descriptions of the waste. The key items identified were information on ^{14}C activity for the various waste classes and information pertaining to the solidification, treatment or encapsulation of each of the waste streams.

Based on the manifest information collected at the Barnwell LLW disposal site, for wastes from all generators, between 1989 and 1994 [Dames & Moore, 1995], estimates of distributions of ^{14}C in various waste forms are presented in Table 3-2. As shown in Table 3-2, ^{14}C is mainly associated with dewatered ion-exchange resins, irradiated hardware, and mixed dry active waste (DAW).

Prior inventory assessments are also shown in the table. Cement waste forms were generally solidified liquids from demineralizers and reverse osmosis concentrates. Sorbents include solutions from various washing operations that have been absorbed into a porous inorganic medium.

The redistribution of ^{14}C amongst the different 10CFR61 classes of LLW based upon these inventory estimates is also shown in Table 3-2. The redistribution results in significantly less ^{14}C in class A and a much greater fraction in B and C. This redistribution is the key reason that performance assessment predictions shown in Section 3.4 are predicted to be reduced. The reason behind this difference lies in the fact the majority of activity is in class B & C wastes, which are placed in durable waste containers (High-Integrity Containers or HICs). Our analysis shows the more durable containers do not eliminate ^{14}C release from a LLW facility, but are predicted to release ^{14}C over a longer period of time and thus reduce and broaden the peak release.

Table 3-2
Distribution of ^{14}C

Waste Form Description (as stated in Manifest)	*Prior Distribution*	*New Distribution*
Ion Exchange Resins		48.8%
Irradiated Hardware[4]		24.1%
Mixed DAW		13.6%
Solidifed Liquids		4.4%
Filter Media		3.6%
Cartridge Filters		2.7%
Solid Non-combustibles		1.2%
Incinerator Ash		1.2%
Air Filters		0.15%
Biological Wastes		0.15%
Cement	43.3%	
Sorbent	5.9%	
None	50.8%	
Total	100%	99.9%
Class		
A	62%	31.3%
B	4%	15.6%
C	32%	53.1%

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[4] Does not include greater than class C wastes.

3.2.2 Chemical Nature of ^{14}C from Nuclear Power Plants

In addition to reclassification of the waste forms and categories as part of the inventory assessment, data on the chemical form of ^{14}C in the waste was also used. Table 3-3 summarizes data from EPRI report TR-105715, "Characterization of Carbon-14 Generated by The Nuclear Power Industry," EPRI 1995. There was little prior characterization of the chemical forms of ^{14}C; values were estimated on the basis of very limited data.

Table 3-3
Summary data on the Chemical Form of ^{14}C in Nuclear Power Plants.

Reactor Coolant Systems	PWR 58 to 95% organic
	BWR 13-48% organic
	Organic radiocarbon in PWR coolant system:
	40% acetaldehyde
	20% methylalcohol
	30% ethylalcohol and acetone
	2% acetic acid [Matsumoto, et al., 1995]
Fuel Cycle Waste	
	75% inorganic (carbonate)
	20% activated metal
	5% organic radiocarbon

The fraction of inorganic versus organic ^{14}C is significant in that it factors into the solubilities, distribution coefficients, and microbial activity models described in Sections 4 and 5. Each of these factors was considered in this study, when assigning radionuclide transport parameters.

3.3 Mass Balance of ^{14}C in Nuclear Power Plant LLW

An effort was made to verify the findings of results presented in Section 3.1 based upon evaluation of waste manifest data at Barnwell versus an estimate of the fraction sent to LLW facilities based upon a mass balance within a nuclear power plant [EPRI, 1995]. Table 3-4 illustrates the findings of the mass balance.

Table 3-4
Summary of ^{14}C mass balance within Nuclear Power Plants

1) Production in Reactor Coolant System

PWR	10 Ci/GW(e)*yr
BWR	15 Ci/GW(e)*yr
On average	11.5 Ci/GW(e)*yr

2) Barnwell 89-94 Manifest

Total LLW received	300,000 ft^3/yr
42.03% volume from Nuclear Power Plants (NPP)	127,000 ft^3/yr
Total ^{14}C reported	32.27 Ci
80.5% from NPP	26 Ci

3) Utilities volumes sent to LLW, based on Barnwell (89-94)

LLW volume produced	434 ft^3/yr/unit
Each unit sent	0.9 Ci/unit

4) Percentage of ^{14}C that remains in LLW

based upon this study using Barnwell data:

<u>Activity each unit sent to LLW : 0.9</u>
Total estimated activity produced: 11.5 ~ 8%

5) Previous industry studies [EPRI, 1995] show that 5~20% of ^{14}C remain in LLW.

The agreement of these two independent paths to estimating the total ^{14}C inventory in LLW from Nuclear Power Plants gives a level of confidence in the findings presented in Section 3.4

3.4 Summary of PA impact of Carbon-14 Inventory and Distributions in Utility LLW Streams

An analysis was performed to determine the effect on performance assessment results of a more detailed description of the inventory. An estimate of inventories based upon the mix of waste forms and classes can be derived from the data that is supplied in both a text version and Microsoft EXCEL file in Appendix B. The key finding was that significantly less ^{14}C was associated with Class A waste than had previously been assumed. The previous inventories with a large amount of ^{14}C in Class A was predicted to cause large releases whenever the facility boundary is breached (at 500 years, shown by the circles in the assessment shown below). The more detailed inventories developed as part of this work result in ^{14}C being associated with the higher integrity of the Class B and C waste containers/forms. This is predicted to result in more gradual release, and hence a much lower peak (see New Inventory values, below). This effect is due only to a redistribution of the inventory, keeping all other performance assessment variables the same. The key result is the illustration of the importance of accurate waste inventory measurements and the benefits more detailed measurements and inventory assessment may have on performance assessment predictions.

4

IMPROVED MECHANISTIC MODELS OF CARBON-14 IN LLW

Chapter Summary

Mechanistic models simulating the release of radionuclides from low level waste forms has been limited. Mechanistic models are attempts to describe in detail the physical (e.g., fluid flow) and chemical (e.g., diffusion) processes that occur in a low-level waste repository that may lead to radionuclide release. The ultimate goal of mechanistic models is to develop defensible predictions of future outcomes. This is in contrast to empirical models that are best suited to interpolated predictions, rather than extrapolations.

Generally, a concept wherein all radionuclides are immediately released from a waste form into any water that comes into contact with the waste form, termed a surface rinse model, is used in the absence of understanding of retention of radionuclides by various waste forms. The waste forms of key interest are those that were identified in the previous section as having high fractional contents of ^{14}C. This chapter addresses mechanistic models for ion-exchange resins, filters and dry active waste, cemented wastes, and metal corrosion, respectively.

The outcome of mechanistic models, as illustrated in Figure 4-1, may show only a modest compliance benefit. None-the-less, from this type of analysis, the models can provide a level of confidence in the conservatism of more simplified approaches.

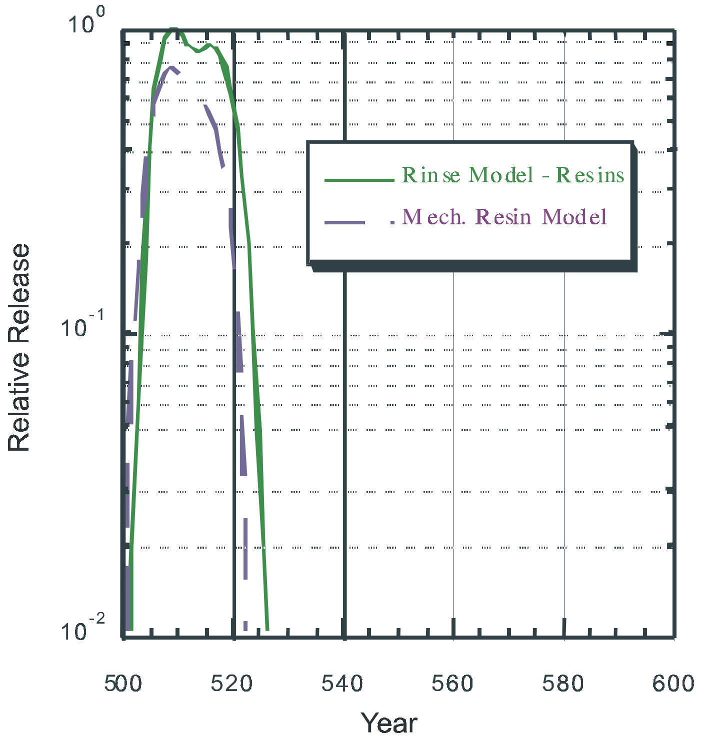

Figure 4-1
Comparison of a Simple “Rinse” Release Model with a Mechanistic Resin Model

4.1 Modeling Carbon-14 Release from Spent Ion-Exchange Resins

4.1.1 Introduction

This analysis proposes a more mechanistic approach to the prediction of releases from ion-exchange resin wastes, which, as described further in this report, that are a significant source of ^{14}C.

As currently practiced, LLWs are disposed in a wide variety of materials and forms, including metals, resins, filters, mixed trash, cement, glass, or sorbent media. The waste form into which the radionuclides are incorporated has a considerable impact on the manner and degree to which they are released or retained in the waste form. The degree of uncertainty or scientific understanding in the analysis of radionuclide release varies among different waste materials. There exists a significant body of work regarding the mechanistic understanding of chemical durability and leaching characteristics of glass, cement, metals or other solidified waste. The unsolidified waste streams, have not been studied to the same level of detail.

Among various types of unsolidified LLW, such as ion-exchange (IX) resins, filters, dry active waste, and incinerator ash, IX resins contain the largest inventory of radioactivity of long-lived, potentially environmentally-mobile radionuclides. According to the shipping manifest information collected at the Barnwell, South Carolina LLW site [Dames and Moore, 1995], the estimated inventory of radioactivity contained in unsolidified IX resins is 47% of the total disposed activity for ^{14}C, 59% for ^{90}Sr, 32% for ^{129}I, and 81% for ^{137}Cs. Even though regeneration of IX resins was practiced in the early history of nuclear power plant operations, the used, or "spent" IX resin beds are now commonly discarded at the time of refueling outage.

For disposal purposes, spent IX resins used to be immobilized in cement. However, problems were encountered in this processing. Current practice is to pre-treat IX resins by a dehydration/dewatering process and store them in high integrity containers (HICs) for disposal.

Release of radionuclides from IX resins is commonly modeled by a simplistic, conservative model known as "surface wash release". In this approach, radionuclide release is assumed to occur instantaneously upon contact by water. The only limits to this release are the solubility limit of the particular chemical form of the various radionuclides, and physical capture of the material in the surrounding materials. In some cases, the solubility limit is very high. The influence of the solubility and the "stickiness" of the surrounding material is represented by a combined term known as the distribution coefficient. The distribution coefficient (K_d) represents the state of equilibrium within the system. Typically very conservative (i.e., low) values of K_d are selected for performance assessment.

In reality, the distribution of activity within bulk resin medium between the solid phase and liquid phase is expected to be dependent on whether the radionuclide "attachment" to the resin matrix is more surface-controlled, ion-exchange-controlled, or filtration-controlled. Also, even with the accurate determination of how much activity is partitioned into the liquid phase, actual release of radionuclides from the waste package will depend on the diffusional transport characteristics within the waste-form, waste package porosity.

This study examines these issues with a goal of better mechanistic understanding and modeling of radionuclide release from IX resins. Mechanisms of radionuclide release from IX resins were examined and a supporting mathematical/computer model was developed.

4.1.2 Background on the Release of Radionuclides from Ion-Exchange Resins

Ion exchange resins are three dimensionally crosslinked hydrocarbon chain material. The chains contain ions that can be exchanged for an equivalent amount of other ions of the same sign when the ion-exchanger is in contact with an electrolyte solution [Helfferich, 1966]. The exchange is a reversible stoichiometric process. This stoichiometric redistribution of **counter-ions**, or ions of equal charge that exchange "places" with each other between the ion exchanger and the contacting liquid phase, is largely responsible both for decontamination capability and predicted mechanistics of radionuclide release after disposal.

One way the characteristics of ion exchange can be determined is by a fluid-solid reaction which produces fluid products. Five basic steps listed below outline the exchange reaction between fluid and particles, also referred to as a mass transfer [Levenspiel, 1961]. Figure 4-2 illustrates the process of mass transfer of ions between bulk liquid and resins.

step 1. Fluid reactant diffuses through the film surrounding the resin solid to the resin surface, i.e., particle *B* diffuses along the dashed arrow to the resin surface.

step 2. Fluid reactant diffuses inside the resin matrix, i.e., circle *B* diffuses along the bold solid arrow inside the resin to the counter-ion site.

step 3. Fluid reactant exchanges the counter-ion species *A* inside the resin (˙*R*), i.e., chemical reaction between fluid reactant *B* and counter-ion *A* as follows.

$$B_{(re\ \sin)} + A \bullet R \leftrightarrow A_{(re\ \sin)} + B \bullet R$$

step 4. The counter-ion species diffuses through the resin matrix to the solid surface, i.e., circle *A* diffuses along the bold solid arrow inside the resin particle to the resin surface.

step 5. The counter-ion species from the resin surface diffuses through the liquid film into the bulk fluid, i.e., particle *A* diffuses along the dashed arrow to the bulk solution.

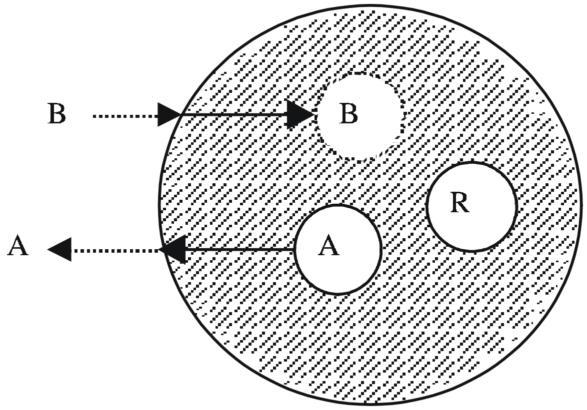

Figure 4-2
Illustration of Ion Exchange

In order for an exchange of counter-ions *A* (carried by resins) and *B* (from liquid phase) to take place, species *B* must migrate from the solution into the interstitial spaces of the ion exchanger, and species *A* must migrate from the exchanger into the solution. For the chemical reaction in step 3, its reaction rate is commonly expressed by the first order reaction rate:

$$d\frac{\lfloor A_{(re\sin)} \rfloor}{dt} = k_l \bullet \left[B_{(re\sin)}\right] \bullet \left[A \bullet R\right]$$

Eq. 4-1

where k_1 is a reaction constant.

The rate-controlling step in the entire ion exchange process could be a chemical reaction, if the concentration of fluid reactant *B* into resins is not enough for ion exchange to occur, or diffusion, if [$B_{(\text{resin})}$] is abundant. Also, the total amount of counter-ion *A* available to be released into the solution is a key factor in determining the leaching amount from the containers.

Diffusion of exchanging counter-ions within ion-exchangers is believed to be slower than in solutions due to the existence of three dimensional framework and tortuous path length in the narrow-mesh regions. Due to the effects of an electric field generated by ionic diffusion, the net flux of the faster and slower ions is balanced to maintain electrical neutrality. This is described by the Nernst-Planck theory [Helfferich, 1962] where the resulting electrical transference is superimposed on ordinary diffusion driven by the concentration gradient [Schlogl, 1957]. The Nernst-Planck equation is

$$\bar{J}_A = -\bar{D}_A \; grad\,\bar{C}_A - \bar{D}_A \,\bar{C}_A \left(\frac{z_A\, F}{GT}\right) grad\varphi \qquad \textbf{Eq. 4-2}$$

where $\bar{C}_A$ = concentration of species A (moles/cm^3)

$\bar{J}_A$ = flux of species A (moles/cm$^{2\cdot}$sec)

$\bar{D}_A$ = diffusion coefficients of species A (cm^2/sec)

F = Faraday constant (coulombs/mole)

G = gas constant (ergs/mole deg)

T = absolute temperature (deg)

z_A = valence of species A (negative for anions)

φ = electric potential (ergs/coulomb)

The Nernst-Planck model can be also substituted by Fick's law when the resins behave with weak functionality [Guria, 1994]. Rinse release could also be responsible for the release of radionuclides from IX resins if the activity is retained through surface phenomena.

4.1.3 Description of the Model

A hypothetical system of IX resins is used to simulate the seepage of radionuclides from IX resins in a waste container with infiltrating ground water flow. This system is shown in Figure 4-3.

As shown in Figure 4-3, the resin system consists of the solid resins (phase I) and the bulk pore liquid (phase II). It is assumed that the infiltrating water enters the top of a container, becomes perfectly mixed with the container volume, and leaves the bottom of the container.

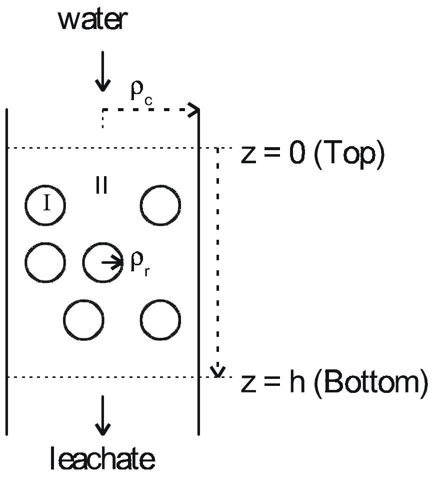

Figure 4-3
Simulated Resin System in a Waste Container (Phases I and II Stand for the Resin Solid, and the Liquid Contiguous to Resins, Respectively)

The two steps involved in the release of radionuclides from waste containers are:

1. transport and release of radionuclides from resin beads to the bulk pore water.

2. the movement of radionuclides in the bulk pore water within waste containers until they leave the container.

The first step requires knowledge of the effects of ionic diffusion, the action of surface rinsing on the physical adherence, and any chemical reactions that may be involved. Movement is affected by the combined effects of diffusion in the bulk pore water, advective transport through the system, sorption during the migration and radioactive decay.

4.2 Modeling ^{14}C Release from Dry Active Waste (DAW) and Filters

For DAW and filters, radionuclides are expected to be attached to these waste materials through surface phenomena, such as physical impingement or adsorption. Surface rinse model is expected to be relevant to these unsolidified absorbing materials.

As previously described, a rinse release model describes the release of radionuclides from the waste forms as immediate release upon water contact. In the rinse process, it is assumed that the water is subject to equilibrium partitioning between the solution and the waste form, represented below.

Using the linear isotherm adsorption model, the solute which is assumed to reach equilibrium quickly in solution can be formulated as:

$$A_s = K_d \bullet A_l \qquad \textbf{Eq. 4-3}$$

where

A_s = the amount of radionuclide adsorbed on solid per gram of solid (Bq/g solid)

A_l = the amount of radionuclide dissolved in solution per ml of solution (Bq/ml solution)

K_d = distribution coefficient (ml/g) defined as the ratio of a radionuclide concentration in a solid phase to that in a liquid phase.

The total amount of radionuclide on solid is represented by

$$S = A_s \bullet v_T \bullet (1 - \varepsilon) \bullet \rho_s \qquad \textbf{Eq. 4-4}$$

where

S = the total amount of radionuclide on solid (Bq)

v_T = the total volume of the waste form, including solid and porous phases (cm^3)

ε = the porosity of the waste form

ρ_s = the density of solid phase (g/cm^3)

and the total amount of radionuclide in solution is given by

$$L = A_l \bullet v_T \bullet \theta_c \qquad \textbf{Eq. 4-5}$$

where

L = the total amount of radionuclide in solution (Bq)

θ_c = the moisture content

It was assumed that $M_t^r(Ci)$ is the amount of radionuclide available in the waste form for rinse at time *t*. Using mass balance based on the above three equations gives:

$$A_s \bullet v_T \bullet (1 - \varepsilon)\rho_s + A_l \bullet v_T \bullet \theta_c = M_t^r \qquad \textbf{Eq. 4-6}$$

The concentration of radionuclide in the solution can be then determined as:

$$A_l = \frac{M_t^r}{v_r \theta_c \left(1 + \frac{K_d \rho_b}{\theta_c}\right)} \qquad \textbf{Eq. 4-7}$$

where

ρ_b = the bulk density (g/cm^3) and

$\rho_b = (1-\varepsilon)\rho_s$

Constrained by the solubility limit, the radionuclide concentration in solution, A_l should be the smaller value out of solubility and solution concentration from the rinse model in (5). The release rate at time t in the rinse model, $Q^R(t)$ (Bq/yr) can be determined as:

$$Q^R(t) = A_l(t) \bullet \mathrm{V}(\mathrm{t}) \bullet \mathrm{B_s}(t) \bullet \theta_\mathrm{c} \qquad \textbf{Eq. 4-8}$$

where

$A_l(t)$ = the time-dependent radionuclide concentration in solution (Bq/ml)

$V(t)$ = the time-dependent linear flow velocity in waste containers (cm/yr)

$B_s(t)$ = the time-dependent breached container area for liquid flow through (cm^2)

4.3 Modeling ^{14}C Release from Cemented Waste

In the case of solidified waste, such as cemented waste, release of radionuclides takes place through diffusion of radionuclides through the pore water in the waste form. This process is known to be independent of the degree of water saturation of the porous matrix but is dependent on the hydraulics of the surroundings which determine the concentration gradient for diffusion. Typically, available models for diffusive release mechanisms assume zero radionuclide concentration at the outer boundary of the waste form to facilitate the derivation of analytical solutions. However, with the trend of designing new facilities with cement backfills, the capability to address the effects of surrounding conditions on the diffusive release of radionuclides is desired. Approaches taken in this study to describe the release of radionuclides from cemented waste include both an analytical-solution model and a numerical model. An analytical model, once developed, is easily implemented. However, analytical models are typically restricted to certain defined conditions. For more generalized conditions that may exist, numerical models are preferred.

4.4 Modeling Release due to Metal Corrosion

Release of radionuclides from metallic waste through corrosion is modeled assuming that radionuclides are released through congruent dissolution of the waste form. The rate of release of all of the radionuclides is assumed to be constant and controlled by the rate of dissolution [Sullivan, 1993]. The release rate by dissolution $Q^U(t)$ (Bq/yr) is given as:

$$Q^U(t) = \frac{v^U M_t^U SA_{waste}^U}{V_{waste}^U}\left(1 - \frac{C(t)}{C_{sat}}\right) \qquad \textbf{Eq. 4-9}$$

where

υ^U is the dissolution rate (cm/yr),

SA^U_{waste} is the surface area of the waste for uniform dissolution release,

V^U_{waste} is the volume of the waste for uniform dissolution release,

M^U_t the amount of radionuclide available for uniform release at time t in the waste form,

$C(t)$= the solution concentration (Bq/cm^3), and

C_{sat} is the solubility limit (Bq/cm^3).

The model is dependent upon the value of the waste form dissolution rate and the surface area to volume ratio of the waste. The possibility of preferential release of constituents is ignored in this model. Although this model was developed, it was not used extensively in this study.

4.5 Conclusion

Mechanistic models have the potential to improve confidence in the predicted outcomes of conservative performance assessments. As shown in the following figure, the differences between the outcomes of an empirical, presumably conservative, “surface rinse” model predicts similar outcomes to the more detailed mechanistic model for the set of assumptions that were made. In the event that the “rinse” model predicts an unacceptable outcome, there is no recourse except for inventory reductions. A mechanistic approach allows for targeting the key parameters or processes that contribute to an outcome and improving understanding of the process. In some cases the mechanistic models and the empirical models may result in the same recommended actions based upon their results. The bottom line in terms of PA is that our knowledge of the behavior of resins has improved, but the impact on PA is not necessarily shown to be significant, as seen below:

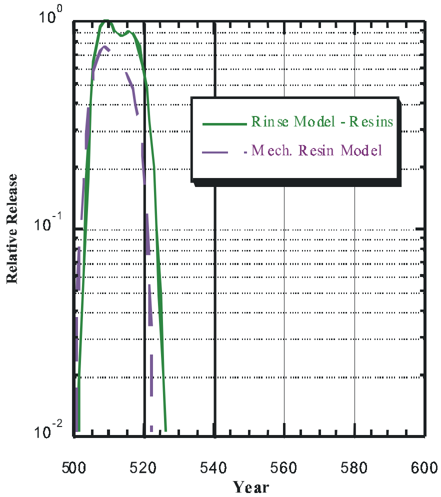

Figure 4-4
Comparison of a Simple “Rinse” Release Model with a Mechanistic Resin Model

5
GAS-PHASE RELEASE OF CARBON-14

Chapter Summary

In current LLW performance assessment models, gaseous release of ^{14}C is not taken into account as a radionuclide release mechanism. This is based upon the well-founded assessment that the most significant pathway for ^{14}C is via a groundwater pathway. However, in actual disposal facilities, amounts of ^{14}C contaminated gases are expected to be generated and this assessment provides a modeling framework to estimate the amounts and impacts.

Gas phase ^{14}C may be available for release at rates greater than would normally be experienced through the liquid (groundwater) pathway. If a significant amount of gaseous ^{14}C activity is released through the air pathway that release will deplete the inventory of ^{14}C available for release through the groundwater pathway. This scenario could have an important impact on the source term. The key finding is illustrated in this figure which indicates that there is the potential for significant depletion of the ^{14}C inventory via biodegradation that leads to gas-phase release.

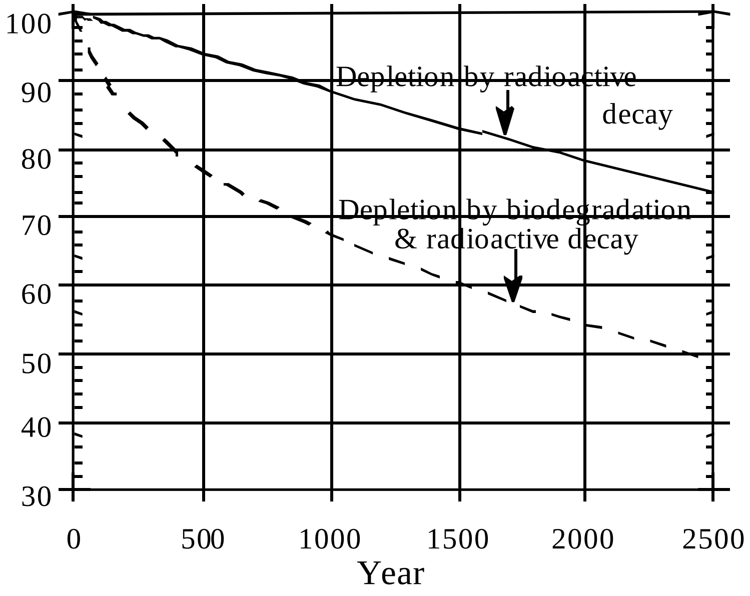

Figure 5-1
Depletion of ^{14}C Inventory by Radioactive Decay and Biodegradation

This chapter summarizes the investigation of the release of ^{14}C through the air pathway and discusses the significant findings. The model is discussed in more detail in the Interim Report, [Yim, 1998]. Figure 5-2, below show that the impact on the overall performance assessment results are expected to be modest, but in some cases, it may be important to assess.

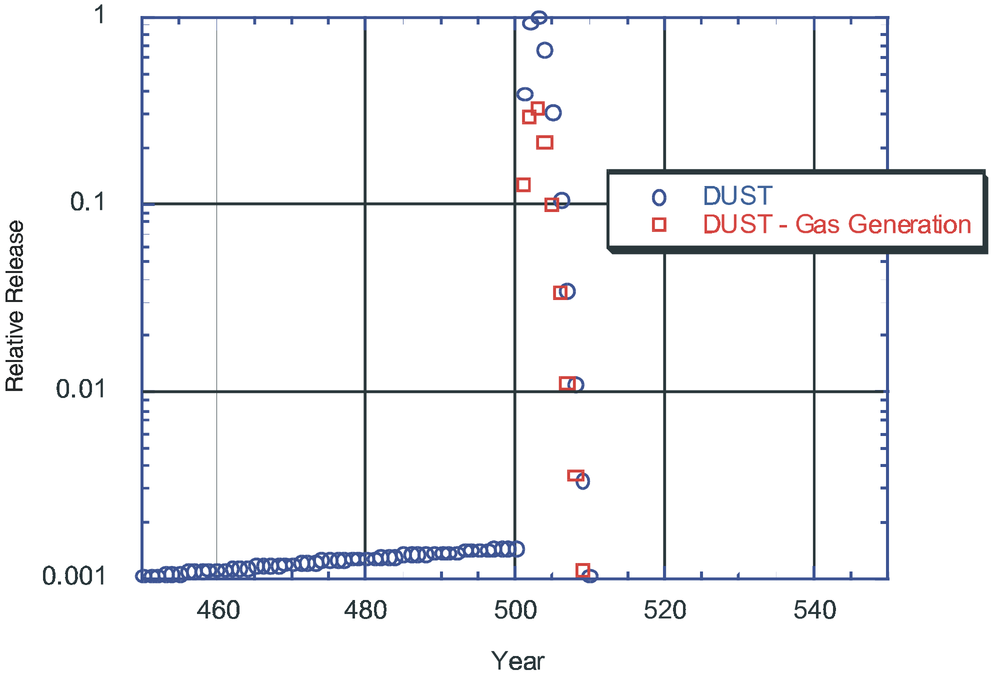

Figure 5-2
Effect on PA Results with Gas

5.1 Behavior of Gases in Disposal Facilities

The presence of contaminated gases in an engineered barrier structure may result in radionuclides entering the atmosphere by diffusion or other mechanisms through the containment structure and soil cover. ^{14}C-contaminated gases may be introduced into the containment structure as a result of the failure of disposal canisters or the decomposition of organic wastes. Because some amount of gaseous and/or organic waste is expected to exist at all disposal facilities, the gas-phase release of ^{14}C was characterized.

Due to the public's unfavorable view of the shallow land burial of LLW, a major share of the LLW disposal facilities being planned by the states and state compacts are to be above- or near-grade bunkered facilities. According to the current design of some proposed facilities, there is a drain in the engineered concrete structure and a standpipe, for monitoring, which is connected to the flow drain. This feature potentially provides a direct pathway for gases to be transported out to the atmosphere from within the facility. The potential radiological dose from the airborne release of ^{14}C for the performance assessment of the newly proposed LLW disposal facilities can be assessed using the modeling approaches given in detail in the interim report [Yim, 1998]. Current performance assessment computer codes without the capability to analyze the gaseous release pathway of ^{14}C provide projections only of the groundwater ^{14}C radiological impacts of these facilities.

If radionuclides in the waste are released to the environment through gases, the source inventory that will be available for the transport through groundwater is reduced. The U.S. Environmental Protection Agency (EPA) has previously estimated [Gruhlke, 1986] that about two-thirds of the total ^{14}C inventory has the potential for atmospheric release, suggesting significant implications of this estimate in overall LLW performance assessments. However, previous to this study the issue has been addressed on a limited basis in performance assessments.

5.2 Characterization of Biodegradable Carbon-14 Inventory

To determine the generation of gaseous ^{14}C, LLW streams need to be characterized in terms of the biodegradable inventory. The element carbon forms organic molecules because of its unique ability to form long chains with itself. Also, carbon forms some inorganic chemical forms, such as carbonate (CO_3^-) and bicarbonate (HCO_3^-). The organic fraction of LLW will have the potential to degrade by microbial processes. To better understand the potential release of ^{14}C activity in the gas-phase, LLW streams need to be characterized in terms of the ^{14}C organic carbon inventory.

Various LLW streams in the form of metals and inorganic substances do not contribute to the organic carbon inventory. These waste streams include equipment, solid non-combustibles, non-cartridge type filter media or incinerator ash. Besides these, many compounds in non utility waste streams are varieties of organic radiocarbon compounds, depending on the type of waste stream. They decompose at various rates when exposed to hydrolysis and microbial action.

For utility LLW, chemical speciation of ^{14}C in dewatered resins is important in determining biodegradable ^{14}C inventory. The chemical speciation of ^{14}C in resins was analyzed for several PWRs and BWRs using simulated mixed-bed demineralizer resins exposed to primary reactor coolant [EPRI, 1995]. The PWR stations exhibited predominantly organic forms of ^{14}C, ranging from 60 to 95%. For the BWR stations, more inorganic forms were observed (42.2 to 100%) in the simulated primary cleanup resins. Reactor coolant chemistry, including hydrogen gas injection and the presence of trace amounts of dissolved organic carbon in the water may be important in the chemical speciation of ^{14}C in the coolant.

Among the total ^{14}C activity in organic compounds, the fraction in a cement waste form in which the pH will remain high is not generally available for microbial degradation. Excluding this fraction, about 40% to 60% of the total ^{14}C activity is estimated to have the potential for gas-phase release through microbial attack based on the manifest information from the Chem Nuclear Facility at Barnwell SC, 1989 through 1994 [Dames & Moore, 1995].

5.3 Computer Model Development for Analysis of Gaseous ^{14}C Release

The objectives of the tasks associated with the analysis of gaseous ^{14}C release include:

- to determine if gaseous carbon compounds are generated in waste containers in a LLW repository;
- to ascertain the amount of ^{14}C in these compounds;
- to determine if the gases are released to the environment;

- to determine the potential dose if ^{14}C gases are released; and
- to consider the implications of gas release to the overall performance assessment of the LLW facility.

To investigate these issues, a computer model that describes microbial gas generation and transport within a generic engineered disposal facility was developed.

Tasks performed in this model development include:

- Classifying biodegradable LLW according to the differences in hydrolysis characteristics and appropriate rate constants developed;
- Modeling ^{14}C-contaminated gases produced by microbial activity using a chemical kinetic description of aerobic and anaerobic decomposition along with modeling of oxygen transport in the waste matrix;
- Developing a computer model that describes microbial gas generation and transport within a waste package and disposal facility. The transport model describes time-dependent effects of diffusion, advection, radioactive decay, and chemical reactions; and
- Making estimates of the ^{14}C inventory which was depleted due to gaseous release and the remainder available for release through the groundwater pathway.

The disposal facility selected for the study is a concrete bunkered facility with a floor drain and a monitoring well. Fluctuations in barometric pressure, which is expected to affect the transport of gas within the facility, are investigated as a major driving mechanism for the release of gases through the monitoring well.

Further details of the model and related analysis are given in the Interim Report [Yim, 1998].

5.4 Impact of Gaseous Release on Hypothetical Performance Assessment

The key finding of gaseous release on performance assessments has been through the reduction of inventory of ^{14}C available for transport in ground water. Figure 5-1 illustrates the reduction observed in the overall inventory. This generally translates into an overall equivalent reduction in release compared to the benchmark performance assessment case, that does not account for the gas pathway. This is illustrated in Figure 5-2 which shows the impact on the hypothetical performance assessment. Modest, yet quantifiable, reductions are observed in the peak dose in the hypothetical model. This effect is most evident when large fractions of ^{14}C are incorporated into organic compounds that are more readily metabolized into gases such as ^{14}CO, $^{14}CO_2$, and $^{14}CH_4$.

6

FEATURE INTEGRATION

Chapter Summary

This chapter summarizes efforts that were undertaken to present new approaches to overall performance assessment. The new approaches were integrated into a performance assessment (PA) model and incorporate the findings of the previous chapters. The new approaches and data (Chapter 3) are briefly recapitulated and a discussion of our new overall performance assessment approach is provided. The chapter finishes with a description of sensitivity of a hypothetical PA to the parameters of the “new” model.

6.1 New Model Approaches Proposed by This Study

The new model developed in this study includes a gaseous release sub-model and a modeling distinction between the waste form and near-field geochemistry. Previous PA models have not integrated these new pieces into the overall PA. The following discussion summarizes Chapters 4 and 5.

6.1.1 Gaseous Release Model

Many of the waste materials that contain ^{14}C are susceptible to microbial attack and can release the activity through the generation of CO_2 and methane. Once gases are generated, they migrate to void spaces in waste containers and can be released by pumping effects of atmospheric pressure changes, if there is a gas release pathway such as vents and drains.

The effects of gas generation on the ^{14}C source term are shown in Figure 5-1. The effects are significant enough to decrease the source term and a hypothetical PA based upon these results shows a modest decrease over the base-case, Figure 5-2. Effects of gas generation are also dependent upon temperature, the water infiltration, initial water content in the waste, the rate of biodegradation, and the mechanism of leaching of ^{14}C from biodegradable materials.

6.1.2 A Distinction Between the Waste-Form and the Near-Field Geochemistry

Backfilling a LLW facility with either cement or soil can have a significant impact on the characteristics of radionuclide release and hold-up of radionuclides in the facility. For radionuclides to reach the bottom boundary of the disposal facility after being released from waste packages, they need to migrate through the backfill. This process will provide spreading and dilution of the plume that are governed by advection, dispersion, and the sorption characteristics of the backfill. If dilution and sorption in the backfill are significant, the source term is significantly reduced in any given period of time.

Geochemistry influences the release of radionuclides from wastes and their fate in the near-field. This is controlled by solubility limits in pore water, complexation with soil minerals, and sorption/retardation during transport.

6.2 Systems Approach

This section describes the alpha version of the computer code developed in this project. The code (RLESS) was used to summarize the outcome of all of the research activities into an integrated modeling approach. Figure 6-1 illustrates the analysis shown in the previous chapters and illustrates the potential benefits of a more rigorous approach to performance assessment. Show in Figure 6-1 is the base-case using previous modeling approaches and data (Chapter 2), improved inventory estimates (Chapter 3), and improved modeling of the source term and near-field chemistry (Chapter 4). The general features of these methods could be applied to any radionuclide of interest, although ^{14}C was the focus of this investigation.

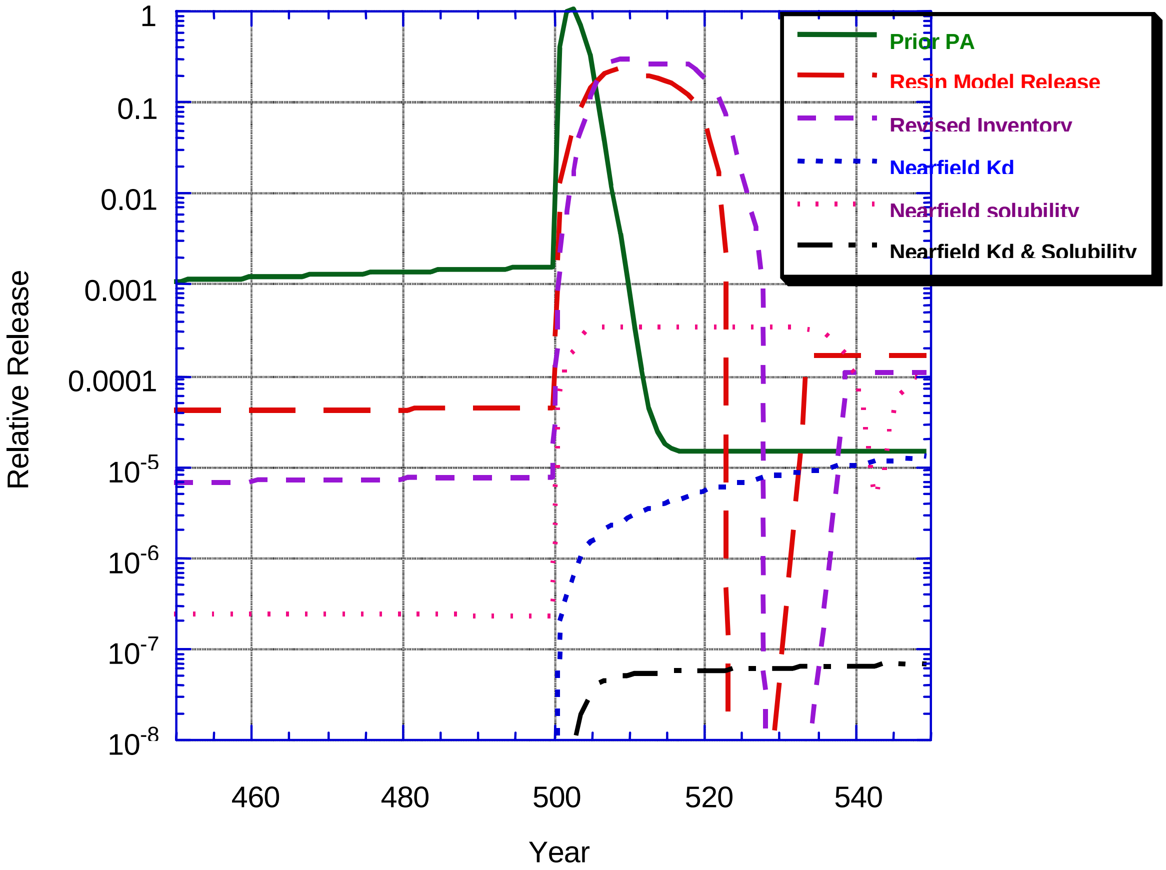

Figure 6-1
Carbon-14 Modeling Summary

As an integrated systems model, RLESS α, also facilitates probabilistic performance assessment approaches. NRC's Branch Technical Position (BTP) on performance assessment suggests the use of a probabilistic approach for uncertainty analysis of combined performance of a number of disposal facility attributes. By integrating and automating subsystem models (or code inputs and outputs) with an overall "system" code, ability to step through successive iterations of the process of evaluating a prospective site's performance and perform uncertainty and sensitivity analyses is significantly enhanced. For this purpose, a systems analysis capability is provided in the new computer model RLESS.

This alpha version of the integrated, new systems analysis model includes the following sub-models:

1. water infiltration with the failure of engineered barriers;
2. waste container failures;
3. radionuclide release from various waste forms;
4. near-field transport;
5. far-field transport, and;
6. human exposure pathways and dose calculations.

This alpha version of the new systems analysis model was used to develop the examples in this report. The disposal system was assumed to be an engineered facility that includes a soil cover system and concrete vault.

6.2.1 Brief Description of the Alpha Model

Modeling Infiltration with the Failure of Engineered Barriers

Modeling the infiltration of water into the waste region is based on the description of key processes and parameters, including:

- the saturated hydraulic conductivity of the cover system and concrete vault,
- the time and rate of failure of the cover system and concrete vault and
- the natural percolation flux at the site.

Modeling Waste Container Failures

Failures of waste containers are described by the time-to- failure approach. The time dependent propagation of the breached area of the container is represented by using specific types of cumulative probability functions to determine the size of the breach with time (termed the breach ratio). The model uses this breach ratio to predict the flow of infiltrating water into waste containers.

Modeling Radionuclide Release from Waste Forms

Release of radionuclides from various LLW materials and forms is predicted based on four different release models, i.e., surface rinse, diffusion, uniform dissolution, and a model for ion-exchange resins.

The rinse release model is based on the concept of surface wash-off by water and equilibrium partitioning between the water and the waste form. The uniform dissolution model assumes uniform, or congruent, dissolution of the waste form using constant release rates. The diffusion model uses the continuity laws at the boundary providing the capability to describe the effects of surrounding conditions on the diffusive release of radionuclides. The model describes two-dimensional diffusion, one-dimensional advection and radioactive decay.

The model for ion-exchange resin is based on the description of resin bead phase (phase 1) and the bulk pore water phase within waste containers (phase 2). In phase 1, radionuclides diffuse from small spherical bead particles to the liquid in containers. In phase 2, radionuclides diffuse from the liquid in the containers to outside of containers controlled by the advection, diffusion, and sorption. A numerical solution approach rather than an explicit analytical model is employed for both diffusion and ion-exchange resin releases to be more accurate. All of the releases are restricted by solubility limits.

Modeling Near-Field Transport

The near-field transport models use a one-dimensional advection-dispersion equation based on a numerical solution technique. Continuity of the concentration of radionuclides are applied as boundary conditions at all interfaces. The near-field transport models predict the release rate of radionuclides at the bottom boundary of a LLW vault.

Modeling Far-Field Transport

The far-field models use two-dimensional advection-dispersion equations based upon an analytical solution approach. This is similar to the approach employed in the PAGAN computer model [Kozak, et al., 1990].

Modeling Exposure Pathways/Human Dose

Radiation exposure of human beings from the waste material disposed in a LLW facility is described by the following pathways:

1. drinking water intake from a well,
2. Consumption of vegetables, fruits, grains which were raised with the contaminated irrigation water from the well,
3. Consumption of animal products (meat and milk) from the animals raised at a farm where contaminated grounwater was used for their drinking water and to raise the feed.

6.2.2 Probabilistic Analysis

A built-in capability for probabilistic Monte Carlo (MC) analysis is provided as part of the model. Distributions that can be used to describe input parameters in the model include normal, lognormal, triangle, uniform, loguniform, and beta. Random sampling and Latin Hypercube sampling techniques are available options that can be used for MC analysis.

6.3 Sensitivity Analysis

Using the systems analysis computer model, a sensitivity analysis was performed to examine the characteristics of the model and to identify major parameters of importance. The analysis is based on a simple single parameter sensitivity analysis where each parameter of interest is varied by 30% from their default input values. The sensitivity of results are defined as:

Table 6-1 shows the results of this analysis. The results indicate that by changing the value of the parameter in the first column of Table 6-1 by 30%, the resulting percentage change in the hypothetical dose is shown in column 2. Large absolute values in the second column indicate sensitive parameters. Zeros in the second column indicate insensitive parameters (or ones that have very little effect on the hypothetical dose.) Examples of input parameters that show very large sensitivity are presented below (the corresponding sensitivity is given in the parenthesis):

- Total activity inventory (29.96); {note, nearly a 1:1 correspondence between changes in this parameter and changes in the dose}
- Natural infiltration at the site (25.84);
- Average linear velocity in the aquifer (22.85);
- Depth of the contaminated region under the facility (22.85);
- Moisture content of soil (22.10);
- K_d of C-14 in the near-field (19.48);
- Density of soil in the near-field (19.48);
- Depth to water table (18.73);
- Height of the waste region (18.73), and;
- Kd of C-14 in the far-field (15.36).

Other parameters of importance identified from the sensitivity analysis include:

1. parameters related to the transport in the aquifer (longitudinal/transverse dispersivity in the aquifer and total porosity in the aquifer),
2. human consumption of contaminated food stuff (drinking water intake, fraction of contaminated water in human drinking, consumption of meat and milk),
3. inventory distributions for rinse release and resin release,
4. animal feed to meat or milk transfer factors, and,
5. consumption of contaminated feed by cattle.

The sensitivity of the inventory in different waste containers is shown to be relatively small. (1.87% for the carbon steel drum inventory, 0.37% for the carbon steel liners, and 4.12% for the HIC inventory). The sensitivity of the inventory for different release mechanisms is somewhat large (5.24% for the rinse release inventory, 3.37% for the dissolution release inventory, 1.50% for the diffusional release inventory, and 7.49% for the resin release inventory).

Figure 6-2 represents a separate sensitivity analysis for the solubility of C-14 in the near-field. In a facility where a cement backfill is not employed, the solubility of C-14 in the near-field is very high and not a controlling factor. But if the cement backfill is used, the solubility of C-14 could become small enough to be a limiting factor. This effect is shown in Figure 6-2. Below the solubility of 2×10^{-9} g/cm^3, the predicted dose was linearly dependent on the changes in the near-field solubility. This indicates the importance of near-field solubility of C-14 when cement backfill is utilized in the facility design.

Figure 6-3 shows another separate sensitivity analysis for the K_d value of C-14 in the near-field. This is also related to whether a cement backfill is employed or not. When the cement backfill is not used, it is assumed that the K_d value of C-14 takes a very small value (no sorption). But if the cement backfill is used, the K_d value of C-14 is expected to be much higher [Dames and Moore, 1995]. The results indicate that the dose is a strong function of the K_d value in the near-field.

Table 6-1
Results of Sensitivity Analysis of the Systems Analysis Performance Assessment Model

Column 1: Parameter	Column 2: Sensitivity
Facility area, m2	0
Height of the facility, cm	18.7
1/2 depth to the water table, cm	18.7
Kd of soil/near-field, ml/g	19.5
Dispersion coeff in soil, cm2	2.62
Density of soil/near-field	19.5
Moisture content of soil	22.1
Height of the container, cm	0.75
Time of complete soil cover failure	0
Time of complete concrete vault failure	0.37
Sat hydraulic conductivity of soil cover, cm/yr	0
Sat hydraulic conductivity of concrete vault, cm/yr	0
Natural infiltration at the site, cm/yr	25.8
Density of waste, g/cm3	0.37
Moisture content of waste	0.75
Drinking water intake, L/yr	7.87
Leafy vegy consumption, kg/yr	0.37
Produce consumption, kg/yr	4.87
Meat consumption, kg/yr	9.74
Milk consumption, L/yr	7.49
Inventory in the carbon steel drum, %	1.87
Inventory in the carbon steel liner, %	0.37
Inventory in the HIC, %	4.12
Inventory for rinse release, %	5.24
Inventory for dissolution release, %	3.37
Inventory for diffusion release, %	1.50
Inventory for resin release, %	7.49
Rate of dissolution, cm/yr	0
Diffusion coefficient in waste, cm2/yr	2.25
Diffusion coefficient in resin, cm2/yr	0
Average linear velocity in the aquifer, m/yr	22.8
Kd in the far-field, ml/g	15.4
Total porosity of the aquifer	11.2
Density of the aquifer, g/cm3	0
Longitudinal dispersivity in the aquifer, m	12.4
Transverse dispersivity in the aquifer, m	12.0
Solubility in the waste, g/cm3	0.37
Kd in the waste for rinse release, ml/g	0
Kd in the waste for dissolution release	0
Kd in the waste for diffusion release	0.37
Kd in the waste for resin release	0
Time of complete failure of carbon steel liners, yr	0
Time of complete failure of HIC, yr	4.87
Depth of the contaminated region under the facility, M	22.8
Total activity inventory, Ci	30.0
Fraction of contamination in human drinking water	7.87
Fraction of contamination in animal drinking water	7.12
Water uptake by milk cow, L/day	3.37
Water uptake by beef cattle, L/day	3.75
Feed to meat transfer, day/kg	9.74
Feed to milk transfer, day/L	7.49
Amount of pasture grass consumed by cattle, kg/d	5.24
Amount of stored feed consumed by cattle, kg/d	5.24

6.4 Conclusion

A comprehensive model of performance assessment has been drafted from the efforts of this research. It allows for complete system modeling and post-modeling analysis such as sensitivity analysis. Although RLESS is not a proven performance assessment code, it can become a very useful tool with additional code validation efforts in the iterations of site evaluation to provide guidance to site developers as to where improvements in the understanding will provide maximum benefits. A more user-friendly version is also needed for general distribution, but this is beyond the scope of the efforts described in this report.

7
CONCLUSIONS

Previous NRC research identified ^{14}C as a potential long term contributor to dose. This research has shown quantitatively that dose projections of ^{14}C can be refined by using the approaches developed in this report. The following describes the key issues addressed by this research and the conclusions reached for each area.

This research addressed the impact of:

- providing an inventory distribution database, assessing the need and impact of a gaseous release model,
- integrating a mechanistic source-term code into an overall systems code,
- developing a specific model for ion exchange [**IX**] resins, and other specific waste forms, and, developing near-field transport models to provide a distinction in geochemistry between the waste form and the near-field, such that different values of K_d and/or solubility could be assigned to a radionuclide for different parts of the calculation"?

on existing state-of-the-art performance assessment models in terms of their impact on ^{14}C dose projections.

An accepted state-of-the-art model was used as the benchmark and the effects of the research are shown relative to this prior benchmark. Since the effects of the improvements are often observed at different time scales, the graphs presented may change in terms of the scaling, but the data that represents the relative prior performance assessment calculation is consistent throughout.

The intention of this presentation of the research is to show the potential that may be realized in PA if the approaches presented are adopted. The magnitude of the results will change based upon specific PA conditions, and thus only qualitative benefits are highlighted.

More Detailed Description of the Inventory

An estimate of inventories based upon the mix of waste forms and classes, see Appendix B – Barnwell data, was conducted. The key finding was that significantly less ^{14}C was associated with Class A waste than had previously been assumed. The previous inventories with a large amount of ^{14}C in Class A predicted large releases whenever the facility boundary is breached (at 500 years, shown by the circles in the figure below). The more detailed inventories developed as part of this work showed the majority of ^{14}C in Class B and C waste which are contained in high-integrity containers and waste forms. This is predicted to result in more gradual release, and hence a much lower peak (see Figure 7-1 below). This effect is due only to a redistribution of the inventory, keeping all other performance assessment variables the same. This emphasizes the importance of accurate waste inventory measurements and the benefits more detailed measurements and inventory assessment may have on performance assessment predictions.

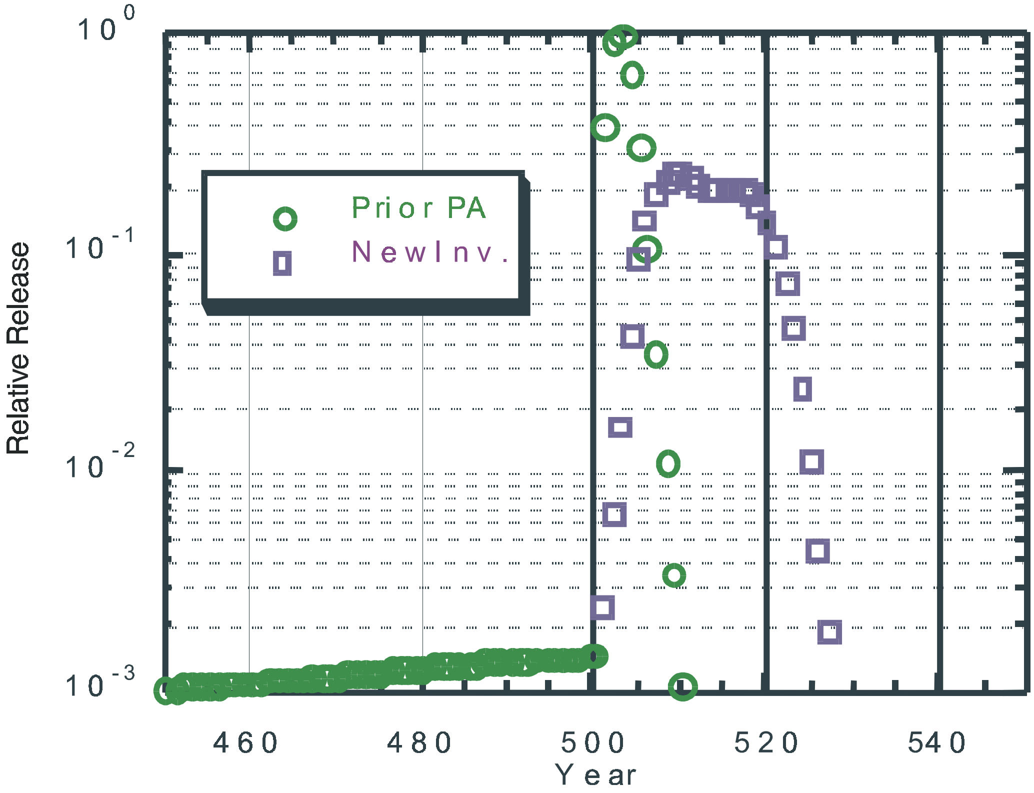

Figure 7-1
Effect on PA Results with Revised Inventory

Use of Mechanistic Models and Analysis

Mechanistic models have the potential to improve confidence in the predicted outcomes of conservative performance assessments. As shown in the following figure, the differences between the outcomes of an empirical, presumably conservative, surface rinse model predicts similar outcomes to the more detailed mechanistic model for the set of assumptions that were made. In the event that the rinse model predicts an unacceptable outcome, there is no recourse except for inventory reductions. A mechanistic approach allows for targeting the key parameters or processes that contribute to an outcome and improving understanding of the process. In some cases the mechanistic models and the empirical models may result in the same recommended actions based upon their results. The bottom line in terms of PA is that our knowledge of the behavior of resins has improved, but the impact on PA is not necessarily shown to be significant, as seen below:

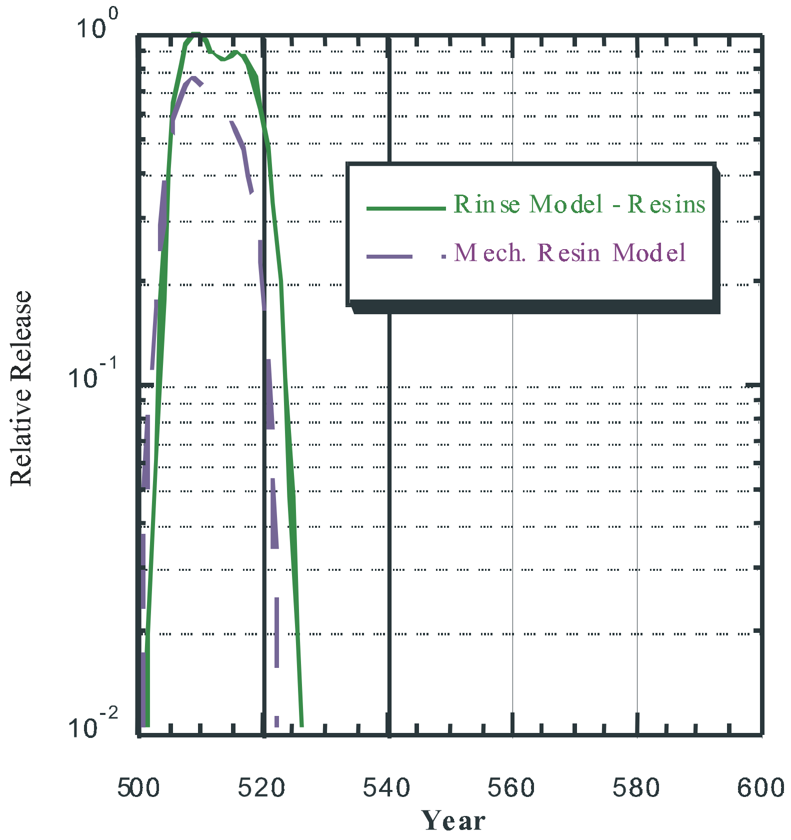

Figure 7-2
Comparison of a Simple “Rinse” Release Model with a Mechanistic Resin Model

Including a Gaseous Release Model

Including a gaseous release model reduces the inventory of ^{14}C available for transport in ground water. This generally translates into an overall equivalent reduction in dose compared to the benchmark performance assessment case, (current PA does not account for the gas pathway). Modest, yet quantifiable, reductions are observed in the peak dose, see Figure 7-3 below.

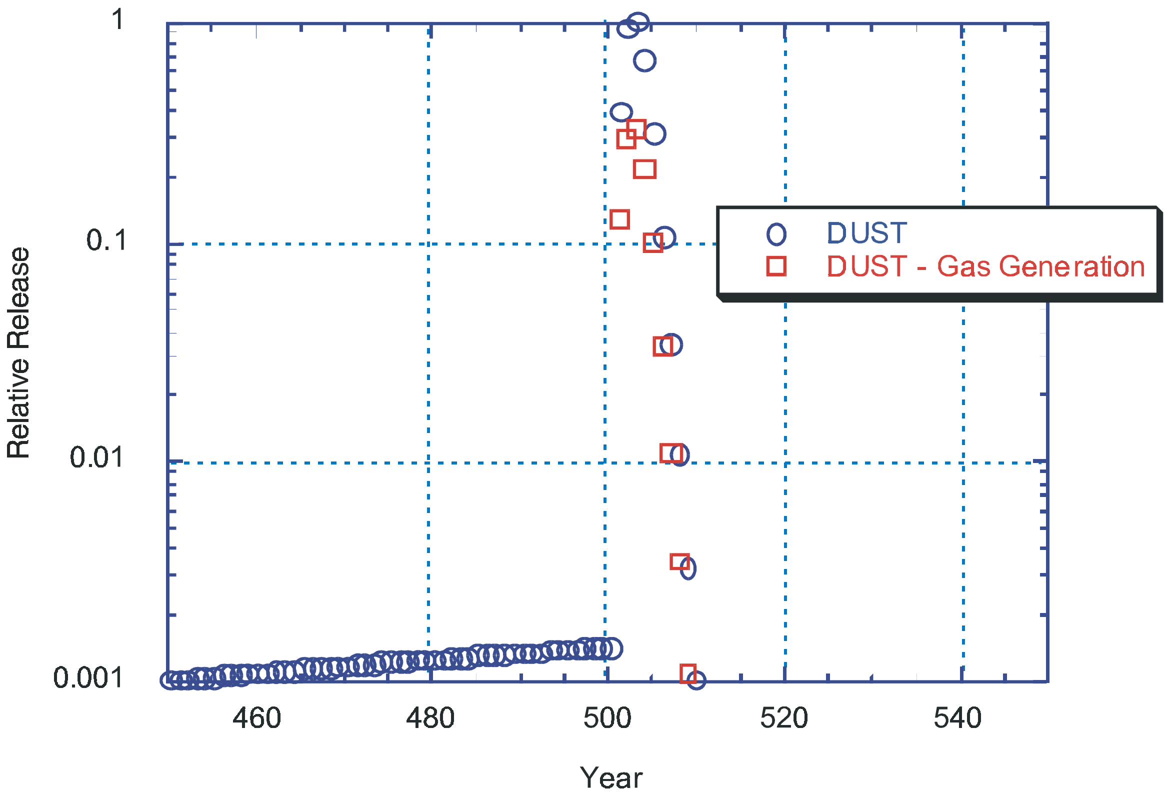

Figure 7-3
Effect of Gas Generation on PA

The intention of this presentation of the research is to show the potential that may be realized in PA if the approaches presented are adopted. The magnitude of the results will change based upon specific PA conditions, however the basis for these approaches are spelled out in detail in the report.

The alpha version of a comprehensive model (RLESS) including these aspects of performance assessment allowed us to describe the efforts of this research in the figures showed above and throughout the report. It allows for complete system modeling and post-modeling analysis such as sensitivity analysis. Although RLESSα is not a proven performance assessment code, it can become a very useful tool with additional code validation efforts. It can be used in the iterations of site evaluation to provide guidance to site developers as to where improvements in the understanding will provide maximum benefits. However, a more user-friendly version is needed for general distribution.

8
REFERENCES

Banba, T., J. Matsumoto, and S. Muraoka, “Leach Behavior of Carbon-14 Contained in Portland Cement,” Cement and Concrete Research, Vol. 22, pp. 381-386, 1992.

Bonka, H., K. Brussermann, and G. Schwarz, Umweitbelatung durch Radiokolenstolf aus Kerntechnichen Analgen, Reactortagung, Berlin, 1974.

Codell, R. B., K. T. Key, and G. Whelan, “A Collection of Mathematical Models for Dispersion in Surface Water and Groundwater, NUREG-0868, U.S. Nuclear Regulatory Commission, 1982.

Crank, J., *The Mathematics of Diffusion*, Oxford University Press, 1957.

Dames & Moore, “Waste Inventory Revision 3, North Carolina Low-Level Radioactive Waste Disposal Facility Project,” DM/DR-002, prepared for Chem-Nuclear Systems, Inc. by Dames & Moore, December 1995.

Davis, W., “Carbon-14 Production in Nuclear Reactors,” in Management of Low-Level Radioactive Waste, edited by M. W. Carter, et.al., Pergamon Press, 1979.

Eichholz, G. G., M. F. Petelka, and J. Whang, “Leaching and Migration of Low-Level Wastes Under Unsaturated Conditions,” Radioactive Waste Management and the Nuclear Fuel Cycle, Vol. 14, pp. 285-310, 1990.

Electric Power Research Institute, “Radwaste Generation Survey Update,” Volume 1 & 2, EPRI NP-5526, Prepared by Analytical Resources, Inc., 1988.

Electric Power Research Institute, “Management Strategies for Treatment and Disposal of Utility-Generated Low-Level Radioactive Waste,” EPRI NP-6225, Prepared by Rogers and Associates Engineering Corporation, 1989.

Electric Power Research Institute, “Soil-to Plant Transfer of Carbon-14 for Environmental Assessment of Radioactive Waste Repositories,” EPRI NP-6946, 1990.

Electric Power Research Institute, “Characterization of Carbon-14 Generated by the Nuclear Power Industry,” EPRI TR-105715, 1995.

Electric Power Research Institute, “Performance Assessment and LLW Characterization,” in Low Level Waste Characterization Guidelines, EPRI TR-107201, 2691, November 1996.

England, R. L., N. W. Kline, K. J. Ekblad, and R. G. Baca, “MAGNUM-2D Computer Code: User’s Guide,” RHO-CR-143 P, Rockwell Hanford Operations, 1986.

Fayer, M. J. and T. L. Jones, “UNSAT-H Version 2.0: Unsaturated Soil Water and Heat Flow Model,” PNL-6779, Pacific Northwest Laboratory, 1990.

Fields, D. E. and C. W. Miller, “User’s Manual for DWNWND - An Interactive Implementation of the Gaussian Plume Atmospheric Transport Model with Eight Dispersion Parameter Options,” ORNL/TM-6874, Oak Ridge National Laboratory, 1980.

Fowler, T. W. and C. B. Nelson, “Health Impact Assessment of Carbon-14 Emissions from Normal Operations of Uranium Fuel Cycle Facilities,” EPA 520/5-80-004, U. S. Environmental Protection Agency, 1981.

Golder Associates, RIP Performance Assessment and Strategy Evaluation Model, Theory Manual and User’s Guide, 1996.

Golder Associates, FRACMAN.

Gruhlke, J. M., J. Neiheisel, and L. Battist, “Estimates of the Quantities, Form, and Transport of Carbon-14 in Low-Level Radioactive Waste,” U. S. Environmental Protection Agency, 1986.

Guppy, R., “Autogenous Healing of Cracks in Concrete and its Relevance to Radwaste Repositories,” NSS/R-105, Nirex, United Kingdom, 1988.

Gureghian, A. B., “A Two-Dimensional Finite Element Model for the Simultaneous Transport of Water and Reacting Solutes Through Saturated and Unsaturated Porous Media,” Technical Report ONWI-465, 1983.

Gureghian, A. B., “Analytical Solution for Multidimensional Transport of Four-Member Radionuclide Decay Chain in Ground Water,” BMI/OCRD-25, 1987.

Habayeb, M. A., “Carbon-14 Releases from Solidified IRN-150 Moderator Resin Wastes,” Nuclear and Chemical Waste Management, Vol. 5, pp. 351-357, 1985.

Healy, R. W., “Simulation of Solute Transport in Variably Saturated Porous Media with Supplemental Information on Modifications to the U.S. Geological Survey’s Computer Program VS2D,” Water Resources Investigation Report 90-4025, U.S. Geological Survey, 1990.

Hoch, A. R., C. C. Naish, S. M. Sharland, A. C. Smith, and K. J. Taylor, “Experiments and Modeling Studies Concerning Localized Corrosion of Carbon Steel and Stainless Steel Containers for Intermediate- and Low-Level Radioactive Waste,” Materials Research Society Symposium Proceedings, Vol. 294, 1993.

Hung, C. Y., “A Model to Simulate Infiltration of Rainwater through the Cover of a Radioactive Waste Trench under Saturated And Unsaturated Conditions,” in <u>Role of the Unsaturated Zone in Radioactive and Hazardous Waste Disposal</u>, J. W. Mercer et. al. (editor), Ann Arbor Science, 1983.

Huyakorn, P. S., J. B. Kool, and J. B. Robertson, "Documentation and User's Guide: VAM2D - Variably Saturated Analysis Model in Two Dimensions," NUREG/CR-5352, HGL/89-01, U. S. Nuclear Regulatory Commission, 1989.

ICRP, "Limits of Intakes of Radionuclides by Workers," ICRP Publication, Part 3, Pergamon Press, 1978.

ICRP, "Limits for Intakes of Radionuclides by Workers," ICRP Publication 30, Part 1, Pergamon Press, New York, 1982.

ICRP, "Recommendations of the International Commission on Radiological Protection," ICRP Publication 60, Pergamon Press, New York, 1990.

Kennedy, W. E., "General Perspective on Performance Assessment," Proceedings of the 18th DOE LLW Management Conference, Salt Lake City, Utah, May 20-22, 1997.

Knisel, W. G., "CREAMS: A Field Scale Model for Chemicals, Runoff, and Erosion from Agricultural Management Systems," U.S. Department of Agriculture Conservation Research Report No. 26, 1980.

Kline, N. W., R. L. England, and R. G. Baca, "CHAINT Computer Code: User's Guide," RHO-CR-144 P, Rockwell Hanford Operations, 1986.

Kozak, M. W., C. P. Harlan, M. S. Y. Chu, and B. L. O'Neal, "Background Information for the Development of a Low-Level Waste Performance Assessment Methodology - Selection and Integration of Models," NUREG/CR-5453, SAND89-2509, Vol. 3, U. S. Nuclear Regulatory Commission, 1989.

Kozak, M. W., M. S. Y. Chu, P. A. Mattingly, J. D. Johnson, and J. T. McCord, "Background Information for the Development of a Low-Level Waste Performance Assessment Methodology - Computer Code Implementation and Assessment," NUREG/CR-5453, SAND90-0375, Vol. 5, U. S. Nuclear Regulatory Commission, 1990.

Kozak, M. W., N. E. Olague, D. P. Gallegos, and R. R. Rao, "Treatment of Uncertainty in Low-Level Waste Performance Assessment," Proceedings of the 13th DOE LLW Management Conference, Atlanta, GA, November 19-21, 1991.

Kozak, M. W., N. E. Olague, R. R. Rao, and J. T. McCord, "Evaluation of a Performance Assessment Methodology for Low-Level Radioactive Waste Disposal Facilities," NUREG/CR-5927, SAND91-2802, Vol. 1, U. S. Nuclear Regulatory Commission, 1993.

Krishnamoorthy, T. M., S. N. Joshi, G. R. Doshi, and R. N. Nair, "Desorption Kinetics of Radionuclides Fixed in Cement Matrix," Nuclear Technology, Vol. 104, pp. 351-357, 1993.

Kunz, C. O., "Radioactive Gas Production and Venting at a Low-Level Radioactive Burial Site," Nuclear and Chemical Waste Management, Vol. 3, pp. 185-190, 1982.

Lappala, E. S., R. W. Healy, and E. P. Weeks, “Documentation of Computer Program VS2D to Solve the Equations of Fluid Flow in Variably Saturated Porous Media,” Water Resources Investigation Report 83-4099, U. S. Geological Survey, 1987.

Leigh, C. D., B. M. Thompson, J. E. Campbell, D. E. Longsine, R. A. Kennedy, and B. A. Napier, “User’s Guide for GENII-S: A Code for Statistical and Deterministic Simulations of Radiation Doses to Humans from Radionuclides in the Environment,” SAND91-0561, Sandia National Laboratories, 1992.

MacDonald, M. G. and A. W. Harbaugh, “A Modular Three-Dimensional Finite-Difference Ground-Water Flow Model,” U.S. Geological Survey Techniques of Water Resources Investigations, Book 6, Chapter A1, 1988.

Matsumoto, J., T. Banba, and S. Muraoka, “Adsorption of Carbon-14 on Mortar,” Mat. Res. Soc. Symp. Proc. Vol. 353, pp. 1029-1035, 1995.

McKinnon, R. J., T. M. Sullivan, C. J. Suen, and S. A. Simonson, “BLT-EC (Breach, Leach, Transport, and Equilibrium Chemistry), a Finite-Element Model for Assessing the Release of Radionuclides from Low-Level Waste Disposal Units: Background, Theory, and Model Description,” NUREG/CR-6305, U. S. Nuclear Regulatory Commission, 1995.

Moore, R. E., C. F. Baes, L. M. McDowell-Boyer, A. P. Watson, F. O. Hoffman, J. C. Pleasant, and C. W. Miller, “AIRDOS-EPA: A Computerized Methodology for Estimating Environmental Concentrations and Dose to Man from Airborne Releases of Radionuclides,” ORNL-5532, Oak Ridge National Laboratory, 1979.

Napier, B. A., D. L. Strenge, R. A. Peloquin, and J. V. Ramsdell, “GENII - The Hanford Environmental Radiation Dosimetry Software System,” PNL-6584 Vol. 1 & 2, Pacific Northwest Laboratory, 1988.

NCRP, “Carbon-14 in the Environment,” NCRP Report No. 81, National Council on Radiation Protection and Measurement, 1985.

Ng, Y. C., C. A. Burton, S. E. Thompsons, R. K. Tandy, H. K. Kretner, M. W. Pratt, “Prediction of the maximum dosage to man from the fallout of nuclear devices. IV. Handbook for estimating the maximum internal dose from radionuclides released to the biosphere,” UCRL-50163, Lawrence Radiation Laboratory, University of California, 1968.

Nichols, W. E. and M. D. White, “MSTS: Multiphase Subsurface Transport Simulator User’s Guide and Reference,” PNL-8637, Pacific Northwest Laboratory, 1993.

North Carolina Regulations for Protection against Radiation, 15A NCAC 11, N. C. Department of Environment, Health, and Natural Resources, Division of Radiation Protection, July, 1993.

Oblath, S. B., “Leaching from Solidified Waste Forms under Saturated and Unsaturated Conditions,” Environmental Science and Technology, Vol. 23, pp. 1098-1102, 1989.

Olague, N. E., D. E. Longsine, J. E. Campbell, C. D. Leigh, "User's Manual for the NEFTRAN II Computer Code," NUREG/CR-5618, SAND90-2089, U. S. Nuclear Regulatory Commission, 1991.

O'Neal, B. L. and C. E. Lee, "IMPACTS-BRC, Version 2.0," NUREG/CR-5517, SAND89-3060, U. S. Nuclear Regulatory Commission, 1990; O. I. Oztunali and G. W. Roles, "Update of Part 61, Impacts Analysis Methodology," NUREG/CR-4370, U.S. Nuclear Regulatory Commission, 1986.

Parks, B. S., "User's Guide for CAP88-PC, Version 1.0," 402-B-92-001, U. S. Environmental Protection Agency, 1992.

Petelka, M. F., "Leaching of Radioactive Waste Forms under Saturated and Unsaturated Flow Conditions," Ph.D. Thesis, Georgia Institute of Technology, 1987.

Pollock, D. W., "Documentation of Computer Programs to Compute and Display Pathlines Using Results from the U.S. Geological Survey Modular Three-Dimensional Finite-Difference Ground-Water Flow Model," U.S. Geological Survey Open File Report 89-381, 1989.

Rogers, V. and C. Hung, "PRESTO-EPA-CPG: A Low-Level Radioactive Waste Environmental Transport and Risk Assessment Code, Documentation and User's Manual," EPA 520/1-87-026, U.S. EPA, December 1987.

Rood, A. S., "GWSCREEN: A Semi-Analytical Model for Assessment of the Groundwater Pathway from Surface or Buried Contamination," EGG-GEO-10797, Rev. 2, 1994.

Runchal, A. K. and B. Sagar, "PORFLOW: A Multifluid Multiphase Model for Simulating Flow, Heat Transfer, and Mass Transport in Fractured Porous Media, User's Manual - Version 2.41," NUREG/CR-5991, CNWRA 92-003, U.S. Nuclear Regulatory Commission, 1993.

Sagendorf, J. F., J. T. Goll, and W. F. Sandusky, "XOQDOQ: Computer Program for the Meteorological Evaluation of Routine Effluent Releases at Nuclear Power Stations," NUREG/CR-2919, PNL-4380, 1982.

Schroeder, P. R., T. S. Dozier, P. A. Zappi, B. M. McEnroe, J. W. Sjostrom, and R. L. Peyton, "The Hydrologic Evaluation of Landfill Performance (HELP) Model: Engineering Documentation for Version 3," EPA/600/9-94/168, U. S. Environmental Protection Agency Risk Reduction Engineering Laboratory, 1994.

Shuman, R., V. C. Rogers, N. Chau, and G. B. Merrell, "The BARRIER Code: A Tool for Estimating the Long-Term Performance of Low-Level Radioactive Waste Disposal Facilities," EPRI NO-7167-CCML, Electric Power Research Institute, May 1991.

Shuman, R., N. Chau, and V. C. Rogers, "Improved Modeling of Engineered Barriers for Low-Level Waste Disposal," Waste Management '91, Vol. 2, pp. 757-762, 1991.

Simmons, C. S., C. T. Kincaid, and A. E. Reisenauer, “A Simplified Model for Radioactive Contaminant Transport: the TRNSS Code,” PNL-6029, Pacific Northwest Laboratory, 1986.

Simonson, S. A. and M. S. Yim, “Verification of Engineering Assumptions in Modeling of Airborne Release of Carbon-14,” Proceedings of 1996 EPRI International LLW Conference, EPRI TR-106929, pp. 203-214, 1996.

Snyder, K. A. and J. R. Clifton, “4SIGHT Manual: A Computer Program for Modeling Degradation of Underground Low Level Waste Concrete Vaults,” NISTIR 5612, National Institute of Standards and Technology, June 1995.

Strenge, D. L., R. A. Peloquin, and G. Whelan, “LADTAP II - Technical Reference and User Guide,” NUREG/CR-4013, PNL-5270, U.S. Nuclear Regulatory Commission, 1986.

Sudicky, E. A. and E. O. Frind, “Contaminant Transport in Fractured Porous Media: Analytical Solutions for a System of Parallel Fractures,” Water Resources Research, Vol. 18, No. 6, pp. 1634-1642, 1982.

Sullivan, T. M. and C. J. Suen, “Low-Level Waste Shallow Land Disposal Source Term Model: Data Input Guides,” NUREG/CR-5387, BNL-NUREG-52206, U. S. Nuclear Regulatory Commission, 1989.

Sullivan, T. M., “Selection of Models to Calculate the LLW Source Term,” NUREG/CR-5773, BNL-NUREG-52295, U. S. Nuclear Regulatory Commission, 1991.

Sullivan, T. M., “Disposal Unit Source Term (DUST) Data Input Guide,” NUREG/CR6041, BNL-NUREG-52375, U. S. Nuclear Regulatory Commission, 1993.

Tang, D. H., E. O. Frind, and E. A. Sudicky, “Contaminant Transport in Fractured Porous Media: Analytical Solution for a Single Fracture,” Water Resources Research, Vol. 17, No. 3, pp. 555-564, 1981.

Travis, B. J., “TRACR3D: A Model of Flow and Transport in Porous/Fractured Media,” LA-9667-MS., Los Alamos National Laboratory, 1984.

U. S. Environmental Protection Agency, “Draft Environmental Impact Statement for Proposed Rules, Vol. 1 Background Information Document, Low-Level and NARM Radioactive Wastes,” EPA 520/1-87-012-1, U. S. EPA, 1988.

U. S. Nuclear Regulatory Commission, “Calculation of Annual Doses to Man from Routine Releases of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I,” Regulatory Guide 1.109, Revision 1, 1977.

U. S. Nuclear Regulatory Commission, “Draft Environmental Impact Statement on 10CFR Part 61,” NUREG-0782, Vol. 4, U. S. Nuclear Regulatory Commission, 1981.

U.S. Nuclear Regulatory Commission, Draft Environmental Impact Statement on 10 CFR Part 61, “Licensing Requirements for Land Disposal of Radioactive Waste,” NUREG-0782, Vol. 1 & 2, September (1981a).

U.S. Nuclear Regulatory Commission, Code of Federal Regulations, Title 10, Chapter 1, Part 61 “Licensing Requirements for Land Disposal of Radioactive Waste,” Fed. Registr. 47, 57446, 1982.

U. S. Nuclear Regulatory Commission, Private Communications, 1996.

U. S. Nuclear Regulatory Commission, “Branch Technical Position on a Performance Assessment Methodology for Low-Level Radioactive Waste Disposal Facilities,” NUREG-1573, 1997.

Voss, C. I., “A Finite-Element Simulation Model for Saturated-Unsaturated Fluid-Density-Dependent Ground Water Flow with Energy Transport or Chemically-Reactive Single-Species Solute Transport,” U.S. Geological Survey Water-Resource Investigations Report 84-4369, 1984.

Walton, J. C., “Performance of Intact and Partially Degraded Concrete Barriers in Limiting Mass Transport,” NUREG/CR-5445, EGG-2662, U. S. Nuclear Regulatory Commission, 1992.

Walton, J. C., L. E. Plansky, and R. W. Smith, “Models for Estimation of Service Life of Concrete Barriers in Low-Level Radioactive Waste Disposal,” NUREG/CR-5542, EGG-2597, U. S. Nuclear Regulatory Commission, 1990.

White, A., Handler, P., Smith, E. L., and Stehen, D., Principles of Biochemistry, 2nd Edition, McGraw-Hill, New York, 1973.

Yeh, G. T. and D. S. Ward, “FEMWATER: A Finite Element Model of Water Flow Through Saturated-Unsaturated Porous Media,” ORNL-5567, Oak Ridge National Laboratory, 1980.

Yeh, G. T. and D. S. Ward, “FEMWASTE: A Finite-Element Model for Waste Transport Through Saturated-Unsaturated Porous Media,” ORNL-5601, Oak Ridge National Laboratory, 1981.

Yim, M.-S. and S.-I. Su, “Source Term Model Evaluations for the Low-Level Waste Facility Performance Assessment,” 17th DOE LLW Management Conference, December 12-14, Phoenix, Arizona, U.S. Department of Energy, 1995a.

Yim, M. S. and S. A. Simonson, “Generation and Release of Radioactive Gases in LLW Disposal Facilities,” Proceedings of the 23rd DOE/NRC Nuclear Air Cleaning and Treatment Conference, NUREG/CP-0141, CONF-940738, 1995b.

Yim, M.-S., “Uncertainty Characterization of EPA’s Groundwater Transport Model for Low Level Waste Performance Assessment,” Transactions of American Nuclear Society, Vol. 73, pp. 85-87, 1995c.

Yim, M.-S., “Carbon-14 Research: Investigation of Technical Issues in C-14 Performance Assessment,” Proceedings of EPRI International LLW Conference, EPRI TR-106929, pp. 195-201, 1996a.

Yim, M.-S., S. A. Simonson, and T. M. Sullivan, “Investigation of C-14 Release from a Generic LLW Disposal Facility,” Nuclear Technology, Vol. 114, No. 2, 1996b.

Yim, M.-S., “Effects of Gas Generation on Low-Level Waste C-14 Source Term,” Transactions of American Nuclear Society, Vol. 75, pp. 59-61, 1996c.

Yim, M. S., “Evaluation of Uncertainties in Low-Level Waste Source Term Analysis,” Proceedings of the 18th DOE LLW Management Conference, Salt Lake City, Utah, May 20-22, 1997.

Yu, C., A. J. Zielen, J.-J. Cheng, Y. C. Yuan, L. G. Jones, et al., “Manual for Implementing Residual Radioactive Material Guidelines Using RESRAD, Version 5.0,” ANL/EAD/LD-2, Argonne National Laboratory, 1993.

Chem Nuclear Systems, Inc., Wake/Chatham County Site Safety Analysis Report, (November, 1994).

Olague, N. E., D. E. Longsine, J. E. Campbell, C. D. Leigh, “User’s Manual for the NEFTRAN II Computer Code,” NUREG/CR-5618, SAND90-2089, Sandia National Laboratory (February 1991).

Oztunali, O. I. and G. W. Roles, “Update of Part 61, Impacts Analysis Methodology,” NUREG/CR-4370, U. S. Nuclear Regulatory Commission (January 1986).

Rogers, V. and C. Hung, “PRESTO-EPA-CPG: A Low-Level Radioactive Waste Environmental Transport and Risk Assessment Code, Documentation and User’s Manual,” EPA 520/1-87-026, U.S. EPA (December 1987).

Sullivan, T. M. and C. J. Suen, “Low-Level Waste Shallow Land Disposal Source Term Model: Data Input Guides,” NUREG/CR-5387, BNL-NUREG-52206, Brookhaven National Laboratory (July, 1989).

Sullivan, T. M., “Selection of Models to Calculate the LLW Source Term,” NUREG/Cr-5773, BNL-NUREG-52295, Brookhaven National Laboratory (October, 1991).

Sullivan, T. M., “Disposal Unit Source Term (DUST) Data Input Guide,” NUREG/CR6041, BNL-NUREG-52375, Brookhaven National Laboratory (May 1993).

U. S. NRC, “Draft Environmental Impact Statement on 10CFR Part 61,” NUREG-0782, Vol. 4, U. S. Nuclear Regulatory Commission (September 1981).

U. S. EPA, “Draft Environmental Impact Statement for Proposed Rules, Vol. 1 Background Information Document, Low-Level and NARM Radioactive Wastes,” EPA 520/1-87-012-1, U. S. EPA (June 1988).

9
GLOSSARY

Adsorption - The adhesion of molecules of gases, liquids, or dissolved substances to the surface of a solid body with which they are in contact.
geochemistry

Advection - The process by which contaminants are transported by the bulk motion of the flowing groundwater.

Aerobic - living or occurring only in the presence of oxygen.

Anaerobic - living or occurring only in the absence of free oxygen.

Aquifer - A formation (or groups of formation) of water-bearing stratum that contains sufficiently permeable rock, sand, or gravel to yield significant quantifies of water to wells and springs.

Average pore velocity, average linear velocity - The measure of seepage movement as defined as Darcy velocity divided by the effective porosity.

Barnwell - The state-licensed low level waste disposal site at Barnwell, South Carolina operated by Chem-Nuclear Systems, Inc.

Biosphere - The part of the earth and its atmosphere in which living things exist.

Cartridge filters - Filter commonly used in pressurized water reactor liquid water processing systems to remove insoluble material. Cotton, nylon, and epoxy-impregnated paper are among the materials commonly used in fabrication of disposable cartridges for nuclear power plants.

Class A waste - A specific class of low level waste defined in 10 CFR 61.

Class B waste - A specific class of low level waste defined in 10 CFR 61. Class B waste contains more activity (typcially short-lived) than Class A waste.

Class C waste A specific class of low level waste defined in 10 CFR 61. Class C waste contains more activity (typcially long-lived) than Class A or Class B waste.

Congruent release - Release takes place coincidingly with the dissolution or corrosion of materials.

Corrosion - the deterioration of mettalic structure, usually with the loss of metal to solution.

Cosmic radiation - The radiation that originate in outer space and impinge on the top of the earth's atmosphere.

Darcy velocity (specific discharge, superficial linear velocity) - Volume of discharge of water per unit of bulk area of a porous medium per unit time.

Demineralizer - An ion exchange vessel used to prepare very low conductivity (demineralized) water. The term is often used to mean any ion exchange vessel used for water cleanup.

Deterministic model - Nonstochastic model, a model whose output is fixed by the mathematical form of its equations and the selection of a single value for each input parameter.

Diffusion - Gradual movement of molecules as a result of random thermal motion. Species move from higher to lower concentration regions.

Dispersion - The spreading of a stream or discrete volume of contaminants as it flows in a porous material.

Dissolution - Indicates the process of release as substances are chemically dissolved or corroded.

Distribution coefficient (Kd, partitioning factor) - The quantity of radionuclide sorbed by a solid per unit weight of the solid, divided by the concentration of the radionuclide dissolved in water.

Dose - A general term denoting the quantity of radiation or energy absorbed. In this report, dose refers to dose equivalent which is the quantity that expresses the effects of all radiation on a common scale for calculating the effective absorbed dose. It is defined as the product of the absorbed dose, the quality factor (radiation weighting factor), and other modifying factors. The unit of dose equivalent is rem. The corresponding SI unit is the sievert (Sv), where 100 rem = 1 Sv.

Dry active waste - Dry, solid, low level radioactive waste.

Electrolyte - Chemical compounds which dissociate or ionize in water to produce a solution which will conduct an electric current.

Equilibrium - A reversible interaction in which the forward and reverse rates of change reach a point where the concentrations of the reactants and products remain constant.

Far-field - Area where natural phenomena dominate in the groundwater transport of radionuclides not affected by the presence of waste, engineered design, and their interactions.

Fracture flow - Groundwater flow through a fractured medium. The medium itself may be porous and permeable, but the flow would be dominated by fractures, cracks, or solution cavities.

Geosphere - The geological systems that connect the biosphere and the radiological source term through groundwater transport.

High integrity containers - specially design low level waste container with minimum structural lifetime of 300 years. The type of high integrity containers expected to be used in the future is the combination of high density polyethylene and concrete overpack.

Hydraulic conductivity - The volume of water that will move per unit time under a unit gradient through a unit cross-sectional area perpendicular to the direction of flow.

Infiltration - Indicates the percolation of precipitated water into the deeper ground media after the runoff, evaporation, and evapotranspiration take place.

Ion - An electrified particle formed when a neutral atom or group of atoms gains or loses one or more electrons.

Ion exchange - A reversible process by which ions are interchanged between an insoluble material and a liquid with no substantial structural changes of the material.

Ion exchange resins - A synthetic, organic insoluble material that has the ability to exchange, reversibly, certain ions in its structure or attached to its surfaces as functional groups, with ions in a surrounding medium.

Leaching - The process of removing soluble constituents from a material by the reaction of a percolating liquid.

Linear isotherm - Description of the relationship between the concentration of contaminants in the liquid phase and the solid phase based on an assumption of instant local equilibrium between the two phase.

Low level waste - Those waste containing sources, special nuclear or byproduct material that are acceptable for disposal in a land disposal facility. It is defined as the radioactive waste not classified as spent nuclear fuel, high-level radioactive waste, transuranic waste, or uranium/thorium mill tailings.

Manifest - Documents that must accompany shipments of low level waste, hazardous waste, and mixed waste.

Mechanistic model - A model that is capable of describing the fundamental underlying mechanisms (chemical and/or physical) of the processes involved.

Model - A physical or mathematical representation of reality.

Moisture content - The fraction of total volume occupied by the water.

Near-field - Region in which waste characteristics and disposal facility (repository) phenomena dominate the transport of radionuclides in groundwater.

Oxidation potential - A measure of ability of an environment to supply electrons to an oxidizing agents, or to take up electrons from a reducing agent.

Parameter - Any one of a set of variables in a model whose values determine model predictions.

Pathway - The route by which a radionuclide travels to give a radiation dose to humans.

Porosity - Total porosity is defined as the ratio of the volume of voids (interstices) to total volume.

Retardation - Slowing down of contaminant movement in groundwater due to sorption.

Reverse osmosis - A water treatment process in which the liquid is forced through a membrane by the application of pressure leaving the impurities behind.

Richland - The state-licensed low level waste disposal site at Richland, Washington operated by U. S. Ecology.

Rinse - Indicates the process of release of substances from surface wash-off.

Saturated zone - That portion of porous ground media where the interconnecting voids (interstices) are filled with water.

Site-specific data - Data, collected for use in radiological assessment models, applicable to the particular location for which the assessment is being performed.

Solubility limit - The limit that reflects the extents to which the reactants (radionuclide in solid) or products (ions) are favored in a dissolution-precipitation reaction.

Sorbent - denotes absorbent waste form materials used for the packaging of low level waste.

Sorption - Interactions that cause radionuclides to migrate at a slower rate than the groundwater itself in transport in the groundwater system.

Source term - The quantity of radionuclide release at the bottom boundary of the disposal unit (performance assessment definition).

Unsaturated zone - The portion of porous medium in the ground where the interconnecting voids (interstices) are only partially filled with fluid.

Valence - The electrical charge that an atom in an ion or molecule has.

A

PERFORMANCE ASSESSMENT MODELS FOR LOW LEVEL RADIOACTIVE WASTE DISPOSAL FACILITIES: A REVIEW

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Abstract

The statutory role of performance assessment of radioactive waste disposal facilities is a technical analysis to evaluate the prospective performance of a disposal site with respect to regulatory compliance. The areas that are covered by performance assessment range from the characterization of the waste and site to estimation of any potential dose to the public due to release of radionuclides over long periods of time (typically thousands of years). In developing new disposal sites, performance assessment has also been used as a screening tool for determining site suitability, a guide in site characterization, as well as a final technical analysis for license application. This paper provides a review of the current status of performance assessment models in the U.S. for civilian low-level radioactive waste (LLW) disposal facilities.

Overview of Performance Assessment

Communities that ultimately accept radioactive or hazardous material storage/disposal sites will need adequate assurance that these sites will not become environmental issues for future generations. Of primary concern is the protection of human health which is evaluated through analysis of the performance of subsystems of engineered sites that will be used to contain radioactive materials and how well they maintain containment. Assessment of radiological safety of these sites is termed “performance assessment”.

The process of conducting performance assessment of radioactive waste disposal facilities is an iterative technical analysis to evaluate the prospective performance of a disposal site during the post-closure phase with respect to regulatory compliance. The primary objectives of performance assessment are[1]: 1) to determine whether reasonable assurance of compliance with quantitative performance objectives can be demonstrated, 2) to identify data, design, and model development needs for reaching defensible decisions about regulatory compliance, and 3) to identify waste acceptance criteria related to quantities of wastes for disposal.

In performance assessment, site-specific data on the characteristics of the site, wastes, and facility are processed in computer models as conceptual representation of physical processes/systems. The results are interpreted as conservative projections of consequences with the aim of providing reasonable assurance that the disposal system will perform in a manner that is in compliance with regulatory requirements. Performance assessment results do not necessarily represent the predictions of expected or actual outcomes. A hypothetical depiction of a facility and the key areas for performance assessment are shown in Figure A-1.

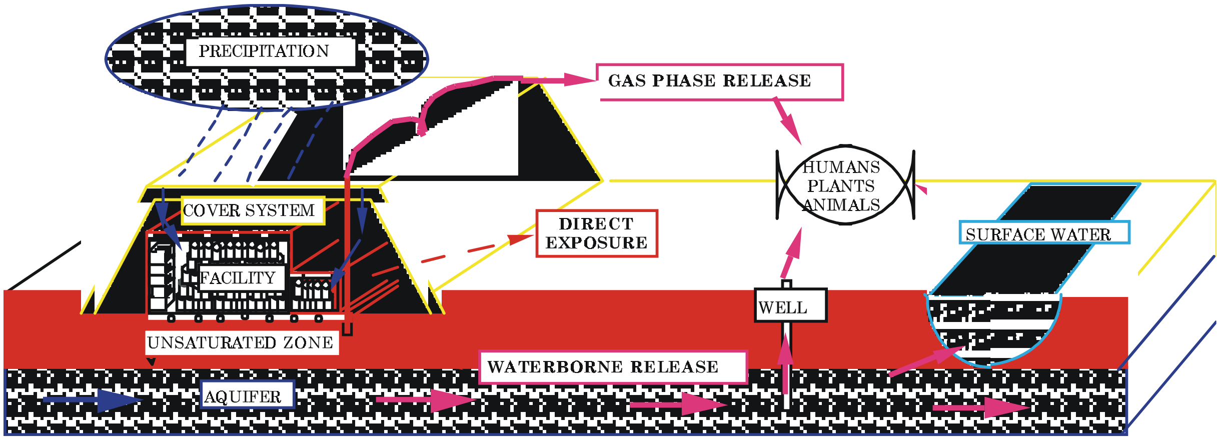

Figure A-1
Depiction of a Hypothetical Facility and the Key Areas for Performance Assessment in LLW

The regulatory requirements on human dose to be complied at any site for disposal of civilian low-level waste have been established by the Nuclear Regulatory Commission[2] as follows:

> *"Concentrations of radioactive material which may be released to the general environment in groundwater, surface water, air, soil, plants or animals must not result in an annual dose exceeding an equivalent of 25 millirems to the whole body, 75 millirems to the thyroid, and 25 millirems to any other organ of any member of the public. Reasonable efforts should be made to maintain releases of radioactivity in effluents to the general environment as low as reasonably achievable."*

States have adopted similar requirements[3]. Demonstration of regulatory compliance at a site is only possible through projections of future conditions based on current knowledge. Therefore predictive mathematical/computer models are relied upon in performance assessment to adequately represent the evolution of physical phenomena that may release, transport and ultimately lead to exposure of humans to radionuclides released from the waste. The models involved in performance assessment are divided into three broad categories for the purposes of this review.

The first category is referred to as near-field models. Near-field models track the degradation of the facility, waste contained therein, and seepage of radionuclides into the surrounding subsoil. The second group of models tracks the movement of radionuclides from the subsoil to potential human exposure sites. These are referred to as transport models. The third group of models track the uptake, exposure and dose equivalent that may result from exposure to any transported radionuclides. These are referred to as exposure/dose models.

Near-field models require a description of facility design, natural site features, aspects of degradation/failure of engineered barriers (soil covers, concrete barriers, waste containers), infiltration of water through engineered barriers, release of radionuclides from various waste forms, and near-field transport.

The transport models include a mathematical description of movement of radionuclides from the facility through the environment via air, surface water, soil, groundwater, and biota[*]. The objective of modeling radionuclide transport is to predict the concentrations at assumed receptor locations.

Exposure/dose models use various scenarios to predict any public exposure to radionuclides released from the site and the resulting human dose. Exposure scenarios combine basic exposure modes - external, inhalation, and ingestion -, demographic data and postulate human activities related to exposure. More detailed discussions on biotic transport, consumption, etc. are presented in later sections.

All the component models are combined to provide an integrated model of the system that is used to project the annual dose to a maximally exposed individual. Regulations require the model projections to project exposure for 1000 years, or more[39]. In the U.S., agreed upon exposure scenarios are assumed to occur, that is the probability of the exposure scenario happening is assumed to be 1. This is different from HLW performance assessment where the probabilities need to be explicitly calculated. In actual performance assessment of a site, linkage between modeling activities, data collection and design through iterative processes is essential to defensibly demonstrate understanding of the overall system performance[1].

This review describes the physical process models of performance assessment and their current status for the application to civilian LLW disposal facilities in the U.S. The following discussions describe each component of a LLW performance assessment.

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[*] Plants, animals, fish, or any other biological species that is related to human exposure pathway.

Review of Performance Assessment Models

A prerequisite for the successful use of performance assessment models is quality input data. These data are comprised of characterizations of emplaced wastes, facility characteristics and materials of construction, and the natural conditions of the selected site as well as data that support the model selections. Characterization of waste determines the inventory of radionuclides in a disposal facility. This issue is further discussed elsewhere[4,5]. Characterization of the site is initially performed to assure compliance with site specific regulatory requirements[2] (10 CFR 61.50). After a site is selected, a more detailed characterization is undertaken for the purpose of obtaining a better understanding of exposure pathways and obtaining data to support a defensible performance assessment. Facility characterization is developed and refined as the facility is designed and built. One benefit of putting together models of the whole system is that "weak-links" in the database can be identified by the models for further refinement. The iterative development of models and data is an important part of reducing the uncertainty in the selection of models and parameter values in performance assessment and ultimately producing technically defensible predictions of the expected behavior of a sited facility. The following sections review the models that are used in performance assessment and a summary of model and data needs is provided. Even though the aspect of data collection is very important, this review focuses on the modeling issues.

Near-Field Models

Modeling Water Infiltration

Infiltration of water into a waste disposal facility and into the waste region is a principal means in inducing the release of radionuclides from a disposal facility. Since infiltrating water flow is dependent on the natural percolation at the site and the performance of engineered barriers, its prediction requires modeling of unsaturated water flow through intact or partially/completely failed components of engineered barriers. The engineered barriers include the cover systems, concrete vault, backfill, waste forms, and overpacks.

The issues to be addressed in these modeling efforts include:

1. What is the infiltrating water flow when the engineered barriers remain intact?;
2. When does each barrier component begin to fail?;
3. What is the rate of change of permeability of each component while failing?, and;
4. When is the barrier considered completely failed and what is the infiltrating water flow when the barrier is completely failed?

Water infiltration models determine the infiltration of water into waste disposal units by following the precipitation of water at a site and its partitioning due to surface runoff, evaporation, and plant uptake/evapotranspiration. Percolation of water into the waste is then determined by describing the flow through the cover system, concrete vault, and other barrier components.

Precise estimation of water flow is extremely complicated due to localized variability in material properties, transient nature of rainfall events, etc. Therefore, simplifications are made in performance assessment with the aim of the obtaining a defensible estimate of water flow.

The vertical steady-state flow of water infiltration in an unsaturated zone is described by the Darcy's law,

$$q = K(\psi)\frac{\partial h}{\partial z} \qquad \textbf{Eq. A-1}$$

where, $K(\Psi)$ represents the hydraulic conductivity in the unsaturated zone as a function of pressure head Ψ. dh/dz represents the hydraulic gradient in the vertical direction. To describe the transient flow, the Darcy equation is combined with the continuity equation to give the Richards equation.

$$\frac{\partial\theta}{\partial_t} = \frac{\partial\theta}{\partial\Psi}\frac{\partial\Psi}{\partial_t} = \frac{\partial}{\partial x}[K(\Psi)\frac{\partial\Psi}{\partial_x}] + \frac{\partial}{\partial_y}[K(\Psi)\frac{\partial\Psi}{\partial_y}] + \frac{\partial}{\partial_z}[K(\Psi)(\frac{\partial\Psi}{\partial_z} + 1)] \qquad \textbf{Eq. A-2}$$

where, θ is moisture content of the soil medium. Various relationships for both hydraulic conductivity and moisture content as functions of pressure head are available to solve the equation. These include, for example, polynomials[6], van Genuchten functions[7], Haverkamp functions[8], and Brooks-Corey functions[9].

There are literally hundreds of computer models available for water infiltration analysis. A few commonly used models in performance assessments include PRESTO[10], HELP[11], UNSAT-H[12], MSTS[13], PORFLOW[14], VAM2D[76], and FEMWATER[79] (see Table A-1).

Even though the variability in precipitation is on a very short time scale (such as hourly or shorter)[15], the infiltration modeling is typically performed on a longer time scale data (such as daily-averaged precipitation rate or much more commonly as historical average over many years). It has been shown that, for the purpose of predicting solute concentrations in a porous media, results from steady-state calculations are in good agreement with those from transient calculations[16,17]. This indicates that use of average bulk water flow is acceptable for performance assessment purposes as long as the correct value of average infiltration rate is used. For the accurate characterization of average infiltration rate, evaporation is found to play a major role[15]. For this purpose, plant growth and water utilization need to be carefully described to accurately predict the flux of water passing below the root zone[15]. As more engineered features such as caps are introduced to the disposal system design, the effects of diverted water run-off in the overall water balance could also be enhanced.

Table A-1
Computer Codes for Modeling Water Infiltration

Code	Features
PRESTO [10,18]	A space independent system equations by using transformation of space-dependent models; Infiltration model includes overland flow, subsurface flow, and atmospheric diffusion systems
HELP [11]	A quasi-2-dimensional empirical moisture routing model
UNSAT-H [12]	1-dimensional finite difference model
MSTS [13]	2-dimensional, 2-phase finite difference model
PORFLOW [14]	2 or 3-dimensional, coupled heat, flow, and mass transport, nodal point integration method

Modeling Flow Through Concrete Vaults

Infiltration predicted by the water infiltration models becomes important only after the failure of a concrete barrier in performance assessment. Before a failure, it is assumed that a concrete vault works as an effective barrier to minimize the contact of water with the waste. The time of failure of a concrete vault is important, along with the time interval between the initiation and completion of the concrete vault failure.

The performance history of modern concrete is short (~100 years) relative to the required time frame of performance assessment, therefore, prediction of concrete service life is difficult[19]. The main concern in predicting the failure of concrete barriers is the formation of cracks which penetrate the barrier thickness. Concrete cracking can be attributed to volume expansion and loss of concrete strength (tensile strength, compressive strength, and modulus of elasticity). Mechanisms leading to cracking include sulfate attack, corrosion of reinforcement steel, freeze-thaw cycling, calcium hydroxide leaching, stress cracking, alkali aggregate reaction, etc. These mechanisms are in turn affected by fluid flow and mass transport through concrete. A detailed review of these processes is beyond the scope of this review and is provided elsewhere in the literature[19]. There is also evidence that self-healing of cracks may occur from carbonation of calcium hydroxide in the cement paste by carbon dioxide[20]. The conditions that would cause autogenous healing to occur are not known. The degree to which the self-healing of cracks can be taken credit for in performance assessment remains an open issue for future research.

Table A-2 lists the computer codes for the analysis of concrete barrier performance that have models for most common degradation modes and have been used in performance assessments. These computer codes include BARRIER[21], 4SIGHT[22], CEMENT[23], and RAESTRICT[24].

Table A-2
Computer Codes for Modeling Concrete Barrier Performance

Code	Features
BARRIER [21]	considers sulfate attack, freeze-thaw cycling, calcium hydroxide leaching, and corrosion of reinforcement steel
4SIGHT [22]	considers sulfate attack and reinforcement steel corrosion
CEMENT [23]	emphasize corrosion of reinforcement steel with capability to consider sulfate attack, freeze-thaw cycling, and alkali aggregate reaction. Use detailed analytical solutions to estimate flow rates in the presence of cracks.
RAESTRICT [24]	Improved version of BARRIER

Cracks could represent a preferential pathway for water and mass flow even though the remaining bulk volume of concrete is intact. However, the role of cracks affecting the flow rate varies greatly depending upon the degree of saturation. In fact, the cracks could serve as barriers to flow under unsaturated conditions[25]. The fact that LLW facilities are located in the unsaturated zone thus complicates the role of cracks in influencing performance of concrete barriers[19]. The conditions at the roof of the vault (i.e., supply of water from above, moisture tension above the vault, the permeability of the vault roof, and the slope of the vault) determines the amount of water flowing through the vault.

Estimation of flow rates in the presence of cracks is modeled by, either numerical solutions of a detailed unsaturated flow code[26] or analytical solutions[27,28]. For the numerical solutions, cracks can be explicitly modeled through proper grid specification[23] or implicitly by modifying the saturated hydraulic conductivity and the moisture release curve to represent crack properties. Calculations have shown that the spacing of fractures is a critical parameter for flow into the vault along with the conditions of the vault roof as described above[23].

In performance assessments, water infiltration through engineered barriers into the waste region is sometimes modeled simply by changing the hydraulic conductivity of the system as a function of the service life of the barriers[29] or by the use of tabular water infiltration data after being estimated by separate calculations[30]. Water infiltration is also modeled by a simple linear relationship between no water flow and natural site infiltration[18].

Modeling Failure of Waste Containers

Waste containers, for some period of time, prevent the contact of groundwater with the waste and the subsequent release of radionuclides from the waste in the event that water comes through the engineered barriers. Waste containers used in burial of LLW include carbon steel drums, liners, and boxes and high integrity containers (HICs). These containers are currently envisioned to be emplaced in a disposal facility with either soil or cement backfills.

The issues that must be addressed by modeling the degradation of containers include:

1. When, and by what mechanism, do the waste containers fail, allowing the contact of infiltrating water with waste?;
2. If the waste containers do not fail all at the same time, how are the failures distributed?, and;
3. If the waste containers are only partially breached, how is the release of radionuclides different from the case of complete failure?

Steel containers can undergo both uniform corrosion and pitting corrosion within the soil systems. A detailed review of data for steel container failure in soil systems is available in the literature[31]. In a cemented system, both types of corrosion can take place but can severely be limited due to depletion of oxygen in the steel-cement interface area[31]. The key process in describing steel corrosion in cement is the description of oxygen and chlorine transport in the system[32]. Modeling and experimental studies of steel corrosion in cementitious anaerobic repository environment are available in the literature.[32,33]. High-integrity containers (HICs) are designed to have a minimum lifetime of 300 years. The type of HIC expected to be used in the future is a combination of both a HDPE and a concrete overpack[90]. HICs eventually are expected to fail by degradation of the concrete casing and creep of the high density polymers.

The distribution of waste container failures is stochastic in nature. However, in current performance assessment, container failures are commonly modeled as a delay time until the onset of release[30, 31, 34]. This assumes that containers have a finite lifetime and at the end of the lifetime all of the containers fail simultaneously (here referred to as “single-time-to-failure (STF)” approach). This STF approach is justified only if the spread of the failure-rate distribution is very narrow.

In a soil backfilled system, the lifetime of carbon steel containers has very little effect on performance assessment because the rate of water infiltration is expected to be very low around the times of container failures. Therefore, there is not much incentive to change the use of STF approach for carbon steel containers in soil systems, if the rate of water infiltration during the early history of a facility is controlled to design objectives. However, if the container failures are taking place when the water infiltration rate is expected to be relatively high, use of the STF approach could result in unrealistic release projections. The DUST[30]-BLT[31] models have container degradation components that simulate container failure over time. The PRESTO[18] model has linear container lifetime functions. Use of the distributed failure (DF) approach is found to have a major impact on the projections of performance assessment calculations when the distribution in failure times is much longer than the mean contaminant residence time in the disposal facility[35, 128]. Adopting a DF approach based on mechanistic understanding of failure for more durable types of waste containers such as HICs needs to be further explored to enhance the technical defensibility of the analysis.

Modeling Radionuclide Release from Various Waste Forms

As currently practiced, LLWs are disposed in a wide variety of materials and forms, including metals, resins, filters, mixed trash, cement, or sorbent media. The waste form into which the radionuclides are incorporated has a considerable impact on the manner and degree to which they are retained in the waste form. Current NRC requirements do not necessarily promote the use of technology to enhance the durability of waste forms resulting in a wide variety of waste forms used in the industry. Current requirements stress the structural but not necessarily the chemical durability of waste forms.

The issues that must be addressed for modeling radionuclide release include:

1. How do we differentiate/describe the release of radionuclides from different LLW forms?;
2. How do we characterize the local geochemistry of the system to describe radionuclide release?;
3. What are the effects of the degree of water saturation and use of different backfill materials on the release of radionuclides from different waste forms?, and;
4. If the radionuclide is volatile or can be released in gas, how should this be addressed?

Mechanisms of release of radionuclides vary between different waste forms and include rinse release, diffusion, dissolution, corrosion, or ion-exchange. Defining the inventory of radionuclides available for each different mechanism is important along with modeling of these mechanisms. Due to the heterogeneity of waste forms in LLW, the release of radionuclides is commonly predicted by a rinse release model. The most conservative rinse release model assumes the entire inventory is “rinsed” off the wastes upon contact with water. Rinse release models are known to be overly conservative for some waste forms.

Existing models for predicting radionuclide release (Table A-3) can be categorized as either K_d based rinse models or mechanistic models. The K_d based rinse models assume that the release is surface wash controlled with water subject to equilibrium partitioning between the waste form and the contacting solution and that this is true for all types of waste forms modeled in LLW streams. Computer models that use this approach include IMPACTS[36], PRESTO[18], and GWSCREEN[37]. Mechanistic models attempt to describe the release of radionuclides from various waste forms based on mechanistic description of the release from specific types ofwaste forms. Typically, mechanistic models have diffusion and/or dissolution release models in addition to a K_d-based partitioning model. These include DUST[30], BLT[31], PAGAN[34]. Equations used to describe the source-term in these computer models are detailed below:

(IMPACTS model)

$$release rate = C_0 V_{water} f_L \frac{1}{K_d \rho_{waste}} t_{contact} \qquad \textbf{Eq. A-3}$$

where, C_0 is the radionuclide concentration of the waste,

V_{water} is the annual volume of water that percolates through the disposal facility cover and contacts the disposed waste,

f_L is the leachability/accessibility factor,

ρ_{waste} is the density of waste, and

$t_{contact}$ is the contact time fraction representing fractional saturation for unsaturated flow.

(PRESTO model)

$$release\,rate = \frac{M}{(1 + \frac{K_d \rho_{waste}}{\varepsilon})\, h_{waste} K} \frac{p^2}{\varepsilon} \qquad \textbf{Eq. A-4}$$

where, M is the mass (activity) of waste,
K_d is the distribution coefficient of radionuclide,
h_{waste} is the height of waste region,
ε is the porosity of the waste,
K is the saturated hydraulic conductivity of the waste trench, and
p is the annual percolation flux.

(GWCREEN model)

$$release\,rate = \frac{M}{(1 + \frac{K_d \rho_{waste}}{\theta})\, h_{waste}} \frac{p}{\theta} \qquad \textbf{Eq. A-5}$$

where, θ is the moisture content of the disposal unit.

(DUST rinse model)

$$release\,rate = \frac{M_{rinse}}{(1 + \frac{K_d \rho_{waste}}{\theta}) \cdot time\,step} \qquad \textbf{Eq. A-6}$$

where, M_{rinse} is the mass available for rinse release.

(Diffusion model in DUST/BLT)

$$release\,rate = \int dS \cdot J_S \qquad \textbf{Eq. A-7}$$

where, J_s is the mass flux at the surface ($J_S = -D \frac{\partial C(x_s)}{\partial x}$) and is obtained through the use of analytical solution of diffusion equation for spherical, cylindrical or rectangular geometry assuming zero concentration at the boundary. x_s denotes a surface of the waste form and dS is the element of the surface area.

(Dissolution model in DUST/BLT)

$$release\,rate = \frac{v_{diss} M_{diss} SA_{diss}}{V_{diss}}$$ **Eq. A-8**

where M_{diss} is the mass available for dissolution. v_{diss} is the dissolution velocity, SA_{diss} is the surface area of the waste form for dissolution release, and V_{diss} is the volume of waste for dissolution release.

Table A-3
Computer Codes for Modeling Radionuclide Release from LLW

Code	Features
PAGAN [34]	Leach rate determined by diffusive release or surface rinse based on Kd, solubility limit considered, coupled with near-field transport (mixing cell cascade model)
NEFTRAN [41]	Leach rate or solubility limit based release, leach rate to be specified by the user, leaching model is coupled with a near-field transport model
IMPACTS [36]	Surface rinse release based on K_d, no near-field transport
PRESTO [18]	Surface rinse release based on K_d, solubility limit considered, no near-field transport
PATHRAE [59]	Leach rate (user-provided) based release, treats area source
GWSCREEN [37]	Surface rinse release based on K_d, no near-field transport
DUST [30]	Diffusive release, rinse release, uniform dissolution release, solubility limit considered, 1-D finite difference model for near-field (advection and dispersion) transport that couples all release processes and feedback effects
BLT [31]	Diffusive release, rinse release, uniform dissolution release, solubility limit considered, 2-D finite element model for near-field transport that couples all release processes and feedback effects (based on FEMWASTE)
DUST-MS [43]	Decay chains can be described in addition to the existing DUST capability
BLT-MS [44]	Decay chains can be described in addition to the existing BLT capability
BLT-EC [42]	In addition to BLT, sequential and branched decay considered, geochemical modeling for release and transport of radionuclides based on HYDROGEOCHEM and MINTEQA2
BARRIER [21]	Includes diffusive release model, but primarily for the prediction of concrete barrier performance (same is also true with RAESTRICT [24])

In PAGAN, a K_d based rinse model for unstabilized waste and a diffusional release model for stabilized waste are employed. The diffusional release is obtained using an analytical solution that assumes only-radial diffusion in a cylinder with a zero- concentration boundary condition.

Among these models, IMPACTS[36], in particular, represents the most commonly used LLW source term model. IMPACTS was developed by U. S. NRC to support the 10CFR Part 61 compliance activities[38] and has been adopted in many LLW disposal facility licensing activities by compacts and states. Radionuclide release from various waste forms are explained in IMPACTS by using various default parameters. And the model has a built-in library for these parameters on which the accuracy of the model prediction is dependent. The partition factors (K_d) used in this library are based on measurements made at Maxey Flats, KY and West Valley, NY that represent near neutral pH and strongly anoxic conditions, where leachates are supersaturated with respect to carbonate and sulfate phases[31]. These values for K_d's are generally higher than those used in more recent performance assessment and are no longer acceptable by the NRC for present use[40]. Post-leaching dilution of radionuclides within the disposal unit is not described in the model.

Cemented waste is an important waste stream from an international perspective due to the widespread use of cement solidification technology. However, in the US, only a small fraction of LLW is currently treated by cementation. This is partly due to the concern for volume increase after solidification and the current NRC regulatory position[45] on LLW forms. The NRC position stresses the structural stability of the waste form, but does not necessarily promote the use of technology to enhance its chemical durability. For the release of radionuclides from cemented waste, bulk diffusion is known as the primary rate-controlling mechanism except for the early time periods when initial washing-off effects are taking place[46]. For the description of diffusion-controlled release, most of the existing models assume zero-concentration at the waste form surface to facilitate the derivation of analytical solutions. This boundary condition leads to release rates that are independent of the surrounding conditions and are conservative. Different analytical solution approaches[47,48] or numerical solution-based models[30,31,49] are available based on continuity of concentration and flux at the surface boundary when effects of surrounding conditions on diffusive release are considered important.

In the current U.S. LLW management practices, dewatered unsolidified ion exchange resins contain a large fraction of activity in LLW[5]. Release of radionuclides from ion exchange resins is commonly modeled by a surface rinse release model due to lack of supporting experimental data. The NRC's Branch Technical Position (BTP) on LLW performance assessment[39] suggests the use of a K_d based partitioning model as an alternative. In either of these cases, the predicted release is dependent upon the value of K_d used in the calculation. The K_d value to be used should be based upon the geochemistry of the waste form environment and is expected to be different from the values used in describing the movement in subsurface environment. The distribution of activity between the solid phase and liquid phase is dependent on whether the contamination is more surface- or ion-exchange-controlled in ion exchange resin matrix. If most of the activity in ion exchange resins is not from surface contamination, radionuclide release from ion exchange resins is expected to be more ion-exchange controlled. A recent work based on a mechanistic modeling approach[50] showed that key processes and parameters in describing radionuclide release from ion exchange resins include diffusion within the bulk pore water phase, flow rate of infiltrating leachant water, concentration of the counter ions in the leachant water, and sorption on resins during the transport in the bulk pore water phase.

Geochemistry controls solubility limits of radionuclides in pore water, sorption/retardation during release, or any chemical degradation/transformation of radionuclides within the waste. The determination of parameters in these processes (i.e., K_d* or solubility limit) is dependent upon the evolution of the geochemistry and the conditions present near the waste when contacted by ground water. A standard approach of treating geochemical uncertainty in predicting these parameters is the use of fixed, conservative values. However, this fixed value may be too conservative (or non-conservative, if not properly selected) in some cases. Separate calculations can be performed with chemical reaction codes for detailed characterization of local geochemistry. A coupled radionuclide release, transport and chemical reaction code[42] is available and should be useful for detailed source term analysis. But wide use of this coupled model is not likely in a typical performance assessment. These codes have the potential to be more accurate and detailed, but, understandably, require extensive input data and are more complex calculations that may require a significant validation effort.

Radionuclide release from solidified wastes is assumed to be independent of the degree of water saturation in the waste[51]. In the case of unsolidified waste, limited data[52,53] show that the degree of water saturation has an influence on the release of radionuclides. This point needs further investigation.

Use of backfills in LLW disposal units affects the flow of water into the waste region and the boundary conditions for the release. This effect becomes much more significant if cement backfills are used. Cement may either prohibit or selectively channel the water flow into the vicinity of the waste. For an extended period of time[58], cement will also generate highly alkaline environments and affects the oxidation potential of the system. Solubility and sorption of radionuclides, corrosion of metallic waste, leachate flow, and gas generation are all influenced by the presence of cement for some period of time. These effects are expected to play an important role in source term analysis with the extensive use of concrete vaults and canisters in the U.S. A detailed analysis of the effects of cement backfills are not explicitly included in all of the computer models listed in Table A-3 except for BLT-EC[42].

Gas generation due to waste form degradation is another mechanism that impacts the release of ^{14}C, ^{3}H, and other volatile nuclides in LLW[54]. Once gases are generated, they migrate to void spaces in waste containers and can be released by the diffusion through the cover system or by the pumping effect of atmospheric pressure changes. In general effects of gas generation are not of significance in the integrity and performance of the facility. However, it has been found that the effects of gas generation on the ^{14}C source term are not negligible[35]. Since none of the models reviewed in Table A-3 address the effects of gas generation, separate analyses are needed to address these effects in performance assessment. Partitioning of radionuclides between gas and liquid phases and its impacts on the inventory available for release to either groundwater or air can be addressed by using mechanistic approaches[55] for individual LLW streams.

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[*] K_d represents the distribution coefficient of the contaminant under equilibrium as the ratio of concentration in the solid phase to the concentration in the liquid phase.

Modeling Near-Field Transport

For radionuclides to reach the bottom boundary of a disposal unit after being released from waste packages, they need to migrate through the remainder of the facility. This process may involve spreading and dilution of radionuclides released from the waste containers. The result of the near-field transport calculations is referred to as the source term[*]. Near-field transport of radionuclides is governed by advection, dispersion, and sorption characteristics of the material surrounding the waste (referred to as the backfill). Near-field transport models are mathematically identical to transport of radionuclides in an unsaturated soil.

The main issues that are addressed by near-field transport models are:

1. What is the radionuclide release rate?;
2. How is near-field chemistry characterized?;
3. How much dilution of plume takes place?, and;
4. Are there geometric considerations such as fracture flow that need to be described?

In some cases, near-field transport is ignored in performance assessment to simplify the analyses. Sometimes, near-field transport is simplified by assuming the transport to be vertically downward and controlled by advection ignoring diffusional transport to avoid a complex numerical solution (referred to as the mixing cell cascade model[30,56]). In this case, the concentration leaving the i_{th} mixing cell, C_i, is determined by the mass balance on the cell, Equation A-9,

$$\frac{dC_i}{dt} = -\frac{v_d}{\theta Rd}\left(C_i - C_{i-1}\right) - \lambda C_i + \frac{Q_i}{\theta RdA_f} \qquad \textbf{Eq. A-9}$$

where, v_d is the Darcy velocity,

R is the retardation factor as $(1+\frac{K_d\rho_b}{\theta})$,

ρ_b is the bulk density of soil, d is the depth of one cell,

A_f is the area of the facility, and

Q_i is the total release rate from the wasteform.

Space-time dependent numerical models, that are the most general treatment, are used in some models to address near-field transport[30,31]. The following one-dimensional equation is commonly used assuming predominantly vertical contaminant movement (i.e., neglect the horizontal dispersion and advection terms of the equation).

$$\frac{\partial C(x,t)}{\partial t} = -\frac{v_d}{R\theta}\frac{\partial C(x,t)}{\partial x} + \frac{D}{R}\frac{\partial^2 C(x,t)}{\partial x^2} - \lambda C + \frac{Q(x,t)}{R\theta} \qquad \textbf{Eq. A-10}$$

where, D is the coefficient of hydrodynamic dispersion.

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[*] Release rate of radionuclides at the bottom boundary of a disposal unit.

An area source, rather than a point source, can be treated by using a superposition principle or explicitly with the use of computer models[41,59] that treat the spatial (horizontal) extent of the source term.

A major challenge in practical application of near-field modeling is obtaining adequate site-specific data on sorption and solubility in the facility[57]. These data depend on the final facility design and emplacement approach employed.

If concrete backfills are adopted, the geochemical properties of pore water in the system buffered by reactions with cement become very different from those waters associated with the local soil and geology. And the solubility limits and distribution coefficients for dissolved radionuclide concentrations in the cementitious system may work favorably for reduction of radionuclide release or migration. Detailed review of these issues along with the characterization of solubility limits and distribution coefficients of selected radionuclides in a cementitious near-field system is provided by a recent work by Krupka and Serne[58]. Generally, concrete is expected to be a factor in the chemical environment of a facility for a long enough period to influence the release of most of the LLW radionuclides except the very longest lived. Concrete is also expected to remain an effective chemical barrier even with significant development of cracks. However, development of cracks in concrete alters the near-field water flow into a fracture flow. The influence of fractured concrete on radionuclide release can be described by using contaminant transport models in fractured porous media[60,61]. Application of these models to LLW facility performance assessment is discussed in the literature[62,23].

Transport Models

Modeling Groundwater Flow

The first task in the analysis of radionuclide transport in groundwater is understanding the groundwater flow system, at the appropriate scale, on the basis of existing hydrogeologic data and interpretations. Groundwater flow rates must be estimated to calculate radionuclide transport. This is often done with separate groundwater flow models. And the results become inputs to contaminant transport models.

The first geologic or natural barrier that would be encountered by radionuclides after release from a disposal facility is termed the unsaturated zone. The unsaturated zone is the region below the facility, but above the local water table. Location above the local water table is required for LLW facilities by 10CFR61[2*] . The unsaturated zone is characterized by a porous system where pore spaces are not fully saturated with water. Due to capillary effects in pores, water pressure in the unsaturated zone is less than the atmospheric pressure. Due to the effects of voids in hindering water movement, hydraulic conductivity in the unsaturated zone is reduced from the fully saturated conditions. Therefore, pore size distribution and moisture content/distribution plays an important role in controlling vertical movement of water and contaminants in an unsaturated zone. The unsaturated zone provides a linkage between atmospheric moisture, groundwater, and seepage of groundwater to streams, lakes, or other surface water bodies.

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[*] 10 CFR 61 allows location of the facility in the water table if diffusion is the dominant transport mechanism.

In an unsaturated zone, obtaining solutions of the flow equation is complicated because moisture content and hydraulic conductivity are nonlinear functions of capillary pressure. This has been discussed earlier in the section for modeling water infiltration into the waste. If the geology near the facility is simple with nearly uniform constant recharge, steady-state vertical flow can be assumed[57]. Often the flow and transport through the unsaturated zone is modeled by a simple delay time and no sorption or dispersion is assumed.

If groundwater pore spaces are fully saturated, the hydrogeologic system is termed a saturated zone. In the saturated zone, water pressure is equivalent to the atmospheric pressure plus any hydraulic head and the transport is mainly a horizontal movement of water and contaminant through the main body of the soil. Depending upon the length of travel and the sorption characteristics, saturated zone transport may provide a significant barrier to radionuclide release to the biosphere through dilution and long travel times that allow for radionuclide decay.

The governing equations for groundwater and contaminant transport models in both the unsaturated and saturated zone are well established. These include the flow equation (result of combination of Darcy's law and the equation of continuity) and the advection-dispersion equation. The flow equation is similar to Equation (A-2) and is represented in a more general form as

$$\nabla \bullet K \nabla h = S_s \frac{\partial h}{\partial t} \qquad \textbf{Eq. A-11}$$

where, S_s is the specific storage which indicates the available volume of water removed per unit volume of rock when the hydraulic head is lowered by unit amount, and h is the hydraulic head. K is the hydraulic conductivity. The contaminant transport equation is the same as Equation (A-10) except that the effective porosity in the saturated system replaces the moisture content in the equation.

In a saturated zone, the mathematical solution techniques are well established. The details of modeling requirements depend on the distance to the receptor location, desired degree of accuracy for the calculation, and hydrogeological characteristics of the site[57]. For example, a simple hydrogeological system such as a confined homogeneous aquifer may be modeled as one-dimensional. If the system is highly inhomogeneous and anisotropic, multidimensional models may be necessary. If transient recharge rates and pumping wells are considered, time dependency needs to be included in the modeling (although this can generally be avoided by appropriate averaging).

Modeling Transport of Radionuclides in Groundwater

Various guidelines or protocols[65-73] are available to help facilitate the use of models for the analysis of groundwater transport in various field situations. The development of a conceptual models for a given problem is one of the most important steps in these modeling exercises. During the development of a conceptual model, preliminary calculations using a simple constitutive relations or analytical solutions can be very useful to identify the probable nature of the dominant processes. For this, review and compilation of all relevant information at both local and regional scales are necessary.

Three of the most common analysis techniques used to analyze contaminant transport in groundwater are[62,63]: (1) numerical solutions of the contaminant transport equation; (2) analytical solutions of the contaminant transport equation, and; (3) the stream-tube approach[41,64]. If the flow velocities are assumed to be approximately constant in space and time, an analytical solution approach or the stream tube approach can be used to estimate the radionuclide transport. For unsteady-state flow or strongly spatially variable flow, numerical solution methods are necessary[57]. Use of complicated flow and transport analyses should only be undertaken if the characteristics of the site merit the extra detail[50].

For some of the actinides, describing decay chains during transport is also important in this modeling and the simplified flow models allow for certain cases to be solved with analytical models. Table A-4 shows computer models that have been developed for performance assessment purposes for the modeling of transport in groundwater. It should be also noted that there are many other computer codes applicable for groundwater transport modeling.

For LLW performance assessment, the stream tube approach was used in the NRC BTP on LLW performance assessment[39]. A stream-tube approach[41,64] assumes that all significant flow and radionuclide transport takes place along pathways that can be simulated as discrete one-dimensional paths. Transport of contaminants in the paths are described by moving groups or packets of particles along the flow field over each time step allowing the packets to spread simultaneously with advective transport. Caution however should be exercised with the use of steam tube models. Calculation of a concentration with a stream-tube approach requires the introduction of a dilution volume, which must be specified by assigning an effective cross-sectional area through which transport occurs[62,63]. To prevent arbitrary selection of this cross-sectional area, a detailed three-dimensional flow model may be necessary to identify the boundaries of pathlines that can be used in assigning the stream tube volume. In general, stream-tube models are better for analyzing radionuclide fluxes than prediction of groundwater concentrations.

One issue which typically poses difficulty in groundwater transport modeling is the description of a fractured system. If the groundwater system is fractured, there exists a preferred pathway for the transport of radionuclides which could accelerate the transport over that found in nonfractured porous medium. Modeling of the fractured system could be based on either an equilibrium continuum approach or discrete fracture modeling[74].

The equilibrium continuum approach assumes that the fractured mass is hydraulically equivalent to a porous medium in which spatially defined values of hydraulic conductivity, porosity, and compressibility can be assigned. This approach is valid as long as the fracture spacing is sufficiently dense that the fractured media acts in a hydraulically similar manner to granular porous media[74]. For the purpose of predicting the spatial distribution of concentration of radionuclides, the equilibrium continuum approach is well accepted[92]. When the fracture density is extremely low and the flow is described in relation to individual fractures or fracture sets, discrete fracture modeling is needed. Characterization of fractures in a system poses a major technical challenge for both the field work and the modeling activities.

Table A-4
Computer Codes Applicable to Modeling Radionuclide Transport in Groundwater in Performance Assessment

Code	Features
NEFTRAN [41]	1-D analytical stream tube approach in saturated zone, steady-state flow, decay chains allowed.
TRNSS [75]	1-D analytical stream tube approach in unsaturated and saturated zone, no decay chain allowed
VAM2D [76]	2-D finite element code for transient or steady state processes in both saturated and unsaturated zones, describe decay chains
VS2DT [77,78]	2-D finite element code for transient or steady state processes in both saturated and unsaturated zones, no decay chain allowed
FEMWATER [79]/ FEMWASTE [80]	2-D finite element code for either transient or steady state processes in both saturated and unsaturated zones, no description of decay chains
MODFLOW [81]/ MODPATH [82]	3 -D finite difference code for saturated zone
MASCOT [83]	analytical solutions for 1, 2, 3-D transport, steady state flow, 4 member decay chain, homogeneous unsaturated and saturated zones
SUTRA [84]	2-D finite element/finite difference code, decay chain modeled, anisotropic unsaturated and saturated zone
TRACR3D [85]	1, 2, 3 D finite difference code for steady state or transient flow and mass transport in heterogeneous porous and fractured media, two-phase (gas and liquid) modeling
MAGNUM2D [86]/ CHAINT [87]	2 or 3 D finite element code for flow and mass transport in a saturated, fractured porous media, transient or steady-state transport in heterogeneous anisotropic system, chain decay allowed
FRACMAN [88]	3 D discrete fracture analysis for flow and mass transport, not available to general public
TRIPM [89]	1, 2 D finite element code, for either transient or steady-state processes in both saturated and unsaturated zones, up to 3 member decay chains
PORFLO [14]	2 or 3D coupled heat, flow, and mass transport in a saturated or unsaturated porous media, equilibrium continuum model, nodal point integration method
GRDFLX [91]	Analytical solution approach to the advection-dispersion (1D advection, 3D dispersion) equation for saturated porous medium based on Green's function method, NRC Codell model
PAGAN [34]	Saturated zone transport is based on the same mathematical model of GRDFLX, with numerical integration Green's function integral, unsaturated zone is assumed to act only as a delay mechanism
GWSCREEN [37]	Saturated zone transport is based on GRDFLX, simple plug flow model for the unsaturated zone transport
PRESTO [18]	1 D analytic plug flow model with a parametric correction for dispersion in the aquifer using a plume-dispersion angle
PATHRAE [59]	1 D analytic plug flow model with a parametric correction for dispersion both in the unsaturated zone and the aquifer

Modeling Atmospheric Transport

Atmospheric and surface water transport are usually less important than the groundwater transport in performance assessment due to large dilution capacity of the medium during the transport and the secondary nature of contamination. However, circumstances arise where atmospheric and/or surface water transport are important. These typically take place after the occurrence of an event that disrupts the integrity of the containment structure and/or exposes waste at the surface[93]. Events that disrupt the integrity of the containment structure and/or soil cover could be naturally occurring (e.g., wind and water erosion, earthquakes and landslides, and containment structure subsidence or collapse) or events resulting from the activities of an intruder. Events resulting from intruder activities are not required to be addressed for civilian LLW disposal sites according to the 10CFR61[2].

Potential sources of airborne contamination are: (1) Contaminated soils, and (2) Radioactive gases. When soils are contaminated, there could be resuspension of particulates allowing entrainment of radionuclides in air. Radioactive gases can be generated within a disposal facility and released to the atmosphere.

Atmospheric transport of these particulates or gases to receptor locations is usually calculated using a Gaussian dispersion model which works well to describe average plume concentration behavior for a relatively short travel distance (less than 80 km). The Gaussian dispersion model is used for most performance assessment calculations. The model assumes a continuous release of point source material and is represented as,

$$\chi(x,y,z) = \frac{Q}{2\pi\sigma_y\sigma_z u} e^{-\frac{y^2}{2\sigma_y^2}} \{e^{[-\frac{(z-h)^2}{2\sigma_z^2}]} + e^{[-\frac{(z+h)^2}{2\sigma_z^2}]}\} \qquad \textbf{Eq. A-12}$$

where, $\chi(x,y,z)$ represents the concentration at a point (x,y,z) downwind, for a plume propagating in the x-direction,

Q is the constant emission rate,
u is the wind speed,
σ_y, σ_z are the standard deviations of the plume dispersion in the transverse and vertical directions, and
h is the effective elevation of the release point above the ground.

Variations of the model can be used to describe the deposition or depletion of the plume[94]. For the calculation of annual average concentrations of contaminants, which are practically more relevant in performance assessment, the sector-average representation of Equation (A-12) is used[95]. Various computer models developed for the atmospheric transport analysis based on the Gaussian dispersion model are shown in Table A-5.

Table A-5
Computer Codes for Modeling Atmospheric Transport in Performance Assessment (All use Gaussian dispersion model)

Code	Features
AIRDOS-EPA [96]	Both point and area source, both wet and dry deposition
XOQDOQ [97]	Point source release, only dry deposition
DWNWND [98]	Point source release, only dry deposition
CAP88 [99]	Uses modified version of AIRDOS-EPA
PRESTO [18]	Uses DWNWND as subroutine
GENII [100]	Point source release, only dry deposition

Modeling Surface Water Transport

Surface water can be contaminated with radionuclides from a disposal facility through groundwater discharge, surface runoff, or deposition of contamination from airborne transport. Modeling approaches vary among different types of surface water bodies depending upon the geometric and dispersive characteristics of the system. Compartment (mixing cell) models are commonly used when the transport is dominated by turbulent mixing and diffusion[57]. This can be represented by the following equation.

$$\frac{dC(t)}{dt} = \frac{Q(t)}{V} - (\lambda + \frac{q_0}{V})C(t) \qquad \textbf{Eq. A-13}$$

where, $C(t)$ is the concentration in the compartment,

$Q(t)$ is the rate at which radionuclides enter the compartment,

V is the volume of water in the compartment,

q_0 is the rate of discharge from the compartment, and

λ is the radioactive decay constant.

The advection-dispersion equation (similar to equation (A-10)) is used when the compartment model is not applicable, i.e., complete mixing is not applicable. Various analytical solutions[95,101] can be used to solve the advection-dispersion equation except in very complex systems such as estuaries for which an elaborate multi-dimensional computer models can be used. Use of complex models is usually discouraged in performance assessment due to difficulty in obtaining the required data. The significance of surface water in performance assessment will depend on the extent of erosion, overland transport of waste, and recharge from contaminated groundwater that are relevant to a particular site[34]. Table A-6 lists computer models for the modeling of radionuclide transport in surface water.

Table A-6
Computer Codes for Modeling Surface Water Transport in Performance Assessment

Code	Features
NRC Codell model[91]	Analytical solution models for 2 D dispersion in nontidal rivers or near-shore lake or simple complete mixing model
LADTAP II [102]	finite difference solution based on the same mathematical models of NRC models
GENII [100]	Based on selected NRC models for nontidal river, near-shore lake or complete mixing model
PRESTO [18]	Only runoff from waste trench to the stream is described. Complete mixing is assumed in the stream, no transport equation used

Modeling Biotic Transport

Radionuclides present in groundwater, surface water, or the air at/near the receptor location can become part of human food-chains through uptake by edible plants, consumption by food animals, or direct consumption by humans. Biotic transport models consider various interactions between the environment, plants, animals, and humans through the food-chain and provide an estimation of the radionuclide intake by humans through different pathways. The models are well established and have been widely used in various radiological assessment applications.

Biotic transfer models and associated parameters including various transfer factors (soil-to-plant, plant-to-animal), bioaccumulation factors, and rate of consumption by animals and humans are compiled in various work[95,103-105]. A large degree of uncertainty is associated with the models depending on deriving site-, species-, or radionuclide-specific parameters in the model.

An additional issue to be noted in the calculation of dose from radionuclides is the special treatment of ^{14}C and ^{3}H. In general, it is assumed that ^{3}H is transferred in environmental media through its association with water as tritiated water (HTO). Carbon-14 follows the conversion of CO_2 and becomes fixed in vegetation and reaches humans primarily through the ingestion pathway[105]. Special models such as the specific activity model[101] can be employed for airborne release and transport of these radionuclides. The specific activity model assumes that the concentrations of nuclide in environmental media (soil, plants, and animal products) have the same specific activity as the contaminating medium. This approach however is not appropriate for groundwater sources or other ground sources of ^{14}C. For ground or groundwater sources of ^{14}C, the concentration in the soil and subsequent uptake by plants or other biota should be explicitly calculated with the use of appropriate transfer factors.

Table A-7 shows the computer codes available for the modeling of biotic transport in performance assessment.

Table A-7
Computer Codes for Modeling Biotic Transport and Exposure/Dose Analysis in Performance Assessment

Code	Features
GENII [100]	Consider both acute and chronic releases. Exposure pathways include inhalation, ingestion, direct exposure via water, soil, and air.
GENII-S [106]	Both statistical and deterministic calculations of dose, based on GENII with improved user interface
PRESTO [18]	Consider mean concentrations in air, stream water, and well water on annual basis. Exposure pathways include inhalation, ingestion, and direct exposure from plume immersion, soil, and trench.
PAGAN [34]	Estimates dose only from ingestion pathway of drinking water (well water and surface water)
MEPAS [107]	Risk computational tool from the release of chemical and radioactive contaminants into the environment and their migration and fate in the groundwater, surface water, overland, and atmospheric pathways.

Exposure/Dose Models

Modeling Human Exposure/Dose

Calculating human doses from exposure to radionuclides through different exposure pathways is the last piece of performance assessment modeling. Among different exposure pathways, ingestion of radionuclides is usually the most important followed by the inhalation pathway. In most cases, direct external exposure can be ignored[*].

Once the intake of radionuclides into human body is estimated, these intakes are translated into dose through dosimetry models. Inhaled or ingested radionuclides generally become distributed throughout many organs and tissues of the body before they are removed through either radioactive decay or biological elimination. Such internally distributed radionuclides lead to radiation doses to the organs or tissues in which they remain or to other parts of the body with the emission of penetrating radiation. The primary quantity of interest is the radiation energy deposited in various organs and tissues of the body by the ionizing radiation emitted from the radionuclides. The dose to the organ or tissue is the deposited energy per mass, averaged over the organ or tissue of interest.

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[*] Direct exposure becomes important for the inadvertent intruder scenario. The inadvertent intruder scenario is treated separately from performance assessment by NRC. DOE and some states consider the intruder scenario as part of the performance assessment.

Application of internal dosimetry models for various radionuclides has resulted in several set of dose conversion factors. Applying these dose conversion factors to the rate of radionuclide intake represents the dose modeling in performance assessment. The basic equation for dose calculation is[103],

$$R_{aipj} = C_{ip} U_{ap} D_{aipj} \qquad \textbf{Eq. A-14}$$

where, R_{aipj} is the annual dose to organ j of an individual of age group a from radionuclide i via pathway p (Sv/yr or mrem/yr),

C_{ip} is the concentration of radionuclide i in the media of pathway p, [coming from the prior pieces of the performance assessment]

U_{ap} is the usage factor (intake rate or exposure time) associated with pathway p for age group a, and

D_{aipj} is the dose conversion factor, specific to age group a, radionuclide i, pathway p, and organ j representing the dose due to unit intake of a radionuclide.

Dose conversion factors can be different depending on the dosimetry model adopted in the derivation. Current standard dose conversion factors are based on the ICRP 30[108] methodology. A new set of dose conversion factors based on the ICRP 60[109] methodology are available, but have not been widely used, yet. One example of the difference between the ICRP 30 and 60 is tissue weighting factors used in calculating effective dose. The major issue in using dose conversion factors is the uncertainties of parameters used in the internal dosimetry model. A formal parameter uncertainty analysis has not been performed to evaluate the uncertainty in existing set of dose conversion factors with few exceptions[110].

Systems Approach

NRC's Branch Technical Position (BTP) on performance assessment[39] suggests the use of a probabilistic approach for uncertainty analysis of combined performance of a number of disposal facility attributes. By integrating and automating subsystem model (or code inputs and outputs) with an overall "system" code, the ability to step through successive iterations of a performance assessment process and perform uncertainty and sensitivity analyses is significantly enhanced. Table A-8 shows the list of available systems computer models for LLW performance assessment. Stochastic driver codes such as @RISK[111] have been also linked to codes such as PATHRAE[59]and PRESTO[18] to obtain probabilistic analysis.

Table A-8
Systems Codes for Performance Assessment

Code	Features
PRESTO [18]	EPA's tool for rule making analysis, not adequate for site-specific analysis
PATHRAE [59]	Primarily for BRC waste disposed in a sanitary landfill or municipal dump and by incineration methods
PAGAN [34]	Screening tool, estimates dose from drinking water ingestion only
RIP [112]	Originally developed for DOE's Yucca Mountain project, probabilistic modeling capability, could be applied to LLW performance assessment. Careful input selection is required especially for the source term analysis. The code is not in public domain.
NRC Methodology [113]	Combination of DUST, NEFTRAN, and other component models for NRC's test case analysis, not publicly available

Uncertainty Analysis

Performance assessment relies upon predictive mathematical/computer models to project the performance of a site for long periods of time. Due to the necessarily limited nature of the available data and models and the need to extrapolate the known database in time, uncertainties naturally arise. If the model and parameter assumption cannot be shown to provide a prudently conservative upper bound, quantification of uncertainty becomes an essential part of performance assessment. Characterization of uncertainty is also necessary for meaningful decision making, i.e., regulatory compliance determination.

With respect to the treatment of uncertainty in LLW performance assessment, U.S. NRC emphasizes[39] a good general understanding of how the facility will perform by conducting a systematic evaluation to identify important facility attributes and their relationship to performance. If a deterministic bounding analysis is to be used to address uncertainty, a reasonable assurance that the estimate conservatively bounds performance needs to be demonstrated. However, simple bounding analyses may not always be an appropriate measure because of the need to rely on conservatism in the analysis as a replacement for quantitatively evaluating uncertainty. Developing more sophisticated models and more realistic parameter ranges that more explicitly quantify the uncertainty in performance estimates is preferable if these lead into a more defensible decision.

Uncertainties in performance assessment[39] include model[*] uncertainty and parameter uncertainty[114]. Model uncertainty refers to the uncertainty regarding abstracting a real system and its evolution into a form that can be mathematically modeled. Model uncertainty results from limitations in the models used to represent complex system behavior, including the system's evolution (future site conditions, processes, and events), for a specific site and engineering design. This includes uncertainty about the interpretation and use of data and assumptions about heterogeneity, system dimensionality, isotropy, and initial boundary conditions[15].

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[*] The model is the conceptual approach or physical phenomena assumed or known to be important to performance assessment, and the mathematical representations of the concepts or phenomena.

Parameter uncertainty refers to the uncertainty in the data, parameters, and coefficients used in the models. Parameter uncertainty is attributed to a number of sources, including: uncertainty associated with laboratory and field measurements; uncertainty in determining parameter and coefficient values used in a model; and uncertainty associated with the intrinsic heterogeneity of natural systems.

Uncertainty analysis of performance assessment has typically been limited to input parameter uncertainties because parameter uncertainty is more readily quantified. Approaches dealing with parameter uncertainty include analytical methods, Monte Carlo methods, response surface methods, and differential methods[115,116].

Analytical methods are based on statistical treatment of error propagation (e.g., Gauss law) and are applicable when the model has a simple structure and the data have known and well-behaved distributional characteristics. Monte-Carlo methods involve sampling values from each input parameter probability distribution and propagating these values through the model. An output distribution is produced by evaluating the output parameter of interest many times based on random or Latin Hypercube sampling. These methods are most useful when statistical data are available to estimate input parameter probability distributions. Response surface methods are based on using experimental design to select values and pairing of input parameters for the execution of the model and the results are used through regression analysis to construct a response surface (correlation). Once the response surface is constructed, various statistical techniques can be employed to quantify the uncertainty in the model. Differential methods require that partial derivatives be estimated for each output parameter with respect to each input parameter of interest. Adjoint analysis[117] is found to be useful to estimate the partial derivatives for a complex model with large number of parameters.

Various mathematical formulations have been proposed to address model uncertainty[118-123]. These formulations can be categorized as model-focused or prediction-focused depending upon whether the attention is directed towards the plausibility of the model hypotheses or to the accuracy of its predictions. However, characterization of model uncertainty still remains a difficult task due to the difficulties in making credible assumptions about likely processes and events, expressing them through selection of appropriate conceptual models and input variables, and choosing the epistemic probability for each selected conceptual models. Bounding analyses and experimental investigations could be included to distinguish between alternative conceptual models and narrowing the options. Bayesian analysis could be used to estimate the likelihood of the fitness of a given conceptual model relative to others[124] or to combine the family of aleatory distributions of models which indicate the analyst's confidence in each selected model[122]. But the burden of relying on expert judgments in the assessment of model uncertainty remains.

Possible improvements in addressing model uncertainty could come from the application of a logic tree approach[125,126]. A logic tree is a generalization of the fault tree and event tree approach in which specific sequences of events or faults are modeled in time where uncertainties in states of nature and in the occurrences of future events are addressed. The approach provides a systematic tool for conceptual representation of uncertain models and scenarios.

One important method of reducing model uncertainty is validation, which is the process by which assurance is obtained that conceptual and mathematical models, as employed in the computer model, are an accurate representation of the process or the system for which the models are intended[124]. Ideally, validation consists of a comparison between model predictions and observations of the real system over temporal and spatial scales relevant to performance assessment. However, the nature of the problem makes such tests very difficult. A proposed validation strategy for performance assessment includes "generic" as well as site-specific validation experiments[127]. These experiments could include laboratory tests, field tests, and natural analogs.

An uncertainty analysis that propagates parameter uncertainty through each credible model of the system, using Monte Carlo methods or some similar technique is suggested as a current NRC position[39] on providing a comprehensive examination of (model and parameter) uncertainty and its effect on compliance demonstrations. For the required specification of the parameter distributions, it is suggested to emphasize the knowledge of the parameter or "degree of belief" rather than concentrating on rigorous statistical efforts to determine distributions.

Characterization of uncertainty in LLW performance assessment requires further contributions. Availability of a more comprehensive systems code that integrates all key component models/ processes of performance assessment for engineered LLW disposal facility is desirable. Specification of parameter distributions, identification and selection of credible model of the system, and developing more systematic framework to address model uncertainty are the areas for further investigations. Developing an analysis framework based on the concept of probabilistic risk assessment to address the probabilities of various events leading into human exposure/dose including an intruder scenario is worthy to be considered in the future as exercised in HLW performance assessment. This would be useful to overcome the unrealistic nature of assumed scenarios and events built into existing methodologies of performance assessment.

Concluding Remarks

Performance assessment tries to project the prospective performance of a complex engineered and natural system over long periods of time. Typically, whenever adequate scientific information is not available, a very conservative approach is employed. It has been found in some conceptual performance assessments that overly conservative assumptions resulted in conceptual non-compliance with regulatory requirements. The approaches that are being taken to remedy this include better characterization and modeling in areas identified by the performance assessments as being the most important to estimating dose to humans. In this way, performance assessment is and will continue to be used to both refine the analyses, as well as be a tool to demonstrate the compliance of a particular site to the regulatory requirements.

Many uncertainties in performance assessment can be eliminated by careful selection and characterization of a candidate site. However, since performance assessment relies heavily on the use of computer models, model uncertainty remains a major portion of uncertainty in any performance assessments. NRC's BTP on performance assessment[39] stresses that model uncertainty should be addressed in all performance assessments and recommends the use of comparing multiple alternative models, where use of unreasonable modeling assumptions must be avoided.

Efforts of developing adequate or enhanced models, for some of the less characterized processes which appear to have major impacts on the performance objectives, should be continued as long as they lead to a more defensible decision. Although performance assessment does not aim to predict dose, in reality, the results of performance assessment are often interpreted by the public as the actual risk involved. In this respect, reducing uncertainty in performance assessment is very important for better communication of risk.

The models reviewed in this study can be important tools for various analysis in performance assessment. Selection and use of these models should be exercised based on the identified needs of a site and the desired goals of the decisions involved.

The key for the successful performance assessment modeling is proper conceptualization of a real-world system maintaining the balance between oversimplification and undersimplification leading to a societally acceptable and technically defensible decision making.

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References

1. W. E. Kennedy, “General Perspective on Performance Assessment,” Proceedings of the 18th DOE LLW Management Conference, Salt Lake City, Utah, May 20-22, 1997.
2. U.S. Nuclear Regulatory Commission, Code of Federal Regulations, Title 10, Chapter 1, Part 61 “Licensing Requirements for Land Disposal of Radioactive Waste,” Fed. Registr. 47, 57446 (1982).
3. North Carolina Regulations for Protection against Radiation, 15A NCAC 11, N.C. Department of Environment, Health, and Natural Resources, Division of Radiation Protection, July, 1993.
4. J. Cline, “Realizing More Accurate Waste Characterization for Disposal Site Dose Risk Assessment,” 1997 EPRI International LLW Conference, Providence, RI, July 21-23, 1997.
5. Electric Power Research Institute, “Performance Assessment and LLW Characterization,” in Low Level Waste Characterization Guidelines, EPRI TR-107201, 2691, November 1996.
6. F. W. Bond, C. R. Cole, and P. J. Gutknecht, “Unsaturated Groundwater Flow Model (UNSAT1D) Computer Code Manual,” CS-2434-CCM, Electric Power Research Institute, 1984.
7. M. Th. van Genuchten, “Calculating the Unsaturated Hydraulic Conductivity with a New Closed-Form Analytical Model,” Water Resource Program, Department of Civil Engineering, Princeton University, Princeton, NJ, 1978; “A Closed-Form Equation for Predicting the Hydraulic Conductivity of Unsaturated Soils,” Soil Science Society of America Journal, Vol. 44 pp. 892-898, 1980.

8. R. Haverkamp, M. Vaclin, J. Touma, P. J. Wierenga, and G. Vachaud, "A Comparison of Numerical Simulation Models for One-Dimensional Infiltration," Soil Science Society of America Journal, Vol. 41, pp. 285-294, 1977.

9. A. T. Corey, "Mechanics of Heterogeneous Fluids in Porous Media," Water Resources Publications, Fort Collins, CO, 1977.

10. C. Y. Hung, "A Model to Simulate Infiltration of Rainwater through the Cover of a Radioactive Waste Trench under Saturated And Unsaturated Conditions," in Role of the Unsaturated Zone in Radioactive and Hazardous Waste Disposal, J. W. Mercer et. al. (editor), Ann Arbor Science, 1983.

11. P. R. Schroeder, T. S. Dozier, P. A. Zappi, B. M. McEnroe, J. W. Sjostrom, and R. L. Peyton, "The Hydrologic Evaluation of Landfill Performance (HELP) Model: Engineering Documentation for Version 3," EPA/600/9-94/168, U. S. Environmental Protection Agency Risk Reduction Engineering Laboratory, 1994.

12. M. J. Fayer and T. L. Jones, "UNSAT-H Version 2.0: Unsaturated Soil Water and Heat Flow Model," PNL-6779, Pacific Northwest Laboratory, 1990.

13. W. E. Nichols and M. D. White, "MSTS: Multiphase Subsurface Transport Simulator User's Guide and Reference," PNL-8637, Pacific Northwest Laboratory, 1993.

14. A. K. Runchal and B. Sagar, "PORFLOW: A Multifluid Multiphase Model for Simulating Flow, Heat Transfer, and Mass Transport in Fractured Porous Media, User's Manual - Version 2.41," NUREG/CR-5991, CNWRA 92-003, U.S. Nuclear Regulatory Commission, 1993.

15. P. D. Meyer, "Application of an Infiltration Evaluation Methodology to a Hypothetical Low-Level Waste Disposal Facility," NUREG/CR-6114, PNL-8842, Vol. 1, U. S. Nuclear Regulatory Commission, 1993.

16. P. J. Wierenga, "Solute Distribution Profiles Computed with Steady-State and Transient Water Movement Models," Soil Sci. Soc. Am. J., Vol. 41 pp. 1050-1055, 1977.

17. F. Beese and P. J. Wierenga, "Solute Transport Through Soil with Adsorption and Root Water Uptake Computed with a Transient and a Constant-Flux Model," Soil Science, Vol. 129, No. 4, pp. 245-252, 1980.

18. V. Rogers and C. Hung, "PRESTO-EPA-CPG: A Low-Level Radioactive Waste Environmental Transport and Risk Assessment Code, Documentation and User's Manual," EPA 520/1-87-026, U.S. EPA, December 1987.

19. J. C. Walton, L. E. Plansky, and R. W. Smith, "Models for Estimation of Service Life of Concrete Barriers in Low-Level Radioactive Waste Disposal," NUREG/CR-5542, EGG-2597, U. S. Nuclear Regulatory Commission, 1990.

20. R. Guppy, "Autogenous Healing of Cracks in Concrete and its Relevance to Radwaste Repositories," NSS/R-105, Nirex, United Kingdom, 1988.

21. R. Shuman, V. C. Rogers, N. Chau, and G. B. Merrell, “The BARRIER Code: A Tool for Estimating the Long-Term Performance of Low-Level Radioactive Waste Disposal Facilities,” EPRI NO-7167-CCML, Electric Power Research Institute, May 1991.

22. K. A. Snyder and J. R. Clifton, “4SIGHT Manual: A Computer Program for Modeling Degradation of Underground Low Level Waste Concrete Vaults,” NISTIR 5612, National Institute of Standards and Technology, June 1995.

23. R. R. Seitz and J. C. Walton, “Modeling Approaches for Concrete Barriers Used in Low-Level Waste Disposal,” NUREG/CR-6070, EGG-2701, U.S. Nuclear Regulatory Commission, 1993.

24. R. Shuman, N. Chau, and V. C. Rogers, “Improved Modeling of Engineered Barriers for Low-Level Waste Disposal,” Waste Management ‘91, Vol. 2, pp.757-762, 1991.

25. J. S. Y. Wang and T. N. Narasimhan, “Hydrologic Mechanisms Governing Fluid Flow in a Partially Saturated, Fractured, Porous Medium,” Water Resour. Res., 21 (12), pp. 1861-1874, 1985.

26. R. R. Seitz, J. C. Walton, C. A. Dicke, and C. S. Smith, “Near-Field Performance Assessment for the Saltstone Disposal Facility,” Scientific Basis for Waste Management XVI, Materials Research Society Symposium Proceedings, 294, pp. 731-736, 1993.

27. S. R. Yates, “Seepage in a Saturated-Stratified Aquifer with Recharge,” Soil Science Society of America Journal, 52, pp. 356-363, 1988.

28. J. C. Walton and R. R. Seitz, “Performance of Intact and Partially Degraded Concrete Barriers in Limiting Fluid Flow,” NUREG/CR-5614, U.S. Nuclear Regulatory Commission, 1993.

29. J. R. Cook and P. D. Hunt, “Radiological Performance Assessment for the E-Area Vaults Disposal Facility,” WSRC-RP-94-218, Westinghouse Savannah River Company, 1994.

30. T. M. Sullivan, “Disposal Unit Source Term (DUST) Data Input Guide,” NUREG/CR6041, BNL-NUREG-52375, U. S. Nuclear Regulatory Commission, 1993.

31. T. M. Sullivan and C. J. Suen, “Low-Level Waste Shallow Land Disposal Source Term Model: Data Input Guides,” NUREG/CR-5387, BNL-NUREG-52206, U. S. Nuclear Regulatory Commission, 1989.

32. A. R. Hoch, C. C. Naish, S. M. Sharland, A. C. Smith, and K. J. Taylor, “Experiments and Modeling Studies Concerning Localized Corrosion of Carbon Steel and Stainless Steel Containers for Intermediate- and Low-Level Radioactive Waste,” Materials Research Society Symposium Proceedings, Vol. 294, 1993.

33. S. M. Sharland, G. P. Marsh, C. C. Naish, and K. J. Taylor, “The Assessment of Localized Corrosion of Carbon and Stainless Steel Containers for Intermediate- and Low-Level Radioactive Waste Under Repository Conditions,” Proc. of Topical Meeting on Nuclear Waste Packaging, Focus ‘91, September 29 - October 2, 1991, Las Vegas, NV, American Nuclear Society, 1991.

34. M. W. Kozak, M. S. Y. Chu, P. A. Mattingly, J. D. Johnson, and J. T. McCord, "Background Information for the Development of a Low-Level Waste Performance Assessment Methodology - Computer Code Implementation and Assessment," NUREG/CR-5453, SAND90-0375, Vol. 5, U. S. Nuclear Regulatory Commission, 1990.

35. M. -S. Yim, "Evaluation of Uncertainties in Low-Level Waste Source Term Analysis," Proceedings of the 18th DOE LLW Management Conference, Salt Lake City, Utah, May 20-22, 1997.

36. B. L. O'Neal and C. E. Lee, "IMPACTS-BRC, Version 2.0," NUREG/CR-5517, SAND89-3060, U. S. Nuclear Regulatory Commission, 1990; O. I. Oztunali and G. W. Roles, "Update of Part 61, Impacts Analysis Methodology," NUREG/CR-4370, U.S. Nuclear Regulatory Commission, 1986.

37. A. S. Rood, "GWSCREEN: A Semi-Analytical Model for Assessment of the Groundwater Pathway from Surface or Buried Contamination," EGG-GEO-10797, Rev. 2, 1994.

38. U. S. NRC, "Draft Environmental Impact Statement on 10CFR Part 61," NUREG-0782, Vol. 4, U. S. Nuclear Regulatory Commission, 1981.

39. U. S. Nuclear Regulatory Commission, "Branch Technical Position on a Performance Assessment Methodology for Low-Level Radioactive Waste Disposal Facilities," NUREG-1573, 1997.

40. U. S. Nuclear Regulatory Commission, "Overview of NRC Staff LLWPA Modeling Efforts," Low-Level Waste Performance Assessment Workshop, Rockville, MD, November 17, 1994.

41. N. E. Olague, D. E. Longsine, J. E. Campbell, C. D. Leigh, "User's Manual for the NEFTRAN II Computer Code," NUREG/CR-5618, SAND90-2089, U. S. Nuclear Regulatory Commission, 1991.

42. R. J. McKinnon, T. M. Sullivan, C. J. Suen, and S. A. Simonson, "BLT-EC (Breach, Leach, Transport, and Equilibrium Chemistry), a Finite-Element Model for Assessing the Release of Radionuclides from Low-Level Waste Disposal Units: Background, Theory, and Model Description," NUREG/CR-6305, U. S. Nuclear Regulatory Commission, 1995.

43. T. M. Sullivan and A. Gard, "DUST-MS - Disposal Unit Source Term - Multiple Species: Data Input Guide," in preparation.

44. T. M. Sullivan, R. J. MacKinnon, R. R. Kinsey, A. Aronson, and M. Divadeenam, "BLT-MS (Breach, Leach and Transport - Multiple Species) Data Input Guide: A Computer Model for Simulating Release of Contaminant from Subsurface Low-Level Waste Disposal Facility," NUREG/CR-6492, BNL-NUREG-52509, 1996.

45. U. S. Nuclear Regulatory Commission, Technical Position on Waste Form (Rev. 1), Technical Branch, Division of Low-Level Waste Management and Decommissioning, 1991.

46. H. Arora and R. Dayal, “Leaching Studies of Cement-Based Low-Level Radioactive Waste Forms,” Topical Report, NUREG/CR-4756, BNL/NUREG-52026, Brookhaven National Laboratory, 1986.

47. C. L. Kim, C. H. Cho, and H. J. Choi, “Equilibrium Concentration and Diffusion Controlled Leaching Model for Cement-Based Waste Forms,” Waste Management, 15 (5/6), 449-455, 1995.

48. M. -S. Yim, “Development of A New Mechanistic Low-Level Waste Source Term Model,” Scientific Basis for Nuclear Waste Management XX, Mat. Res. Soc. Symp., Vol. 465, pp. 1059-1066, 1997.

49. S. -I. Su and M. -S. Yim, “Source Term Analysis Methodology for LLW Disposal Facilities,” to be submitted to Radioactive Waste Management and Environmental Restoration, 1998.

50. S. -I. Su and M. -S. Yim, “Analysis of Radionuclide Release from Spent Ion-Exchange Resins,” (accepted for publication), Nuclear Technology, 1999.

51. S. B. Oblath, “Leaching from Solidified Waste Forms under Saturated and Unsaturated Conditions,” Environmental Science and Technology, Vol. 23, pp. 1098-1102, 1989.

52. G. G. Eichholz, M. F. Petelka, and J. Whang, “Leaching and Migration of Low-Level Wastes Under Unsaturated Conditions,” Radioactive Waste Management and the Nuclear Fuel Cycle, Vol. 14, pp. 285-310, 1990.

53. M. F. Petelka, “Leaching of Radioactive Waste Forms under Saturated and Unsaturated Flow Conditions,” Ph.D. Thesis, Georgia Institute of Technology, 1987.

54. M. -S. Yim and S. A. Simonson, “Generation and Release of Radioactive Gases in LLW Disposal Facilities,” Proceedings of the 23rd DOE/NRC Nuclear Air Cleaning and Treatment Conference, NUREG/CP-0141, CONF-940738, 1995.

55. M.-S. Yim, “Gas-Phase Release of Radionuclides from Low Level Waste Disposal Facilities,” Sc.D. Thesis, Harvard University School of Public Health, 1994.

56. T. M. Sullivan, “Selection of Models to Calculate the LLW Source Term,” NUREG/CR-5773, BNL-NUREG-52295, U. S. Nuclear Regulatory Commission, 1991.

57. M. W. Kozak, C. P. Harlan, M. S. Y. Chu, and B. L. O’Neal, “Background Information for the Development of a Low-Level Waste Performance Assessment Methodology - Selection and Integration of Models,” NUREG/CR-5453, SAND89-2509, Vol. 3, U. S. Nuclear Regulatory Commission, 1989.

58. K. M. Krupka and R. J. Serne, “Effects of Radionuclide Concentrations by Cement/Ground-Water Interactions in Support of Performance Assessment of Low-Level Radioactive Waste Disposal Facilities,” NUREG/CR-6377, PNNL-11408, U. S. Nuclear Regulatory Commission.

59. V. C. Rogers and C. Hung, "PATHRAE-EPA: A Low-Level Radioactive Waste Environmental Transport and Risk Assessment Code," EPA 520/1-87-028, U. S. Environmental Protection Agency, 1987.

60. D. H. Tang, E. O. Frind, and E. A. Sudicky, "Contaminant Transport in Fractured Porous Media: Analytical Solution for a Single Fracture," Water Resources Research, Vol. 17, No. 3, pp. 555-564, 1981.

61. E. A. Sudicky and E. O. Frind, "Contaminant Transport in Fractured Porous Media: Analytical Solutions for a System of Parallel Fractures," Water Resources Research, Vol. 18, No. 6, pp. 1634-1642, 1982.

62. J. C. Walton, "Performance of Intact and Partially Degraded Concrete Barriers in Limiting Mass Transport," NUREG/CR-5445, EGG-2662, U. S. Nuclear Regulatory Commission, 1992.

63. M. W. Kozak, N. E. Olague, R. R. Rao, and J. T. McCord, "Evaluation of a Performance Assessment Methodology for Low-Level Radioactive Waste Disposal Facilities," NUREG/CR-5927, SAND91-2802, Vol. 1, U. S. Nuclear Regulatory Commission, 1993.

64. M. W. Kozak, M. S. Y. Chu, and P. A. Mattingly, "A Performance Assessment Methodology for Low-Level Waste Facilities," NUREG/CR-5532, SAND90-0375, U. S. Nuclear Regulatory Commission, 1990.

65. American Society of Civil Engineers (ASCE), "Groundwater Quality: Guideline for Selection of Commonly Used Groundwater Models," ASCE, New York, 1995.

66. American Society for Testing and Materials (ASTM), "Guide for Application of a Ground-Water Flow Model to a Site-Specific Problem," ASTM, Philadelphia, PA, 1993.

67. U. S. Environmental Protection Agency, "Assessment Framework for Ground-Water Model Applications (Draft)," Office of Solid Waste and Emergency Response, U. S. EPA, 1992.

68. T. E. Reilly, O. L. Franke, H. T. Buxton, and G. D. Bennett, "A Conceptual Framework for Ground-Water Solute-Transport Studies with Emphasis on Physical Mechanisms of Solute Movement," U. S. Geological Survey Water-Resources Investigations Report 87-4191, 1987.

69. J. Weaver, C. G. Enfield, S. Yates, D. Kreamer, and C. White, "Predicting Subsurface Contaminant Transport and Transformation: Considerations for Model Selection and Field Validation," EPA/600/2-89/045, Robert S. Kerr Environmental Research Laboratory, U. S. EPA, Ada, OK, 1989.

70. National Research Council, "Ground Water Models: Scientific and Regulatory Applications," National Academy Press, Washington, DC, 1990.

71. P. K. M. van der Heijde and O. A. Elnawawy, "Quality Assurance and Quality Control in the Development and Application of Ground-Water Models," EPA/600/R-93/011, U. S. EPA, Ada, OK, 1992.

72. M. P. Anderson and W. W. Woessner, "Applied Groundwater Modeling: Simulation of Flow and Advective Transport," Academic Press, 1992.

73. C. Zheng and G. D. Bennett, "Applied Contaminant Transport Modeling: Theory and Practice," Van Nostrand Reinhold, 1995.

74. R. A. Freeze and J. A. Cherry, "Groundwater," Prentice Hall, 1979.

75. C. S. Simmons, C. T. Kincaid, and A. E. Reisenauer, "A Simplified Model for Radioactive Contaminant Transport: the TRNSS Code," PNL-6029, Pacific Northwest Laboratory, 1986.

76. P. S. Huyakorn, J. B. Kool, and J. B. Robertson, "Documentation and User's Guide: VAM2D - Variably Saturated Analysis Model in Two Dimensions," NUREG/CR-5352, HGL/89-01, U. S. Nuclear Regulatory Commission, 1989.

77. E. G. Lappala, R. W. Healy, and E. P. Weeks, "Documentation of Computer Program VS2D to Solve the Equations of Fluid Flow in Variably Saturated Porous Media," Water Resources Investigation Report 83-4099, U. S. Geological Survey, 1987.

78. R. W. Healy, "Simulation of Solute Transport in Variably Saturated Porous Media with Supplemental Information on Modifications to the U.S. Geological Survey's Computer Program VS2D," Water Resources Investigation Report 90-4025, U.S. Geological Survey, 1990.

79. G. T. Yeh and D. S. Ward, "FEMWATER: A Finite Element Model of Water Flow Through Saturated-Unsaturated Porous Media," ORNL-5567, Oak Ridge National Laboratory, 1980.

80. G. T. Yeh and D. S. Ward, "FEMWASTE: A Finite-Element Model for Waste Transport Through Saturated-Unsaturated Porous Media," ORNL-5601, Oak Ridge National Laboratory, 1981.

81. M. G. MacDonald and A. W. Harbaugh, "A Modular Three-Dimensional Finite-Difference Ground-Water Flow Model," U.S. Geological Survey Techniques of Water Resources Investigations, Book 6, Chapter A1, 1988.

82. D. W. Pollock, "Documentation of Computer Programs to Compute and Display Pathlines Using Results from the U.S. Geological Survey Modular Three-Dimensional Finite-Difference Ground-Water Flow Model," U.S. Geological Survey Open File Report 89-381, 1989.

83. A. B. Gureghian, "Analytical Solution for Multidimensional Transport of Four-Member Radionuclide Decay Chain in Ground Water," BMI/OCRD-25, 1987.

84. C. I. Voss, “A Finite-Element Simulation Model for Saturated-Unsaturated Fluid-Density-Dependent Ground Water Flow with Energy Transport or Chemically-Reactive Single-Species Solute Transport,” U.S. Geological Survey Water-Resource Investigations Report 84-4369, 1984.

85. B. J. Travis, “TRACR3D: A Model of Flow and Transport in Porous/Fractured Media,” LA-9667-MS., Los Alamos National Laboratory, 1984.

86. R. L. England, N. W. Kline, K. J. Ekblad, and R. G. Baca, “MAGNUM-2D Computer Code: User’s Guide,” RHO-CR-143 P, Rockwell Hanford Operations, 1986.

87. N. W. Kline, R. L. England, and R. G. Baca, “CHAINT Computer Code: User’s Guide,” RHO-CR-144 P, Rockwell Hanford Operations, 1986.

88. W. Dershowitz, G. Lee, J. Geier, T. Foxford, and P. LaPointe, Fracman Interactive Discrete Fracture Data Analysis, Geometric Modeling and Exploration Simulation: User Documentation Version 2.6, Golder Associates Inc., 1997.

89. A. B. Gureghian, “A Two-Dimensional Finite Element Model for the Simultaneous Transport of Water and Reacting Solutes Through Saturated and Unsaturated Porous Media,” Technical Report ONWI-465, 1983.

90. Personal Communications, J. W. Latham, Chem-Nuclear Systems, Inc., 1996.

91. R. B. Codell, K. T. Key, and G. Whelan, “A Collection of Mathematical Models for Dispersion in Surface Water and Groundwater, NUREG-0868, U.S. Nuclear Regulatory Commission, 1982.

92. Personal Communications, D. Evans, Department of Marine, Earth, and Atmospheric Science, North Carolina State University, 1997.

93. L. R. Shipers and C. P. Harlan, “Background Information for the Development of a Low-Level Waste Performance Assessment Methodology,” NUREG/CR-5453, SAND89-2509, Vol. 2, U.S. Nuclear Regulatory Commission, 1989.

94. S. R. Hanna, G. A. Briggs and R. P. Hosker, Handbook on Atmospheric Diffusion, DOE/TIC-11223 (DE82002045), U.S. Department of Energy, 1982.

95. R. E. Faw and J. K. Shultis, “Radiological Assessment,” PTR Prentice Hall, 1993.

96. R. E. Moore, C. F. Baes, L. M. McDowell-Boyer, A. P. Watson, F. O. Hoffman, J. C. Pleasant, and C. W. Miller, “AIRDOS-EPA: A Computerized Methodology for Estimating Environmental Concentrations and Dose to Man from Airborne Releases of Radionuclides,” ORNL-5532, Oak Ridge National Laboratory, 1979.

97. J. F. Sagendorf, J. T. Goll, and W. F. Sandusky, “XOQDOQ: Computer Program for the Meteorological Evaluation of Routine Effluent Releases at Nuclear Power Stations,” NUREG/CR-2919, PNL-4380, 1982.

98. D. E. Fields and C. W. Miller, “User’s Manual for DWNWND - An Interactive Implementation of the Gaussian Plume Atmospheric Transport Model with Eight Dispersion Parameter Options,” ORNL/TM-6874, Oak Ridge National Laboratory, 1980.

99. B. S. Parks, “User’s Guide for CAP88-PC, Version 1.0,” 402-B-92-001, U. S. Environmental Protection Agency, 1992.

100. B. A. Napier, D. L. Strenge, R. A. Peloquin, and J. V. Ramsdell, “GENII - The Hanford Environmental Radiation Dosimetry Software System,” PNL-6584 Vol. 1 and 2, Pacific Northwest Laboratory, 1988.

101. J. E. Till and H. R. Meyer, “Radiological Assessment, A Textbook on Environmental Dose Analysis,” NUREG/CR-3332, ORNL-5968, U. S. Nuclear Regulatory Commission, 1983.

102. D. L. Strenge, R. A. Peloquin, and G. Whelan, “LADTAP II - Technical Reference and User Guide,” NUREG/CR-4013, PNL-5270, U.S. Nuclear Regulatory Commission, 1986.

103. U. S. Nuclear Regulatory Commission, “Calculation of Annual Doses to Man from Routine Releases of Reactor Effluents for the Purpose of Evaluating Compliance with 10 CFR Part 50, Appendix I,” Regulatory Guide 1.109, Revision 1, 1977.

104. U. S. Nuclear Regulatory Commission, “Guidance on Using Decision Methods for Dose Assessment to Comply With Radiological Criteria for License Termination,” NUREG-1549, Vol. 2 (Draft), 1998.

105. C. Yu, et. al., Manual for Implementing Residual Radioactive Material Guidelines Using RESRAD, Version 5.0, ANL/EAD/LD-2, Environmental Assessment Division, Argonne National Laboratory, 1993.

106. C. D. Leigh, B. M. Thompson, J. E. Campbell, D. E. Longsine, R. A. Kennedy, and B. A. Napier,” User’s Guide for GENII-S: A Code for Statistical and Deterministic Simulations of Radiation Doses to Humans from Radionuclides in the Environment,” SAND91-0561, Sandia National Laboratories, 1992.

107. J. W. Buck, G. Whelan, J. G. Droppo, D. L. Strenge, K. J. Castleton, J. P. McDonald, C. Sato, and G. P. Streile, “Multimedia Environmental Pollutant Assessment System (MEPAS) Application Guidance, *Guidelines for Evaluating MEPAS Input Parameters for Version 3.1*,” PNL-10395, Pacific Northwest Laboratory, 1995.

108. ICRP 30, “Limits for Intakes of Radionuclides by Workers,” ICRP Publication 30, Part 1, Pergamon Press, New York, 1982.

109. ICRP 60, “Recommendations of the International Commission on Radiological Protection,” ICRP Publication 60, Pergamon Press, New York, 1990.

110. M. Baek, S. K. Lee, U. C. Lee, and C. S. Kang, “A probabilistic Approach to Quantify the Uncertainties in Internal Dose Assessment Using Response Surface and Neural Network,” International Conference on Nuclear Engineering, Vol. 3, pp. 361-373, ASME, 1996.

111. Palisade Corporation, @RISK, 1-800-432-7475, New Field, NY.

112. Golder Associates, RIP Performance Assessment and Strategy Evaluation Model, Theory Manual and User's Guide, 1996.

113. Private Communications, U. S. Nuclear Regulatory Commission, 1996.

114. M. W. Kozak, N. E. Olague, D. P. Gallegos, and R. R. Rao, "Treatment of Uncertainty in Low-Level Waste Performance Assessment," Proceedings of the 13th DOE LLW Management Conference, Atlanta, GA, November 19-21, 1991.

115. S. J. Maheras and M. R. Kotecki, "Guidelines for Sensitivity and Uncertainty Analyses of Performance Assessment Computer Codes," DOE/LLW-100, National Low Level Waste Management Program, 1990.

116. D. C. Cox and P. Baybutt, "Methods for Uncertainty Analysis: A Comparative Survey," Risk Analysis, Vol. 1, No. 4, pp. 251-258, 1981.

117. B. A. Worley, Deterministic Uncertainty Analysis, ORNL-6428, 1987.

118. G. E. Apostolakis, "The Concept of Probability in Safety Assessments of Technological Systems," Science, 250, pp. 1359-1364, 1990.

119. G. E. Apostolakis, "A Commentary on Model Uncertainty," Proceedings of Workshop I on Advanced Topics in Risk and Reliability Analysis, A Mosleh, N. Siu, C. Smidts and C. Lui (eds.), NUREG/CP-0138, U.S. Nuclear Regulatory Commission, Annapolis, MD, Oct. 20-22, 1993.

120. R. L. Winkler, "Model Uncertainty: Probabilities for Models?", Proceedings of Workshop I on Advanced Topics in Risk and Reliability Analysis, A Mosleh, N. Siu, C. Smidts and C. Lui (eds.), NUREG/CP-0138, U.S. Nuclear Regulatory Commission, Annapolis, MD, Oct. 20-22, 1993.

121. E. Zio, G. E. Apostolakis, D. Okrent, "Towards a Quantitative Treatment of Model Uncertainty in the Performance Assessment of High-Level Radioactive Waste Repositories," Proceedings of WM'95, Tucson, Arizona, 1995.

122. E. Zio and G. E. Apostolakis, "A Norm-Based Approach to the Quantification of Model Uncertainty," Proceedings of the 7th HLW Conference, Las Vegas, 1996.

123. E. Zio, "Quantitative Analysis of Model Uncertainty: Estimating the Credibility of Alternate Model Structures," Proceedings of the International Topical Meeting on Probabilistic Safety Assessment, PSA'96, pp. 1219-1226, American Nuclear Society, Park City, UT, September 29 - October 3, 1996.

124. P. Davis, E. J. Bonano, K. K. Wahi and L. L. Price, "Uncertainties Associated with Performance Assessment of High-Level Radioactive Waste Repositories," NUREG/CR-5211, SAND88-2703, U. S. Nuclear Regulatory Commission, 1990.

125. Electric Power Research Institute, “Demonstration of a Risk-Based Approach to High-Level Waste Repository Evaluation: Phase 2,” EPRI TR-100384, 1992.

126. Electric Power Research Institute, “Yucca Mountain Total System Performance Assessment, Phase 3,” EPRI TR-107191, 1996.

127. P. A. Davis and M. T. Goodrich, “Technical Basis for Judging the Validity of Models for Performance Assessment of HLW Repositories,” NUREG/CR-5537, SAND90-0575, U. S. Nuclear Regulatory Commission, 1991.

128. Personal Communications, Terry M. Sullivan, Brookhaven National Laboratory, 1999.

B

INVENTORY DATABASE

The data described in the following spreadsheet was used for the comparisons and analyses discussed in Chapter 3 of the report. The inventory is based upon manifest data and has been summarized so that different combinations of wastes could be chosen in a particular inventory makeup. The basis for this inventory estimate may become dated as disposal and operational practices change, but the methodology of computing detailed inventory estimates and distributions in a LLW facility is important for performance assessment.

Table B-1

LLW C-14 Inventory

		Form	Class	Chelates	6yr avg 89–94 Ann Vol m3	Vol %	C14 Ci/m3	Ci	Ci absrbing w	Ci activted me	Ci trash	Ci solidified w	Ci inciner/sol	Ci in CsD	Ci in CsL	Ci inHIC	Fpaper DAW csd resin
Utility	Resins	Dewatered	AS	No	6.41E+02	7.474013957	2.70E-03	1.7307	1.7307					0	0	1.7307	0
			AU	No	5.91E+02	6.891017548	2.03E-03	1.19973	1.19973					0.959784	0.239946	0	0.959784
			B	No	1.58E+02	1.842268651	2.86E-02	4.5188	4.5188							4.5188	
			C	No	3.33E+01	0.388275608	2.28E-01	7.5924	7.5924							7.5924	
		Cement	AS	No	5.41E+00	0.063080211	1.77E-03	0.0095757				0.0095757		0	0	0.0095757	
			AU	No	1.20E+01	0.139919138	6.56E-05	0.0007872				0.0007872		0.0007872	0		
			C	No	4.23E-01	0.00493215	4.46E-03	0.00188658				0.00188658		0	0.00188658		
		Dewatered	AS	Yes	1.26E+01	0.146915095	1.04E-03	0.013104	0.013104					0	0	0.013104	0
			AU	Yes	9.40E-01	0.010960332	2.06E-04	0.00019364	0.00019364					0.000154912	0.000038728		0.000154912
			B	Yes	3.22E+00	0.037544969	9.30E-03	0.029946	0.029946							0.029946	
		Cement	AU	Yes	3.02E-01	0.003521298	4.92E-04	0.000148584				0.000148584		0.000148584	0		
		Dow media	AS	Yes	5.90E-01	0.006879358	9.72E-04	0.00057348				0.00057348		0	0	0.00057348	
	Solid combustible	solid	AU	No	9.00E-02	0.001049394		0			0			0	0		
ft3	SolidnonComb	sealed source	AS	No	4.83E-02	0.000563175		0		0				0	0		
			AU	No	3.30E-02	0.000384778		0		0				0	0		
			B	No	3.42E-02	0.00039877		0		0					0	0	
			C	No	3.00E-01	0.003497978	1.10E-02	0.0033		0.0033					6.60E-05	0.003234	
		Dewatered	AS	No	5.72E-01	0.006669479	1.42E-03	0.00081224	0.00081224					0	0	0.00081224	
			AU	No	9.05E-01	0.010552235	1.29E-05	1.17E-05	1.17E-05					9.34E-06	2.33E-06		
			B	No	2.75E-01	0.00320648	1.21E-04	0.000033275	0.000033275						6.66E-07	3.26E-05	
		cement	AU	No	5.85E-01	0.006821058	8.83E-05	5.17E-05				5.17E-05		5.17E-05			
			AS	No	1.80E-01	0.002098787		0				0		0	0		
			B	No	9.06E-01	0.010563895	1.30E-03	0.0011778				0.0011778		0	0.0011778		
		bitumen	AU	No	4.20E-02	0.000489717		0				0		0	0		
		solid	AS	No	6.08E-02	0.000708924		0			0			0	0		
			AU	No	1.15E+02	1.335061775	2.48E-04	0.028396			0.028396			0.0227168	0.0056792		
			B	No	9.05E-01	0.010552235	4.15E-03	0.00375575			0.00375575				7.51E-05	0.003680635	
			C	No	3.69E-01	0.004302513	8.32E-02	0.0307008			0.0307008				0.000614016	0.030086784	
	Filter media	dewatered	AS	No	2.91E+01	0.33930391	6.84E-03	0.199044	0.199044					0	0	0.199044	
			AU	No	4.13E+00	0.048097204	2.52E-03	0.010395	0.010395					0.008316	0.002079		
			B	No	2.66E+00	0.031015409	5.65E-02	0.15029	0.15029							0.15029	
			C	No	4.56E+00	0.053169272	8.80E-03	0.040128	0.040128							0.040128	
		cement	AU	No	1.41E+00	0.016393859	1.06E-04	0.000149036				0.000149036		0.000149036	0		
			C	No	6.13E-01	0.007147536	4.48E-04	0.000274624				0.000274624		0	0.000274624		
		bitumen	AS	No	1.42E+00	0.016557098	8.20E-04	0.0011644				0.0011644		0.0011644	0		
		solid	AU	No	1.85E-01	0.002157087	5.42E-05	0.000010027				0.000010027		8.02E-06	2.01E-06		
	Cartrdge flter	dewatered	AU	No	1.13E+01	0.131757188	1.40E-04	0.001582	0.001582					0.0012656	0.0003164		
			AS	No	4.48E+00	0.052236478	1.74E-03	0.0077952	0.0077952					0.00623616	0.00155904		
			B	No	1.99E+01	0.232032571	5.28E-03	0.105072	0.105072							0.105072	
			C	No	2.79E+01	0.325311996	2.52E-02	0.70308	0.70308							0.70308	
		cement	AS	No	7.98E-01	0.009304623	3.57E-04	0.000284886				0.000284886		0.000284886	0		
			AU	No	1.47E-02	0.000171401	1.14E-04	1.68E-06				1.68E-06		1.68E-06	0		
			B	No	2.65E-01	0.003089881	1.87E-02	0.0049555				0.0049555		0	0.0049555		
			C	No	7.57E-01	0.008826566	2.31E-03	0.00174867				0.00174867		0	0.00174867		
	Solidfd liquid	cement	AS	No	8.06E+00	0.093979021	1.09E-02	0.087854				0.087854		0.087854	0		
			AU	No	3.53E+01	0.411595464	1.06E-03	0.037418				0.037418		0.037418	0		
			B	No	1.65E+00	0.019238881	8.66E-04	0.0014289				0.0014289		0	0.0014289		
			C	No	5.52E-01	0.00643628		0				0		0	0		
		bitumen	AS	No	3.10E+01	0.361457773	3.28E-03	0.10168				0.10168		0.10168	0		
			AU	No	1.08E+01	0.125927224	4.15E-03	0.04482				0.04482		0.04482	0		

Table B-1
LLW C-14 Inventory (continued)

0.7	Fpapr solidCom	0.3	Forg resinUt	0.55	Forg resinNU	0.8	forg other	0.8	Forg filtmed	0.2								
csd paper	**csd biolog**	**csd liquid**	**csd others**	**csd cement/sol**	**csd metal**	**csl resin**	**csl paper**	**csl biolog**	**csl liquid**	**csl other**	**csl cement/sol**	**csl metal w**	**hic resins**	**hic paper**	**hic other**	**hic cement**	**hic metal w**	**hic biolog**
						0							1.7307					
						0.239946												
						0							4.5188					
						0							7.5924					
				0							0					0.0095757		
				0.0007872							0							
											0.00188658							
						0							0.013104					
						0.000038728												
													0.029946					
				0.000148584														
				0												0.00057348		
															0			
				0														
				0														
												6.60E-05					0.003234	
					0							0					0.00081224	
					9.34E-06							2.33E-06						
												6.66E-07					3.26E-05	
				5.17E-05														
0																		
											0.0011778							
														0				
				0														
			0.0227168							0.0056792								
										7.51E-05					0.003680635			
										0.000614016					0.030086784			
			0							0					0.199044			
			0.008316							0.002079								
															0.15029			
															0.040128			
				0.000149036														
											0.000274624							
				0.0011644														
			8.02E-06							2.01E-06								
0.0012656							0.0003164											
0							0							0.0077952				
														0.105072				
														0.70308				
																0.000284886		
				1.68E-06														
											0.0049555							
											0.00174867							
																0.087854		
				0.037418														
											0.0014289							
																0.10168		
				0.04482														

Table B-1
LLW C-14 Inventory (continued)

subtotal	csd resinbio	csd otherbio	csl resinbio	csl otherbio	hic resinbio	hic otherbio	V in CSD	V in CSL	V in HIC	% contri	Co60 Ci/m3	C14/Co60
1.7307	0		0		0.951885	0	0	0.00E+00	6.41E+02	5.363635277	8.16E-01	3.31E-03
1.19973	0.5278812		0.1319703			0	472.8	1.18E+02		3.718099122	1.18E-01	1.72E-02
4.5188					2.48534	0		0.00E+00	1.58E+02	14.00427289	7.83E+00	3.65E-03
7.5924					4.17582	0		0.00E+00	3.33E+01	23.52970733	3.74E+01	6.10E-03
0.0095757					0	0	5.41E+00	0.00E+00		0.029676179	2.98E+00	5.94E-04
0.0007872					0	0	1.20E+01	0.00E+00		0.002439622	1.17E-01	5.61E-04
0.00188658					0	0	0	4.23E-01		0.005846725	1.20E-01	3.72E-02
0.013104	0		0		0.0072072	0	0	0.00E+00	1.26E+01	0.04061078	1.44E+00	7.22E-04
0.00019364	8.52E-05		2.13E-05			0	0.752	1.88E-01	0.00E+00	0.000600112	4.71E-02	4.37E-03
0.029946					0.0164703	0	0.00E+00	0.00E+00	3.22E+00	0.092806045	7.37E+00	1.26E-03
0.000148584					0	0	3.02E-01	0.00E+00		0.000460479	1.18E-02	4.17E-02
0.00057348					0	0	5.90E-01	0.00E+00		0.001777279	1.78E+00	5.46E-04
0					0	0	0.072	1.80E-02		0	1.68E-01	0.00E+00
0					0	0			4.83E-02	0		
0					0	0	0.0264	6.60E-03		0	1.69E+01	0.00E+00
0					0	0	0.02736	6.84E-03		0	2.28E-01	0.00E+00
0.0033					0	0		6.00E-03	0.294	0.010227074	7.51E-01	1.46E-02
0.00081224					0	0	0.4576	1.14E-01		0.002517224	2.26E-04	6.28E+00
1.17E-05					0	0				3.62E-05	4.12E+00	3.13E-06
0.000033275				5.32E-07	0	0		5.50E-03	0.2695	0.000103123	2.01E-03	6.02E-02
5.17E-05					0	p[oi	0.468	1.17E-01		0.000160086	4.61E-05	1.92E+00
0					0	0	0.144	3.60E-02		0	6.16E+00	0.00E+00
0.0011778					0	0		1.81E-02	0.88788	0.003650136	1.71E-03	7.60E-01
0					0	0		8.40E-04	0.04116	0	3.89E-03	0.00E+00
0					0	0	0.04864	1.22E-02		0	1.65E-02	0.00E+00
0.028396		0.01817344		0.00454336	0	0	91.6	2.29E+01		0.088002419	6.46E-01	3.84E-04
0.00375575				6.01E-05	0	0.002944508	0.724	1.81E-01		0.011639495	2.16E+00	1.92E-03
0.0307008				0.000491213	0	0.024069427		7.38E-03	0.36162	0.095145256	6.82E-01	1.22E-01
0.199044		0		0	0	0.1592352		5.82E-01	28.518	0.616859895	5.93E-02	1.15E-01
0.010395		0.0016632		0.0004158	0	0	3.3	8.25E-01		0.032215282	4.07E-01	6.19E-03
0.15029					0	0.030058	2.128	5.32E-01		0.465765728	2.71E+00	2.08E-02
0.040128					0	0.0080256		9.12E-02	4.4688	0.124361216	9.10E-02	9.67E-02
0.000149036					0	0		2.81E-02	1.37788	0.000461879	1.96E-01	5.41E-04
0.000274624					0	0	0.4904	1.23E-01		0.000851091	1.42E-02	3.15E-02
0.0011644					0	0	1.136	2.84E-01		0.003608607	4.32E-03	1.90E-01
0.000010027		1.60E-06		4.01E-07	0	0		3.70E-03	0.1813	3.11E-05	6.29E-02	8.62E-04
0.001582					0	0	9.04	2.26E+00		0.004902797	9.15E-01	1.53E-04
0.0077952					0	0	3.584	8.96E-01		0.024158207	7.16E+00	2.43E-04
0.105072					0	0		3.98E-01	19.502	0.325630026	5.64E+00	9.36E-04
0.70308					0	0		5.58E-01	27.342	2.178924534	4.47E+00	5.64E-03
0.000284886					0	0	0.6384	1.60E-01		0.000882894	7.30E-01	4.89E-04
1.68E-06					0	0	0.01176	2.94E-03		5.19E-06	2.96E+00	3.85E-05
0.0049555					0	0	0.212	5.30E-02		0.015357656	6.25E+00	2.99E-03
0.00174867					0	0	0.6056	1.51E-01		0.005419326	6.39E-01	3.62E-03
0.087854					0	0	6.448	1.61E+00		0.272269494	1.15E-01	9.48E-02
0.037418					0	0		7.06E-01	34.594	0.115962619	6.17E+00	1.72E-04
0.0014289					0	0		3.30E-02	1.617	0.004428323	4.34E-01	2.00E-03
0					0	0	0.4416	1.10E-01		0	4.86E-02	0.00E+00
0.10168					0	0	24.8	6.20E+00		0.315117834	8.32E-02	3.94E-02
0.04482					0	0	8.64	2.16E+00		0.138902255	7.83E-01	5.30E-03

Table B-1
LLW C-14 Inventory (continued)

		Form	**Class**	**Chelates**	**6yr avg 89–94 Ann Vol m3**	**Vol %**	**C14 Ci/m3**	**Ci**	**Ci absrbing w**	**Ci activted me**	**Ci trash**	**Ci solidified w**	**Ci inciner/sol**	**Ci in CsD**	**Ci in CsL**	**Ci inHIC**	**Fpaper DAW csd resin**
	Equipment	cement	AS	No	1.41E+00	0.016440499	1.03E-04	0.00014523				0.00014523		0.00014523	0		
			C	No	1.80E-01	0.002098787	5.49E-03	0.0009882				0.0009882		0	0.0009882		
		solid	AS	No	1.03E+01	0.12009726	3.37E-05	0.00034711		0.00034711				0.000277688	0.000069422		
			AU	No	7.47E-01	0.008709966	3.78E-02	0.0282366		0.0282366				0.02258928	0.00564732		
			B	No	2.33E+00	0.027167633	1.27E-03	0.0029591		0.0029591					5.92E-05	0.002899918	
			C	No	1.57E+01	0.183060872	4.93E-01	7.7401		7.7401					0.154802	7.585298	
	Inciner ash	cement	AU	No	7.13E+00	0.083135288	2.41E-03	0.0171833				0.0171833		0.0171833	0		
		solid	AU	No	3.91E+01	0.455903191	2.67E-03	0.104397					0.104397	0.104397	0		
	Air filters	cement	AU	No	1.88E+00	0.021920665	1.77E-06	3.33E-06				3.33E-06		3.33E-06	0		
		solid	AS	No	6.13E-02	0.000714754		0			0			0	0		
			AU	No	2.93E+01	0.341635895	9.34E-05	0.00273662			0.00273662			0.002189296	0.000547324		
	Mixed DAW	cement	AS	No	6.42E-01	0.007485674	3.87E-04	0.000248454				0.000248454		0.000248454	0		
		solid	AS	No	3.41E+01	0.397603551	4.97E-03	0.169477			0.169477			0.1355816	0.0338954		
			AU	No	1.66E+03	19.37880062	6.80E-04	1.13016			1.13016			0.904128	0.226032		
			B	No	1.54E+00	0.017956289	4.21E-02	0.064834			0.064834				0.00129668	0.06353732	
			C	No	3.03E+00	0.035329582	1.10E-02	0.03333			0.03333				0.0006666	0.0326634	
	Asbestos	solid	AS	No	3.45E-02	0.000402268	4.84E-05	1.67E-06			1.67E-06			1.34E-06	3.34E-07		
			AU	No	1.97E+01	0.229700585	7.13E-05	0.00140461			0.00140461			0.001123688	0.000280922		
Industry	Resin	dewatered	AS	No	8.95E-01	0.010435636	5.22E-05	0.000046719	0.000046719					3.74E-05	9.34E-06		
			AS	Yes	2.17E-01	0.002530204	5.04E-03	0.00109368	0.00109368					0.000874944	0.000218736		
			AU	No	7.04E+00	0.082085894	1.83E-04	0.00128832	0.00128832					0.001030656	0.000257664		0.001030656
		cement	AU	No	4.81E-02	0.000560843		0				0		0	0		
	solid combstbl	solid	AU	No	1.04E+00	0.012126325	5.75E-04	0.000598			0.000598			0.0004784	0.0001196		
		cement	AU	No	1.17E+00	0.013642116	0.00E+00	0				0		0	0		
	soild noncomb	sealed source	AU	No	1.70E+00	0.019821878		0		0				0	0		
			AS	No	8.46E-02	0.00098643		0		0				0	0		
			B	No	2.44E+00	0.028450225	1.36E-06	3.32E-06		3.32E-06					6.64E-08	3.25E-06	
			C	No	2.40E+00	0.027983828	1.11E-04	0.0002664		0.0002664					5.33E-06	0.000261072	
		cement	AU	No	7.96E+01	0.928130282	1.09E-05	0.00086764				0.00086764		0.00086764	0		
			AS	No	3.53E-02	0.000411595		0				0		0	0		
			B	No	3.22E-03	3.75E-05		0				0		0	0		
			C	No	5.13E-02	0.000598154	9.94E-02	0.00509922				0.00509922		0	0.00509922		
		solid	AS	No	5.22E+00	0.060864825		0			0			0	0		
			AU	No	5.87E+02	6.844377836	5.40E-04	0.31698			0.31698			0.253584	0.063396		
			B	No	3.00E-02	0.000349798		0			0				0	0	
			C	No	1.50E-02	0.000174899	1.67E-03	0.00002505			0.00002505				5.01E-07	0.000024549	
	Filter media	dewatered	AU	No	2.55E+01	0.297328168	1.56E-04	0.003978	0.003978					0.0031824	0.0007956		
		cement	AU	No	2.50E+02	2.914982042	1.99E-05	0.004975				0.004975		0.004975	0		
			B	No	3.02E-02	0.00035213		0				0		0	0		
		solid	AU	No	1.04E+00	0.012126325	1.60E-06	0.000001664			0.000001664			1.33E-06	3.33E-07		
	Cartrdge flter	dewatered	AU	No	4.51E+00	0.052586276	3.96E-03	0.0178596	0.0178596					0.01428768	0.00357192		
			C	No	3.62E-01	0.004220894	2.63E-02	0.0095206	0.0095206							0.0095206	
		cement	C	No	3.53E-02	0.000411595	2.99E-02	0.00105547				0.00105547		0.00105547	0		
		solid	AU	No	3.58E-01	0.004174254		0			0			0	0		
	Solidfd liqd	cement	AS	No	8.14E-02	0.000949118	1.10E+00	0.08954				0.08954		0.08954	0		
			AU	No	6.89E+01	0.803369051	2.68E-03	0.184652				0.184652		0.184652	0		
			B	No	2.37E-02	0.00027634		0				0		0	0		
			C	No	5.96E-01	0.006949317	1.37E+00	0.81652				0.81652		0	0.81652		
			AU	Yes	1.39E-01	0.00162073	2.02E-01	0.028078				0.028078		0.028078	0		
	Equipment	cement	AU	No	1.68E+00	0.019588679	5.27E-05	0.000088536				0.000088536		0.000088536	0		
		solid	AU	No	3.12E-01	0.003637898	5.33E-06	1.66E-06		1.66E-06				1.33E-06	3.33E-07		
			B	No	6.88E-02	0.000802203	1.84E-02	0.00126592		0.00126592					2.53E-05	0.001240602	

Table B-1
LLW C-14 Inventory (continued)

0.7	Fpapr solidCom	0.3	Forg resinUt	0.55	Forg resinNU	0.8	forg other	0.8	Forg filtmed	0.2								
csd paper	**csd biolog**	**csd liquid**	**csd others**	**csd cement/sol**	**csd metal**	**csl resin**	**csl paper**	**csl biolog**	**csl liquid**	**csl other**	**csl cement/sol**	**csl metal w**	**hic resins**	**hic paper**	**hic other**	**hic cement**	**hic metal w**	**hic biolog**
																0.00014523		
											0.0009882							
																	0.00034711	
					0.02258928							0.00564732						
												5.92E-05					0.002899918	
												0.154802					7.585298	
				0.0171833														
				0.104397														
				3.33E-06														
0.002189296							0.000547324											
																0.000248454		
														0.1186339	0.0508431			
0.6328896			0.2712384				0.1582224			0.0678096								
							0.000907676			0.000389004				0.04447612	0.019061196			
							0.00046662			0.00019998				0.02286438	0.00979902			
																1.67E-06		
				0.001123688							0.000280922							
													4.6719E-05					
													0.00109368					
						0.000257664												
0.00014352			0.00033488				0.00003588			0.00008372								
												6.64E-08					3.25E-06	
												5.33E-06					0.000261072	
				0.00086764														
											0.00509922							
					0.253584							0.063396						
												5.01E-07					0.000024549	
			0.0031824							0.0007956								
				0.004975														
			1.33E-06							3.33E-07								
0.01428768							0.00357192											
														0.0095206				
				0.00105547														
																0.08954		
				0.184652														
											0.81652							
				0.028078														
				0.000088536														
					1.33E-06							3.33E-07						
												2.53E-05					0.001240602	

Table B-1
LLW C-14 Inventory (continued)

subtotal	csd resinbio	csd otherbio	csl resinbio	csl otherbio	hic resinbio	hic otherbio	V in CSD	V in CSL	V in HIC	% contri	Co60 Ci/m3	C14/Co60
0.00014523					0	0	1.128	2.82E-01		0.00045008	5.82E+00	1.77E-05
0.0009882					0	0	0.144	3.60E-02		0.00306254	5.76E-01	9.53E-03
0.00034711					0	0	8.24	2.06E+00		0.00107573	4.02E+01	8.38E-07
0.0282366					0	0	0.5976	1.49E-01		0.08750842	7.91E+01	4.78E-04
0.0029591					0	0	1.864	4.66E-01		0.00917059	3.24E+03	3.92E-07
7.7401					0	0	12.56	3.14E+00		23.9874464	2.82E-02	1.75E+01
0.0171833					0	0	5.704	1.43E+00		0.05325299	6.18E-02	3.90E-02
0.104397					0	0	31.28	7.82E+00		0.32353812	1.63E-02	1.64E-01
3.33E-06					0	0	1.504	3.76E-01		1.03E-05	1.49E-03	1.19E-03
0					0	0				0	2.94E-03	0.00E+00
0.00273662					0	0				0.0084811	2.95E-01	3.17E-04
0.000248454					0	0	0.5136	1.28E-01		0.00076999	2.56E-01	1.51E-03
0.169477					0	0.04067448	27.28	6.82E+00		0.52522841	1.61E-02	3.09E-01
1.13016		0.21699072		0.05424768	0	0				3.50249381	8.09E-01	8.41E-04
0.064834				0.000311203	0	0.015248957	1.232	3.08E-01		0.20092791	1.83E+00	2.30E-02
0.03333				0.000159984	0	0.007839216	2.424	6.06E-01		0.10329344	9.92E-03	1.11E+00
1.67E-06					0	0				5.17E-06	2.20E-02	2.20E-03
0.00140461					0	0	15.76	3.94E+00		0.00435305	2.76E-03	2.58E-02
0.000046719					2.57E-05	0	0.716	1.79E-01		0.00014479	2.63E-02	1.98E-03
0.00109368					0.000601524	0				0.00338944	8.87E-03	5.68E-01
0.00128832	0.000824525		0.000206131			0	5.632	1.41E+00		0.00399265	2.04E-03	8.97E-02
0					0	0	0.03848	9.62E-03		0	2.22E-03	0.00E+00
0.000598		0.000267904		0.000066976	0	0				0.00185327		
0					0	0	0.936	2.34E-01		0	4.89E-03	0.00E+00
0					0	0	1.36	3.40E-01		0		
0					0	0				0	2.67E+02	0.00E+00
3.32E-06					0	0	1.952	4.88E-01		1.03E-05	1.55E-01	8.77E-06
0.0002664					0	0				0.0008256	1.25E-05	8.88E+00
0.00086764					0	0	63.68	1.59E+01		0.00268892		
0					0	0	0.02824	7.06E-03		0		
0					0	0	0.002576	6.44E-04		0		
0.00509922					0	0	0.04104	1.03E-02		0.01580306		
0					0	0	4.176	1.04E+00		0	3.88E-03	0.00E+00
0.31698					0	0				0.98235691		
0					0	0	0.024	6.00E-03		0		
0.00002505					0	0	0.012	3.00E-03		7.76E-05	1.33E-03	1.26E+00
0.003978		0.00063648		0.00015912	0	0	20.4	5.10E+00		0.01232827	2.11E-03	7.39E-02
0.004975					0	0	200	5.00E+01		0.01541809	1.93E-03	1.03E-02
0					0	0				0	6.58E-04	0.00E+00
0.000001664		2.66E-07		6.66E-08	0	0	0.832	2.08E-01		5.16E-06	1.43E-02	1.12E-04
0.0178596					0	0	3.608	9.02E-01		0.05534892	1.57E+00	2.52E-03
0.0095206					0	0	0.2896	7.24E-02		0.02950542	6.70E-01	3.93E-02
0.00105547					0	0	0.02824	7.06E-03		0.00327102	2.60E-03	1.15E+01
0					0	0	0.2864	7.16E-02		0		
0.08954					0	0	0.06512	1.63E-02		0.2774946	2.70E-03	4.07E+02
0.184652					0	0	55.12	1.38E+01		0.57225746		
0					0	0	0.01896	4.74E-03		0		
0.81652					0	0	0.4768	1.19E-01		2.53048794	7.21E-05	1.90E+04
0.028078					0	0	0.1112	2.78E-02		0.0870169		
0.000088536					0	0	1.344	3.36E-01		0.00027438	5.39E-03	9.78E-03
1.66E-06					0	0		6.24E-03	0.30576	5.15E-06	1.39E+02	3.83E-08
0.00126592					0	0	0.05504	1.38E-02		0.00392323	9.08E+01	2.03E-04

Table B-1
LLW C-14 Inventory (continued)

		Form	Class	Chelates	6yr avg 89–94 Ann Vol m3	Vol %	C14 Ci/m3	Ci	Ci absrbing w	Ci activted me	Ci trash	Ci solidified w	Ci inciner/sol	Ci in CsD	Ci in CsL	Ci inHIC	Fpaper DAW csd resin
			C	No	2.07E-01	0.002413605	2.50E-02	0.005175		0.005175					0.0001035	0.0050715	
	Biological	solid	AU	No	1.77E+00	0.020638073	4.87E-03	0.0086199			0.0086199			0.0086199	0		
	Inciner ash	cement	AU	No	7.13E+00	0.083135288	1.79E-03	0.0127627				0.0127627		0.0127627	0		
		solid	AU	No	6.50E+01	0.757895331	3.78E-03	0.2457					0.2457	0.2457	0		
	Air filters	solid	AU	No	1.17E+01	0.13642116	3.92E-03	0.045864			0.045864			0.0366912	0.0091728		
	Mixed DAW	solid	AS	No	4.90E+00	0.057133648		0			0			0	0		
			AU	No	2.46E+03	28.6834233	8.93E-04	2.19678			2.19678			1.757424	0.439356		
			B	No	7.22E-02	0.000841847		0			0				0	0	
			C	No	1.37E+01	0.159741016	6.98E-03	0.095626			0.095626				0.00191252	0.09371348	
		cement	AU	No	2.41E+00	0.028100427		0				0		0	0		
			AS	No	3.53E-02	0.000411595		0				0		0	0		
	Asbestos	solid	AU	No	4.25E+00	0.049554695	2.55E-05	0.000108375			0.000108375			0.0000867	0.000021675		
Government	Resin	dewatered	AS	No	5.23E+00	0.060981424	2.09E-02	0.109307	0.109307					0.0874456	0.0218614		0.0874456
			AU	No	1.29E+01	0.150413073	2.34E-04	0.0030186	0.0030186					0.00241488	0.00060372		0.00241488
			C	No	1.80E-01	0.002098787	2.25E-01	0.0405	0.0405							0.0405	
		cement	AS	No	5.37E+00	0.062613814	9.46E-02	0.508002				0.508002		0.508002	0		
			AU	No	5.18E+00	0.060398428	5.33E-04	0.00276094				0.00276094		0.00276094	0		
			AS	Yes	1.06E-01	0.001235952	1.57E-05	1.66E-06				1.66E-06		1.66E-06	0		
			Au	Yes	1.42E-01	0.00165571	3.53E-05	5.01E-06				5.01E-06		5.01E-06	0		
	Solid combustible	solid	AU	No	5.72E+00	0.066694789	8.76E-07	5.01E-06			5.01E-06			4.01E-06	1.00E-06		
	Solid noncomb	sealed source	AS	No	4.72E-04	5.50E-06		0		0				0	0		
			B	No	8.10E-01	0.009444542		0		0					0	0	
			C	No	7.47E-01	0.008709966	2.45E-05	1.83E-05		1.83E-05					3.66E-07	1.79E-05	
		solid	AU	No	9.06E+01	1.056389492	1.44E-04	0.0130464			0.0130464			0.01043712	0.00260928		
			B	No	1.66E+00	0.019355481	4.01E-06	6.66E-06			6.66E-06				1.33E-07	6.52E-06	
			C	No	1.04E-02	0.000121263		0			0				0	0	
		cement	AS	No	1.80E-01	0.002098787	3.35E-03	0.000603				0.000603		0.000603	0		
			AU	No	2.09E+00	0.02436925	1.80E-04	0.0003762				0.0003762		0.0003762	0		
	Filter media	dewatered	AS	No	1.07E+01	0.124761231	2.64E-02	0.28248	0.28248					0.225984	0.056496		
			AU	No	6.04E+00	0.070425966	8.70E-05	0.00052548	0.00052548					0.000420384	0.000105096		
			B	No	6.42E-01	0.007485674	2.28E-01	0.146376	0.146376							0.146376	
		cement	AS	No	2.47E+00	0.028800023	1.30E-01	0.3211				0.3211		0.3211	0		
			AU	No	3.42E+00	0.039876954	8.10E-04	0.0027702				0.0027702		0.0027702	0		
	Cartrdg filter	dewatered	AS	No	8.87E-01	0.010342356	2.34E-03	0.00207558	0.00207558					0.001660464	0.000415116		
			AU	No	2.71E+00	0.031598405	1.14E-04	0.00030894	0.00030894					0.000247152	0.000061788		
			C	No	1.83E-01	0.002133767	3.27E-02	0.0059841	0.0059841							0.0059841	
		cement	AS	No	1.00E+00	0.011659928	2.05E-03	0.00205				0.00205		0.00205	0		
			AU	No	2.17E-01	0.002530204	1.59E-03	0.00034503				0.00034503		0.00034503	0		
		dewatered	AU	Yes	4.72E-03	5.50E-05		0	0					0	0		
		cement	AS	Yes	3.53E-02	0.000411595	4.71E-05	1.66E-06				1.66E-06		1.66E-06	0		
		solid	AU	No	1.42E-01	0.00165571	1.18E-05	1.68E-06			1.68E-06			1.34E-06	3.35E-07		
	Solidfd liquid	cement	AU	No	1.47E+01	0.171400944	2.41E-05	0.00035427				0.00035427		0.00035427	0		
			AS	No	2.23E-01	0.002600164	1.19E-02	0.0026537				0.0026537		0.0026537	0		
			AS	Yes	9.32E-01	0.010867053	1.79E-06	1.67E-06				1.67E-06		1.67E-06	0		
			AU	Yes	2.08E-01	0.002425265	2.41E-05	5.01E-06				5.01E-06		5.01E-06	0		
			B	No	1.42E-02	0.000165571	1.18E-04	1.68E-06				1.68E-06		0	1.68E-06		
		solid	AU	No	1.77E-01	0.002063807	1.04E-03	0.00018408			0.00018408			0.000147264	0.000036816		
	Equipment	solid	AU	No	7.53E+00	0.087799259	4.74E-04	0.00356922		0.00356922				0.002855376	0.000713844		
			B	No	9.43E-04	1.10E-05		0		0				0	0		
		cement	AU	No	3.89E-01	0.004535712	8.78E-04	0.000341542				0.000341542		0.000273234	6.83E-05		
		sealed source	AU	No	1.14E-01	0.001329232		0		0				0	0		
			B	No	3.07E-02	0.00035796		0		0				0	0		

Table B-1
LLW C-14 Inventory (continued)

0.7	Fpapr solidCom	0.3	Forg resinUt	0.55	Forg resinNU	0.8	forg other	0.8	Forg filtmed	0.2									
csd paper	**csd biolog**	**csd liquid**	**csd others**	**csd cement/sol**	**csd metal**	**csl resin**	**csl paper**	**csl biolog**	**csl liquid**	**csl other**	**csl cement/sol**	**csl metal w**	**hic resins**	**hic paper**	**hic other**	**hic cement**	**hic metal w**	**hic biolog**	
												0.0001035					0.0050715		
	0.0086199																		
				0.0127627															
				0.2457															
0.0366912							0.0091728												
1.2301968			0.5272272				0.3075492			0.1318068									
							0.001338764			0.000573756				0.06559944	0.028114044				
																0.000108375			
						0.0218614													
						0.00060372													
													0.0405						
																0.508002			
				0.00276094															
																1.66E-06			
				5.01E-06															
1.20E-06			2.81E-06				3.01E-07			7.02E-07									
												3.66E-07					1.79E-05		
					0.01043712							0.00260928							
												1.33E-07					6.52E-06		
																0.000603			
				0.0003762															
															0.28248				
			0.000420384							0.000105096									
															0.146376				
																0.3211			
				0.0027702															
														0.00207558					
0.000247152							0.000061788												
														0.0059841					
																0.00205			
				0.00034503															
																1.66E-06			
1.34E-06							3.35E-07												
				0.00035427															
																0.0026537			
																1.67E-06			
				5.01E-06															
											1.68E-06								
		0.000147264							0.000036816										
					0.002855376							0.000713844							
				0.000273234							6.83E-05								

Table B-1
LLW C-14 Inventory (continued)

subtotal	csd resinbio	csd otherbio	csl resinbio	csl otherbio	hic resinbio	hic otherbio	V in CSD	V in CSL	V in HIC	% contri	Co60 Ci/m3	C14/Co60
0.005175					0	0	0.1656	4.14E-02		0.01603791		
0.0086199					0	0	1.416	3.54E-01		0.02671405	1.98E-02	2.46E-01
0.0127627					0	0	5.704	1.43E+00		0.03955305	1.07E-02	1.67E-01
0.2457					0	0	52	1.30E+01		0.76145212	3.59E-03	1.05E+00
0.045864					0	0	9.36	2.34E+00		0.14213773		
0					0	0				0	2.72E-03	0.00E+00
2.19678		0.42178176		0.10544544	0	0				6.80806997		
0					0	0	0.05776	1.44E-02		0		
0.095626				0.000459005	0	0.022491235		2.74E-01	13.426	0.2963558	7.36E-03	9.48E-01
0					0	0	1.928	4.82E-01		0		
0					0	0	0.02824	7.06E-03		0	3.38E-03	0.00E+00
0.000108375					0	0	3.4	8.50E-01		0.00033587	1.05E+00	2.43E-05
0.109307	0.04809508		0.01202377		0	0	4.184	1.05E+00		0.33875477	1.00E-02	2.09E+00
0.0030186	0.001328184		0.000332046		0	0	10.32	2.58E+00		0.00935498	1.07E+01	2.19E-05
0.0405					0.022275	0	0.144	3.60E-02		0.12551409	4.61E+00	4.88E-02
0.508002					0	0	4.296	1.07E+00		1.57435572	2.53E-02	3.74E+00
0.00276094					0	0	4.144	1.04E+00		0.00855647	4.55E-04	1.17E+00
1.66E-06					0	0	0.0848	2.12E-02		5.16E-06	1.47E-03	1.07E-02
5.01E-06					0	0	0.1136	2.84E-02		1.55E-05	3.30E-05	1.07E+00
5.01E-06		2.24E-06		5.61E-07	0	0	4.576	1.14E+00		1.55E-05		
0					0	0	0.000378	9.44E-05		0		
0					0	0	0.648	1.62E-01		0	1.40E+01	0.00E+00
1.83E-05					0	0	0.5976	1.49E-01		5.67E-05	6.89E-02	3.56E-04
0.0130464					0	0	72.48	1.81E+01		0.04043227	1.03E-03	1.40E-01
6.66E-06					0	0	1.328	3.32E-01		2.06E-05	1.00E-06	4.01E+00
0					0	0	0.00832	2.08E-03		0		
0.000603					0	0	0.144	3.60E-02		0.00186877	2.23E-01	1.50E-02
0.0003762					0	0				0.00116589	7.92E-03	2.27E-02
0.28248					0	0.225984	8.56	2.14E+00		0.87543751	1.31E+00	2.02E-02
0.00052548		8.41E-05		2.10E-05	0	0	4.832	1.21E+00		0.00162852	4.11E-03	2.12E-02
0.146376					0	0.1171008	0.5136	1.28E-01		0.4536358	1.52E+01	1.50E-02
0.3211					0	0	1.976	4.94E-01		0.99512526	6.35E+00	2.05E-02
0.0027702					0	0				0.00858516	3.66E-02	2.21E-02
0.00207558					0	0	0.7096	1.77E-01		0.00643246	1.17E-01	2.00E-02
0.00030894					0	0	2.168	5.42E-01		0.00095744	5.29E-03	2.16E-02
0.0059841					0	0				0.0185454	1.48E+00	2.21E-02
0.00205					0	0				0.00635318	7.95E-02	2.58E-02
0.00034503					0	0	0.1736	4.34E-02		0.00106929	6.01E-02	2.65E-02
0					0	0	0.003776	9.44E-04		0		
1.66E-06					0	0	0.02824	7.06E-03		5.15E-06	2.59E-03	1.82E-02
1.68E-06					0	0	0.1136	2.84E-02		5.19E-06	1.20E-03	9.83E-03
0.00035427					0	0	11.76	2.94E+00		0.00109792	1.67E-03	1.44E-02
0.0026537					0	0				0.00822412	5.74E-01	2.07E-02
1.67E-06					0	0	0.7456	1.86E-01		5.17E-06	1.14E-04	1.57E-02
5.01E-06					0	0	0.1664	4.16E-02		1.55E-05	1.85E-03	1.30E-02
1.68E-06					0	0	0.01136	2.84E-03		5.19E-06		
0.00018408					0	0				0.00057049	4.54E-02	2.29E-02
0.00356922					0	0	6.024	1.51E+00		0.01106142	4.22E-02	1.12E-02
0					0	0				0		
0.000341542					0	0				0.00105848	4.08E-02	2.15E-02
0					0	0	0.0912	2.28E-02		0		
0					0	0	0.02456	6.14E-03		0		

Table B-1
LLW C-14 Inventory (continued)

		Form	Class	Chelates	6yr avg 89–94 Ann Vol m3	Vol %	C14 Ci/m3	Ci	Ci absrbing w	Ci activted me	Ci trash	Ci solidified w	Ci inciner/sol	Ci in CsD	Ci in CsL	Ci inHIC	Fpaper DAW csd resin
	Biological	solid	AU	No	1.17E+00	0.013642116	2.13E-03	0.0024921			0.0024921			0.00199368	0.00049842		
	Inciner ash	solid	AU	No	5.04E-01	0.005876604	5.04E-04	0.000254016					0.000254016	0.000203213	5.08E-05		
	Air filters	solid	AU	No	3.02E+00	0.035212983	3.31E-06	1.00E-05			1.00E-05			8.00E-06	2.00E-06		
	Mixed DAW	solid	AS	No	1.64E+01	0.191222822	3.72E-04	0.0061008			0.0061008			0.00488064	0.00122016		
			AU	No	9.72E+02	11.33345018	2.22E-04	0.215784			0.215784			0.1726272	0.0431568		
			B	No	1.19E-01	0.001387531	1.40E-03	0.0001666			0.0001666				3.33E-06	0.000163268	
			C	No	1.65E-02	0.000192389	1.21E+00	0.019965			0.019965				0.0003993	0.0195657	
		cement	AS	No	5.85E-01	0.006821058	8.79E-04	0.000514215				0.000514215		0.000514215	0		
			AU	No	9.58E+00	0.111702112	5.91E-06	5.66E-05				5.66E-05		5.66E-05	0		
		solid	AS	Yes	2.53E+00	0.029499618	7.23E-06	1.83E-05			1.83E-05			1.46E-05	3.66E-06		
	Asbestos	solid	AU	No	3.57E+01	0.416259436	4.29E-06	0.000153153			0.000153153			0.000122522	3.06E-05		
Academic	Resin	dewatered	AU	No	2.48E-01	0.002891662		0	0					0	0		
	solid noncomb	sealed source	AU	No	8.33E-02	0.000971272	2.00E-05	0.000001666		0.000001666				1.33E-06	3.33E-07		
			B	No	1.05E-01	0.001224292	0.00E+00	0		0					0	0	
			C	No	4.70E-01	0.005480166	4.18E-04	0.00019646		0.00019646					3.93E-06	0.000192531	
		solid	AU	No	4.77E-01	0.005561786	3.50E-06	1.67E-06			1.67E-06			1.34E-06	3.34E-07		
		cement	AS	No	3.22E-03	3.75E-05		0				0		0	0		
			AU	No	7.36E-03	8.58E-05		0				0		0	0		
	Filter media	cement	AU	No	5.10E-02	0.000594656		0				0		0	0		
		dewatered	AU	No	2.83E-02	0.000329976		0	0					0	0		
	Solidfd liquid	cement	AS	No	9.00E-02	0.001049394	3.37E-03	0.0003033				0.0003033		0.0003033	0		
			AU	No	6.29E+00	0.073340948	3.02E-03	0.0189958				0.0189958		0.0189958	0		
			B	No	7.08E-02	0.000825523		0				0		0	0		
			C	No	2.26E-02	0.000263514		0				0		0	0		
		solid	AU	No	6.85E-01	0.007987051	3.92E-03	0.0026852			0.0026852			0.0026852	0		
	Biological	solid	AU	No	2.04E+01	0.237862535	1.63E-03	0.033252			0.033252			0.033252	0		
			AS	No	5.23E-02	0.000609814	3.88E-03	0.000202924			0.000202924			0.000162339	4.06E-05		
	Inciner ash	solid	AU	No	3.43E-01	0.003999355	9.92E-04	0.000340256					0.000340256	0.000272205	6.81E-05		
	Air filters	solid	AU	No	9.43E-03	0.000109953		0			0			0	0		
	Mixed DAW	gas	B	No	1.32E-01	0.001539111		0		0				0	0		
	Mixed DAW	solid	AU	No	8.74E+01	1.019077722	4.99E-03	0.436126			0.436126			0.3489008	0.0872252		
Medical	solid noncomb	sealed source	AU	No	8.22E-02	0.000958446		0		0				0	0		
			AS	No	3.53E-02	0.000411595		0		0				0	0		
			B	No	1.27E-01	0.001480811		0		0					0	0	
			C	No	1.16E-01	0.001352552		0		0					0	0	
		cement	AU	No	2.46E-02	0.000286834		0				0		0	0		
		solid	AU	No	5.47E-02	0.000637798	3.04E-05	1.66E-06			1.66E-06			1.33E-06	3.33E-07		
	soldfd lquid	cement	AU	No	6.46E-01	0.007532314	1.83E-03	0.00118218				0.00118218		0.00118218	0		
			C	No	1.46E-02	0.000170235		0				0		0	0		
		solid	AU	No	5.43E-02	0.000633134		0			0			0	0		
	Biological	solid	AU	No	3.95E+00	0.046056716	6.13E-04	0.00242135			0.00242135			0.00193708	0.00048427		
	Inciner ash	Cement	AU	No	1.77E-02	0.000206381		0				0		0	0		
		solid	AU	No	1.50E-01	0.001748989	1.57E-03	0.0002355					0.0002355	0.0001884	0.0000471		
	Mixed DAW	solid	AU	No	1.04E+01	0.121263253	2.00E-03	0.0208			0.0208			0.01664	0.00416		
				tot V	8.58E+03		Subtotal Ci	32.26729468	16.92747965	7.785440759	4.882824009	2.320623487	0.350926772	6.882602937	2.247092539	23.1375992	1.050830048

Table B-1
LLW C-14 Inventory (continued)

0.7	Fpapr solidCom	0.3	Forg resinUt	0.55	Forg resinNU	0.8	forg other	0.8	Forg filtmed	0.2								
csd paper	csd biolog	csd liquid	csd others	csd cement/sol	csd metal	csl resin	csl paper	csl biolog	csl liquid	csl other	csl cement/sol	csl metal w	hic resins	hic paper	hic other	hic cement	hic metal w	hic biolog
	0.00199368							0.00049842										
				0.000203213							5.08E-05							
8.00E-06							2.00E-06											
														0.00427056	0.00183024			
0.12083904			0.05178816				0.03020976			0.01294704								
							2.33E-06			1.00E-06				0.00011429	4.90E-05			
							0.00027951			0.00011979				0.01369599	0.00586971			
																0.000514215		
				5.66E-05														
														1.28E-05	5.49E-06			
				0.000122522							3.06E-05							
					1.33E-06							3.33E-07						
												3.93E-06					0.000192531	
					1.34E-06							3.34E-07						
																0.0003033		
				0.0189958														
		0.0026852																
	0.033252																	
																		0.00020292
				0.000272205							6.81E-05							
0.24423056			0.10467024				0.06105764			0.02616756								
					1.33E-06							3.33E-07						
				0.00118218														
	0.00193708							0.00048427										
				0.0001884							0.0000471		0					
0.011648			0.004992				0.002912			0.001248								
2.294638988	0.04580266	0.002832464	0.994898623	0.71333805	0.289480445	0.262707512	0.576654649	0.00098269	0.000036816	0.250697316	0.834626985	0.227437101	13.9265904	1.10319496	0.967657197	1.125243005	7.599441842	0.00020292

Table B-1
LLW C-14 Inventory (continued)

subtotal	csd resinbio	csd otherbio	csl resinbio	csl otherbio	hic resinbio	hic otherbio	V in CSD	V in CSL	V in HIC	% contri	Co60 Ci/m3	C14/Co60
0.0024921					0	0	0.936	2.34E-01		0.0077233		
0.000254016					0	0				0.00078722	2.38E-02	2.12E-02
1.00E-05					0	0				3.10E-05	2.66E-04	1.24E-02
0.0061008					0	0.001464192	13.12	3.28E+00		0.01890707	1.73E-02	2.15E-02
0.215784		0.041430528		0.010357632	0	0				0.66873905	8.04E-04	2.76E-01
0.0001666				8.00E-07	0	3.92E-05	0.0952	2.38E-02		0.00051631		
0.019965				9.58E-05	0	0.004695768	0.0132	3.30E-03		0.0618738		
0.000514215					0	0	0.468	1.17E-01		0.00159361	4.39E-02	2.00E-02
5.66E-05					0	0	7.664	1.92E+00		0.00017547	5.94E-04	9.95E-03
1.83E-05					0	4.39E-06	2.024	5.06E-01		5.67E-05	4.22E-04	1.71E-02
0.000153153					0	0				0.00047464	1.83E-04	2.34E-02
0					0	0	0.1984	4.96E-02		0	7.06E-04	0.00E+00
0.000001666					0	0	0.06664	1.67E-02		5.16E-06	2.96E+00	6.76E-06
0					0	0	0.084	2.10E-02		0	3.06E+01	0.00E+00
0.00019646					0	0				0.00060885	1.05E+00	3.98E-04
1.67E-06					0	0				5.17E-06		
0					0	0				0		
0					0	0				0		
0					0	0				0	2.29E-04	0.00E+00
0					0	0				0	2.94E-02	0.00E+00
0.0003033					0	0				0.00093996		
0.0189958					0	0				0.05887014	9.72E-04	3.11E+00
0					0	0				0	8.95E+00	0.00E+00
0					0	0				0	6.57E+00	0.00E+00
0.0026852					0	0				0.00832174		
0.033252					0	0				0.10305171		
0					0	0				0.00062888		
0.000340256					0	0				0.00105449		
0					0	0				0		
0					0	0				0		
0.436126		0.083736192		0.020934048	0	0				1.35160386	4.71E-04	1.06E+01
0					0	0				0	3.45E-04	0.00E+00
0					0	0				0		
0					0	0				0		
0					0	0				0	7.91E-04	0.00E+00
0					0	0				0		
1.66E-06					0	0				5.15E-06		
0.00118218					0	0				0.00366371		
0					0	0				0		
0					0	0				0		
0.00242135					0	0				0.00750404		
0					0	0				0		
0.0002355					0	0				0.00072984		
0.0208				0.0009984	0	0				0.06446156	1.46E-05	1.37E+02
32.26709175	0.57821419	0.784768416	0.144553548	0.198769165	7.659624719	0.659874958	1358.062	338.0811	981.3552	100		136.9863014
							0.507213	0.126268	0.366519	2677.49821		

C
MECHANISTIC MODELS

C.1 Modeling Radionuclide Release from Spent Ion-Exchange Resins

C.1.1 Introduction

Source-Term is the phrase that is used to refer to the prediction of the release of radionuclides from the engineered boundaries of a low level waste disposal.

Predicted results of source term releases directly dictate the subsequent analysis of geosphere and biosphere transport in performance assessment. Nonetheless, source term analysis remains the most uncertain step and one of the biggest contributors to the conservatism in an overall performance assessment. The uncertainty is attributed to inadequacies in our understanding, limitations in current modeling approaches and uncertainties of the parameters involved in the models used in the absence of defendable, mechanistic understanding of the phenomena involved, conservative approaches in the analysis must be applied. This analysis proposes a more mechanistic approach to the prediction of releases from ion-exchange resin wastes, which, as described further in this report, contain a large quantity of significant radionuclides.

As currently practiced, LLWs are disposed in a wide variety of materials and forms, including metals, resins, filters, mixed trash, cement, glass, or sorbent media. The waste form into which the radionuclides are incorporated has a considerable impact on the manner and degree to which they are released or retained in the waste form. The degree of uncertainty or scientific understanding in the analysis of radionuclide release varies among different waste materials. There exists a significant body of work into the mechanistic understanding of chemical durability and leaching characteristics of glass, cement, metals or other solidified waste. The unsolidified waste streams, in contrast, remain as being very little studied.

Among various types of unsolidified LLW such as ion-exchange (IX) resins, filters, dry active waste, and incinerator ash, IX resins contain the largest inventory of radioactivity of long-lived, potentially environmentally mobile radionuclides. According to the shipping manifest information collected at Barnwell, South Carolina, LLW site[1], the estimated inventory of radioactivity contained in unsolidified IX resins is 47% of the total disposed activity for ^{14}C, 59% for ^{90}Sr, 32% for ^{129}I, 81% for ^{137}Cs, and 98% for ^{237}Np. Even though regeneration of IX resins was practiced in the early history of nuclear power plant operations, the used, or “spent” IX resin beds are now commonly discarded at the time of refueling outage.

For disposal purposes, spent IX resins used to be immobilized into cement. However, problems were encountered in this processing. When IX resins are placed in wet, alkaline cement, resins absorb water and undergoe chemical reactions that result in swelling [2Swelling results in tensile stresses in the encapsulating cement causing it to crack and expose the resin beads to further environmental attack.. Current practice is to pretreat IX resins by a dehydration/dewatering process and store them in high integrity containers (HICs) for disposal. This is likely to continue until the development of a better immobilization technique.

Release of radionuclides from IX resins is commonly modeled by a simplistic, conservative model known as "surface wash release". In this approach, radionuclide release is assumed to occur instantaneously upon contact by water. The only limits to this release are the solubility limit of the particular chemical form of the various radionuclides, and physical capture of the material in the surrounding materials. In some cases, the solubility limit is very high. The influence of the solubility and the "stickiness" of the surrounding material is represented by a combined term known as the distribution coefficient. The distribution coefficient (K_d) represents the state of equilibrium within the system. Typically very conservative (i.e., low) values of K_d are selected for performance assessment.

In reality, the distribution of activity within bulk resin medium between the solid phase and liquid phase is expected to be dependent on whether the radionuclide "attachment" to the resin matrix is more surface-controlled, ion-exchange-controlled, or filtration-controlled. Also, even with the accurate determination of how much activity is partitioned into the liquid phase, actual release of radionuclides from the waste package will depend on the diffusional transport characteristics within the waste-form, waste package porosity.

This study examines these issues with a goal of better mechanistic understanding and modeling of radionuclide release from IX resins. Mechanisms of radionuclide release from IX resins were examined and a supporting mathematical/computer model was developed.

C.1.2 Background on the Release of Radionuclides from Ion-Exchange Resins

[comment: this first paragraph could benefit from a better articulated figure (than Figure C-1) that illustrates the phenomena being described. The terminology is not descriptive enough for the typical reader]

IX resins are three dimensionally crosslinked hydrocarbon chain material consisting of an irregular macromolecule of alkyl groups which also carries fixed ionic groups within the matrix. The charge of the group is balanced by mobile counter ions making them neutral polyelectrolyte material. These counter ions can be exchanged for an equivalent amount of other ions of the same sign when the ion-exchanger is in contact with an electrolyte solution [3]. The exchange is a reversible stoichiometric process. This stoichiometric redistribution of counterions between the ion exchanger and the contacting liquid phase is largely responsible both for decontamination capability and predicted mechanistics of radionuclide release after disposal.

One way the characteristics of ion exchange can be determined is by a fluid-solid reaction which produces fluid products. The five basic steps outline the exchange reaction between fluid and particles, also referred to a mass transfer [4]. Figure C-1 illustrates the process of mass transfer of ions between bulk liquid and resins.

step 1. Fluid reactant diffuses through the film surrounding the resin solid to the resin surface, i.e., particle *B* diffuses along the dashed arrow to the resin surface.

step 2. Fluid reactant diffuses inside the resin matrix, i.e., circle *B* diffuses along the bold solid arrow inside the resin to the counterion site.

step 3. Fluid reactant exchanges the counterion species *A* inside the resin ($^{\cdot}R$), i.e., chemical reaction between fluid reactant *B* and counterion *A* as follows.

$$B(re\,\mathrm{sin}) + A?R \cap A(re\,\mathrm{sin}) + B?R$$

step 4. The counterion species diffuses through the resin matrix to the solid surface, i.e., circle *A* diffuses along the bold solid arrow inside the resin particle to the resin surface.

step 5. The counterion species from the resin surface diffuses through the liquid film into the bulk fluid, i.e., particle *A* diffuses along the dashed arrow to the bulk solution.

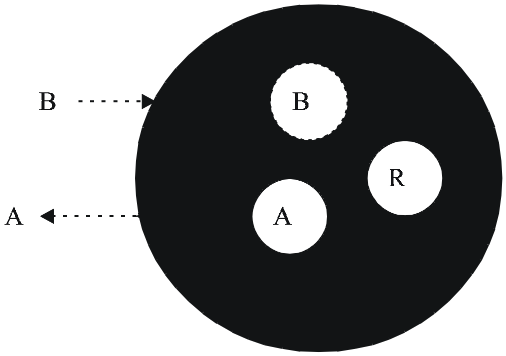


Figure C-1
Illustration of Ion Exchange

In order for an exchange of counterions *A* (carried by resins) and *B* (from liquid phase) to take place, species *B* must migrate from the solution into the interior of the ion exchanger, and species *A* must migrate from the exchanger into the solution. For the chemical reaction in step 3, its reaction rate is commonly expressed by the first order reaction rate:

$$\frac{dA_{(re\,\mathrm{sin})}}{dt} = k_1 ?\left[B_{(re\,\mathrm{sin})}\right]?[A?R] \qquad \textbf{Eq. C-1}$$

where k_1 is a reaction constant.

The rate controlling step in the entire ion exchange process could be the chemical reaction if the concentration of fluid reactant *B* into resins is not enough for the ion exchange to occur or the diffusion if [$B_{(\mathrm{re}\ sin)}$] is abundant. Also, the total amount of counterion *A* available to be released into the solution is a key factor in determining the leaching amount from the containers.

Diffusion of exchanging counter ions within ion-exchangers is believed to be slower than in solutions due to the existence of three dimensional framework and tortuous path length in the narrow-mesh regions. Due to the effects of electric field generated by ionic diffusion, the net flux of the faster and slower ions is balanced to maintain electric neutrality. This is described by the Nernst-Planck theory [5] where the resulting electric transference is superimposed on ordinary diffusion driven by the concentration gradient [6]. The Nernst-Planck equation is

$$\bar{J}_A = -\ \bar{D}_A\ grad\bar{C}_A - \bar{D}_A\bar{C}_A\ \frac{z_A F}{GT}\ \underset{\hookleftarrow}{\surd}\ grad\varphi \qquad \textbf{Eq. C-2}$$

where $\bar{C}_A$ = concentration of species A (moles/cm^3)

$\bar{J}_A$ = flux of species A (moles/cm$^{2\cdot}$sec)

$\bar{D}_A$ = diffusion coefficients of species A (cm^2/sec)

F = Faraday constant (coulombs/mole)

G = gas constant (ergs/mole$^{\cdot}$ (deg)

T = absolute temperature (deg)

z_A = valence of species A (negative for anions)

φ = electric potential (ergs/coulomb)

The Nernst-Planck model can be also substituted by Fick's law when the resins behave with weak functionality [7]. Rinse release could also be responsible for the release of radionuclides from IX resins if the activity is retained through surface phenomena.

C.1.3 Description of the Model

A hypothetical system of IX resins is used to simulate the seepage of radionuclides from IX resins in a waste container with infiltrating ground water flow. This system is shown in Figure C-2.

As shown in Figure C-2, the resin system consists of the solid resins (phase I) and the bulk pore liquid (phase II). It is assumed that the infiltrating water enters the top of a container, becomes perfectly mixed with the container volume, and leaves the bottom of the container.

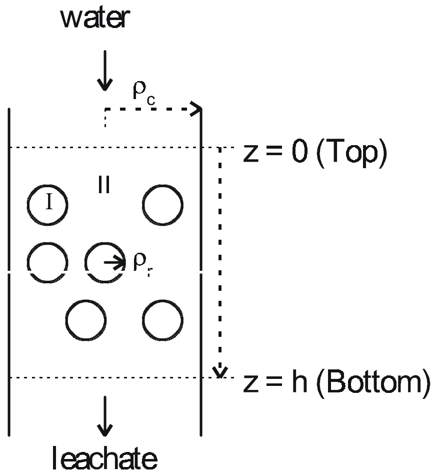


Figure C-2
Simulated Resin System in a Waste Container (Phases I and II stand for the resin solid, and the liquid contiguous to resins, respectively)

The two steps involved in the release of radionuclide from waste containers are:

1. transport and release of radionuclide from resin beads to the bulk pore water.
2. the movement of radionuclide in the bulk pore water within waste containers until they leave the container

The first step requires knowledge of the effects of ionic diffusion; the action of surface rinsing on the physical adherence, and any chemical reactions that may be involved. Movement is affected by the combined effects of diffusion in the bulk pore water, advective transport through the system, sorption during the migration and radioactive decay.

C.1.3.1 Modeling of Resin System

Resin beads are modeled in spherical coordinates with radius ρ_r (cm). The bulk pore water within the waste container is in a cylindrical coordinate with radius ρ_c (cm) and height h (cm). The symbols of coordinates hereafter used for this resin system include:

1. r-coordinate for the radial direction in the resin bead;
2. ρ-coordinate for the radial direction in cylindrical containers; and
3. z-coordinate for the downward direction in cylindrical containers.

The key assumptions made in modeling the release of radionuclides from the resin system include the following:

1. The resin bead is isotropic and spherical with constant diameter.
2. The ion exchange process in resin beads is isothermal.

3. The chemical reaction among ions within resins reaches equilibrium quickly.

4. The individual diffusivity of ions in the resin phase is constant.

5. Diffusion coefficients and retardation coefficients are constant in the whole resin system.

6. Local equilibrium of each reaction is maintained at all times and all locations in the bulk pore liquid within the container.

7. Equilibrium between liquid and resins is reached quickly as reaction begins. [comment: is this redundant with (3) ?]

8. The radionuclide concentration distributes symmetrically in the polar direction of cylindrical containers.

9. Water flow through the container is in a net downward *z*-direction.

10. The resin system is fully saturated with water.

11. The distribution of radionuclide concentration within resins is identical for all resin beads in the same container.

12. A linear isotherm partition concept is used to describe the sorption effect. [comment: ????]

13. No chemical reactions (i.e., adsorption) take place on the container walls.

[comment: summarize, conservative, mechanistic, requires supporting data.]

Equations for the radionuclide transport in the resin system can be formulated as follows:

<u>Phase I (Resin phase)</u>:

$$\frac{fC_1}{ft} = \frac{\overline{D}_i}{r^2}\frac{f}{fr}\left(r^2\frac{fC_1}{fr}\right) - \lambda C_1 + S_1 \qquad \textbf{Eq. C-3}$$

<u>Phase II (Bulk pore water phase)</u>:

$$\frac{fC_2}{ft} = \frac{D_2}{R}\frac{1}{\rho}\frac{f}{f\rho}\overline{\rho}\frac{fC_2}{f\rho}\surd_{\hookleftarrow} + \frac{D_2}{R}\frac{f^2C_2}{fz^2} - \frac{v}{R}\frac{fC_2}{fz} - \lambda C_2 + S_2 \qquad \textbf{Eq. C-4}$$

where C_1 and C_2: concentrations of radionuclide in phases I and II (mole/cm^3)

$\overline{D}_i$: interdiffusion coefficient in resins (cm^2/sec)= $\frac{\overline{D}_A\overline{D}_B(z_A^2\overline{C}_A + z_A^2\overline{C}_A)}{z_A^2\overline{C}_A\overline{D}_A + z_A^2\overline{C}_A\overline{D}_B}$

(where, subscript B represents the counter ion in the solution. The others are defined the same as in Equation (C-2))

D_2: diffusion coefficient in solution (cm^2/sec)

R: retardation coefficient in solution

v : flow velocity (cm/sec)

λ: decay constant of radionuclide (sec^{-1})

S_1: source term in phase I = $\underset{i}{\;}k_i C_{c,i}[I]$ (mole/cm^{3}sec)

$C_{c,i}$: concentration of counter ion species (mole/cm^3)

I: concentration of available ion-exchange sites (sites/cm^3)

k_i: reaction constant for counter-ion i (cm^3/sec)

S_2: source term in phase II, release from resins (mole/cm^{3}sec)

The retardation coefficient, R, is represented by $R = 1 + \frac{K_d \rho_b}{\theta}$, where K_d is the distribution coefficient, ρ_b is the bulk density, and θ is the moisture content.

The radioactivity of resins available for surface rinse release is initially assumed to be uniformly distributed in the solution phase. The amount of radionuclide retained in resins is initially homogeneous. Therefore the initial conditions in both phases can be expressed as:

$$C_1(r, \rho, z, t = 0) = \frac{(1 - f_0)wC_0}{V_I} \qquad \textbf{Eq. C-5}$$

$$C_2(\rho, z, t = 0) = \frac{f_0 w C_0}{V_{II}\varepsilon} \qquad \textbf{Eq. C-6}$$

where C_o: the concentration of radionuclide in resins before leaching (Ci/g of resins)

ε: porosity in phase II

w: total resin weight (g)

V_I: total volume of phase I (cm^3)

V_{II}: total volume of phase II (cm^3)

f_o: the fraction of surface rinse

Boundary conditions in this model are considered as follows.

1. The concentration distribution is symmetric at the center (*r* = 0) in phase I which means the concentration distribution is angular-independent.

$$-\overline{D}_i \frac{f\,C_1}{f\,r}\bigg|_{r=0} = 0 \qquad \textbf{Eq. C-7}$$

2. The radionuclide flux at the interface between phases I and II obeys the continuity law. It can be conceptually expressed as:

$$\text{radionuclide flux from phase I to II} = -\overline{D}_i \frac{f\,C_1}{f\,r}\bigg|_{r=\rho_r} \qquad \textbf{Eq. C-8}$$

3. The concentration distribution in phase II is symmetric at the center (ρ = 0) of the radial container plane.

$$-\,D_2 \frac{f\,C_2}{f\,\rho}\bigg|_{\rho=0} = 0 \qquad \textbf{Eq. C-9}$$

4. There is no concentration flux across the radial boundary ($\rho = \rho_c$) of phase II.

$$-\,D_2 \frac{f\,C_2}{f\rho}\bigg|_{\rho=\rho_c} = 0 \qquad \textbf{Eq. C-10}$$

5. The continuity of radionuclide flux applies at the top of the container.

$$-\frac{D_2}{R}\frac{f\,C_2(x,z,t)}{f\,z}\bigg|_{z=0^+} + \frac{vD_2}{R}\bigg|_{z=0^+} = -\frac{D_2}{R}\frac{f\,C_2(x,z,t)}{f\,z}\bigg|_{z=0^-} + \frac{vD_2}{R}\bigg|_{z=0^-} \qquad \textbf{Eq. C-11}$$

6. The continuity of radionuclide flux applies at the bottom of the container.

$$-\frac{D_2}{R}\frac{f\,C_2(x,z,t)}{f\,z}\bigg|_{z=h+0^+} + \frac{vD_2}{R}\bigg|_{z=h+0^+} = -\frac{D_2}{R}\frac{f\,C_2(x,z,t)}{f\,z}\bigg|_{z=h+0^-} + \frac{vD_2}{R}\bigg|_{z=h+0^-} \qquad \textbf{Eq. C-12}$$

C.1.3.2 Numerical Solution Technique

For equations (C-3) and (C-4), numerical solution approach is employed in this study due to the limitations with the analytical solution approach.

In the numerical approach, spherical resin beads are uniformly divided into N_r grids in radial directions. The cylindrical container is designed with N_z and N_ρ divisions in vertical and radial directions. The divisions of the resin bead and the container are schematically shown in

Figures C-3 and C-4, respectively. Except for natural radioactive decay, the amount of radionuclide disappearing from resins is assumed to enter the liquid phase adjacent to the resins. Similarly, radionuclides leave the container either through radioactive decay or by leaving the container from the bottom. Hence, the amount of radionuclide release from the resin container can be determined by a massbalance.

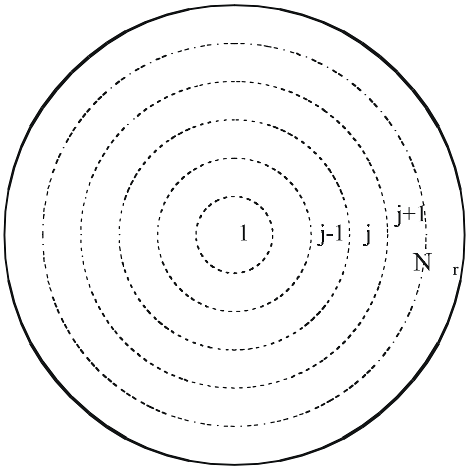


Figure C-3
Resin Grids in Numerical Approach

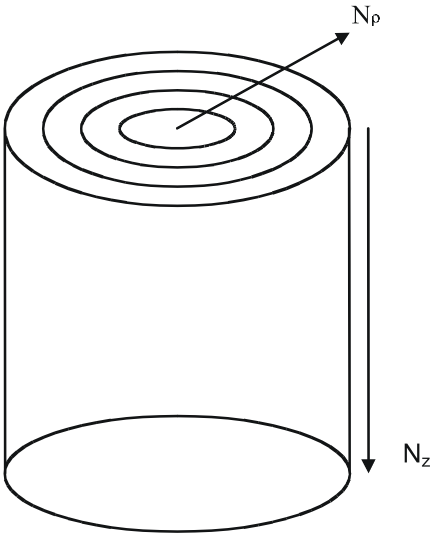


Figure C-4
Container Nodes in Numerical Approach

The partial differential equation of resin beads from (3) is rewritten as:

$$\frac{\partial C_1}{\partial t} = \overline{D}_i \frac{\partial^2 C_1}{\partial r^2} + \frac{2\overline{D}_i}{r} \frac{\partial C_1}{\partial r} - \lambda C_1 + S_1$$ **Eq. C-13**

The finite difference approach used here incorporates the Crank-Nicholson scheme in time coordinates and the central difference scheme in spatial coordinates. The symbols for time interval, radial grid length, and radius at grid j are Δt, Δr, and r_j, respectively. The governing finite difference equation can be expressed as:

$$
\begin{aligned}
\frac{C_{1_j}^{n+1} - C_{1_j}^{n}}{\Delta t} = {} & \frac{1}{2}\overline{D}_i \frac{C_{1_{j+1}} - 2C_{1_j} + C_{1_{j-1}}}{\Delta r^2} + \overline{D}_i \frac{C_{1_{j+1}} - C_{1_{j-1}}}{r_j \Delta r} - \lambda C_{1_j}{}^{n+1} \\
& + \frac{1}{2}\overline{D}_i \frac{C_{1_{j+1}} - 2C_{1_j} + C_{1_{j-1}}}{\Delta r^2} + \overline{D}_i \frac{C_{1_{j+1}} - C_{1_{j-1}}}{r_j \Delta r} - \lambda C_{1_j}{}^{n} \\
& + \frac{1}{2}\left(S_{1_j}^{n} + S_{1_j}^{n+1}\right)
\end{aligned}
\qquad \textbf{Eq. C-14}
$$

where n and n+1 represent the current time and the next time steps, respectively
j stands for the number of grids in phase I (1 through N_r)

$$\Delta r = \frac{\rho_r}{N_r}$$

$$r_j = \left(j - \frac{1}{2}\right) ?\, \Delta r$$

This implicit finite difference equation of phase I can be arranged into the following matrix equation.

$$\overline{R}\,\overline{L} \leftrightarrow \overline{C}_1^{n+1} = \overline{R}\,\overline{R} \leftrightarrow \overline{C}_1^{n} + \overline{R}\,\overline{S} \qquad \textbf{Eq. C-15}$$

where $\overline{R}\,\overline{R}$ = coefficient matrix for the current concentration distribution

$\overline{R}\,\overline{L}$ = coefficient matrix for the concentration distribution of the next time step

$\overline{C}_1^{n}$ = matrix of the current concentration distribution in phase I

$\overline{C}_1^{n+1}$ = matrix of the concentration distribution of the next time step in phase I

$\overline{R}\,\overline{S}$ = source term matrix in phase I

Using mass balance, the radionuclide release from resin to solution for the next step, $S_{2_j}^{n+1}$, can be determined by the difference of total radionuclide amount retained in resins between two consecutive time steps corrected by decay effect. The source term for the liquid phase, $S_{2_j}^{n+1}$ in the container node j, can be interpreted by the following equation (C-16):

$$S_{2_j}^{n+1} = \frac{\overset{N_r}{\underset{k=1}{}} C_{1k}^{n} ? V_k ? e^{-\lambda\Delta t} - \overset{N_r}{\underset{k=1}{}} C_{1k}^{n+1} ? v_k}{Volume\ _j ?\varepsilon ?\Delta t}$$

Eq. C-16

where v_k: volume at grid *k* in phase I

$Volume_j$: volume at node *j* in phase II

ε: porosity in phase II

After radionuclide releases from phases I to II, its fate is determined by the partial differential equation in phase II from (C-4).

$$\frac{fC_2}{ft} = \frac{D_2}{R}\frac{f^2C_2}{f\rho^2} + \frac{D_2}{R\rho}\frac{fC_2}{f\rho} + \frac{D_2}{R}\frac{f^2C_2}{fz^2} - \frac{v}{R}\frac{fC_2}{fz} - \lambda C_2 + S_2$$

Eq. C-17

Similar to phase I, the finite difference approach used here exploits the Crank-Nicholson scheme in time coordinates and the central difference scheme in spatial coordinates.

The implicit finite difference equation is reorganized into:

$$-\frac{-D_2\Delta\dagger}{2R\Delta z^2} - \frac{v\Delta\dagger}{4R\Delta z}\underset{\lrcorner}{\surd} C_{2j-N_p}^{n+1} + \overline{\ } -\frac{D_2\Delta\dagger}{2R\Delta\rho^2} + \frac{D_2\Delta\dagger}{4R\rho_j\Delta\rho}\underset{\lrcorner}{\surd} C_{2j-1}^{n+1} + \overline{\ }1 + \frac{D_2\Delta\dagger}{R\Delta\rho^2} + \frac{D_2\Delta\dagger}{R\Delta z^2} + \frac{\lambda\Delta\dagger}{2}\underset{\lrcorner}{\surd} C_{2j}^{n+1}$$

$$-\frac{-D_2\Delta\dagger}{2R\Delta\rho^2} + \frac{D_2\Delta\dagger}{4R\rho_j\Delta\rho}\underset{\lrcorner}{\surd} C_{2j-1}^{n+1} + \overline{\ } -\frac{D_2\Delta\dagger}{2R\Delta z^2} + \frac{v\Delta\dagger}{4R\Delta z}\underset{\lrcorner}{\surd} C_{2j+N_p}^{n+1}$$

Eq. C-18

$$= \frac{-D_2\Delta\dagger}{2R\Delta z^2} + \frac{v\Delta\dagger}{4R\Delta z}\underset{\lrcorner}{\surd} C_{2j-N_p}^{n} + \frac{-D_2\Delta\dagger}{2R\Delta\rho^2} - \frac{D_2\Delta\dagger}{4R\rho_j\Delta\rho}\underset{\lrcorner}{\surd} C_{2j-1}^{n} + \overline{\ }1 - \frac{D_2\Delta\dagger}{R\Delta\rho^2} - \frac{D_2\Delta\dagger}{R\Delta z^2} - \frac{\lambda\Delta\dagger}{2}\underset{\lrcorner}{\surd} C_{2j}^{n}$$

$$+\frac{-D_2\Delta\dagger}{2R\Delta\rho^2} + \frac{D_2\Delta\dagger}{4R\rho_j\Delta\rho}\underset{\lrcorner}{\surd} C_{2j-1}^{n} + \frac{-D_2\Delta\dagger}{2R\Delta z^2} - \frac{v\Delta\dagger}{4R\Delta z}\underset{\lrcorner}{\surd} C_{2j+N_p}^{n} + \frac{\Delta\dagger}{2}\left(S_{2_j}^{n} + S_{2_j}^{n+1}\right)$$

where *n* and *n*+1 represent the current time and the next time step, respectively

j stands for the number of nodes in phase II (1 through $N_{\rho z}$)

$N_{\rho z} = N_\rho \cdot N_z$

$S_{2_j}^{n}$ = the source term in container node j from resins based on the mass balance in (C-16) at time step *n*.

As in phase I, the right hand side stands for the current situation, while the left hand side represents the status for the next time step. The above equation can be arranged into matrix form as:

$$\overline{B}\overline{L} \leftrightarrow \overline{C}_2^{\,n+1} = \overline{B}\overline{R} \leftrightarrow \overline{C}_2^{\,n} + \overline{B}\overline{S} \qquad \textbf{Eq. C-19}$$

where $\overline{B}\overline{R}$ = five diagonal coefficient matrix for the current concentration distribution

$\overline{B}\overline{L}$ = five diagonal coefficient matrix for the concentration distribution of the next time step

$\overline{C}_2^{\,n}$ = matrix of the current concentration distribution in phase II

$\overline{C}_2^{\,n+1}$ = matrix of the concentration distribution of the next time step in phase II

$\overline{B}\overline{S}$ = source term in phase II

If either p or q is greater than $N_{\rho z}$ or not greater than zero in the elements $\overline{B}\overline{L}_{p,q}$ and $\overline{B}\overline{R}_{p,q}$ in the coefficient matrix, then both elements should be zero.

The amount of radionuclide released from the container for the next time step, $Release^{n+1}$, can be determined by the difference in the total amount of radionuclide retained in phase II between two consecutive time steps and corrected for radioactive decay as follows.

$$Release^{n+1} = {}_{j=1}^{N\,pz}\left(C_{2j}^{n} e^{-\lambda\Delta t} - C_{2j}^{n+1}\right) ?\ Volume_j\ ?\ \varepsilon\ ? \qquad \textbf{Eq. C-20}$$

The variables in (C-20) have been previously defined.

Boundary conditions are then applied to the governing equations, (C-15) and (C-19), to derive the specific numerical equation for this resin system. Hence, the procedure to analyze the resin system begins with the initial conditions, (C-5) and (C-6), for time n=0. The distribution of radionuclide concentration in resins can be determined by (C-15) for the first time step. Then, the amount of radionuclide released from the resin beads is known. With the updated S_{2j} values, the concentration distribution in the solution can be calculated by (C-19). Then, the updated amount of release is found using the mass balance in (C-20). As shown in Figure C-5, the calculation repeats steps 2 to 5 in order to determine the release in step 5 for each time interval.

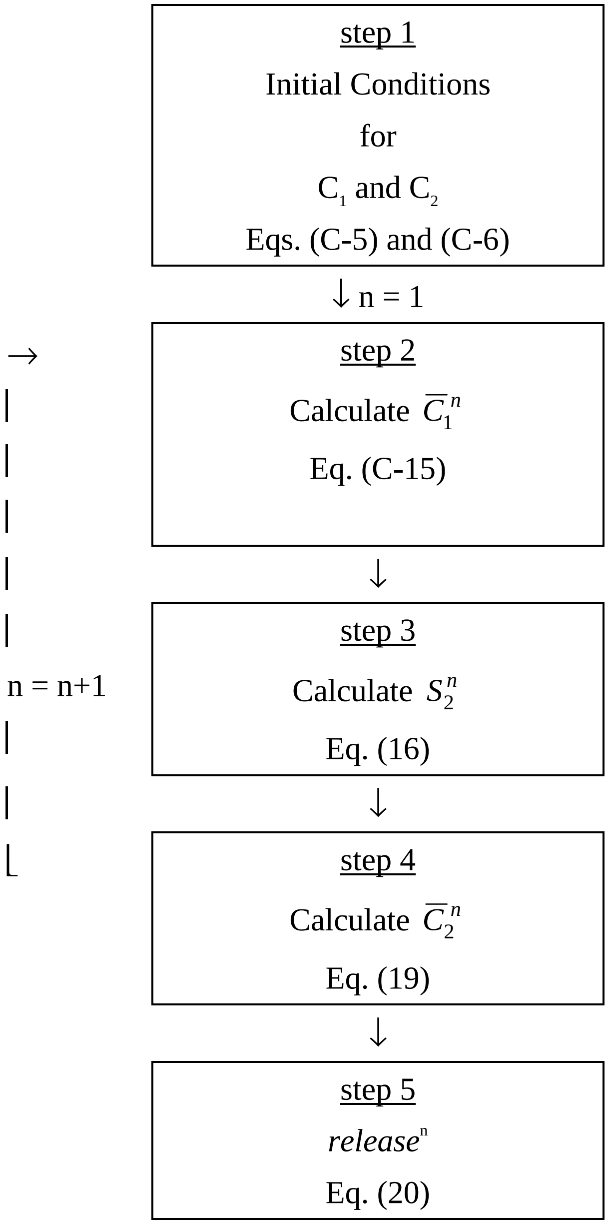


Figure C-5
Procedure of Numerical Solution for the Resin System [8]

C.1.4 Validation of the Model

The model and accuracy of numerical methods for the ion-exchange resin model are examined in this section. For this purpose, comparisons with appropriate analytical solutions or physical understanding of the release processes based on various hypothetical test cases are performed. These include description of diffusion in a sphere (for the resin bead release, Phase 1), diffusion in cylinder (for the bulk pore water phase) and interaction between resin particles and the bulk medium within a cylindrical container. Details of these comparisons are in the following

For the diffusional release of radionuclide from a sphere, phase I, the fraction of diffusing substance leaving a sphere can be analytically determined as [Crank, 1957]:

$$\text{Release} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{x} \frac{1}{n^2} e^{-\bar{D}_i n^2 \pi^2 t/\rho_r^2} \qquad \textbf{Eq. C-21}$$

For this expression, a uniform initial concentration and zero boundary conditions are assumed. The variables in this equation have been defined in section C.1. Figure C-6 represents the comparison of predicted release fraction between the model and the analytical solution for the spherical diffusion. The non-dimensional time in Figure C-6 is represented by $(\bar{D}_i t)^{0.5} / \rho_r$. The comparison shows good agreement between the analytical and numerical solutions in the spherical diffusion.

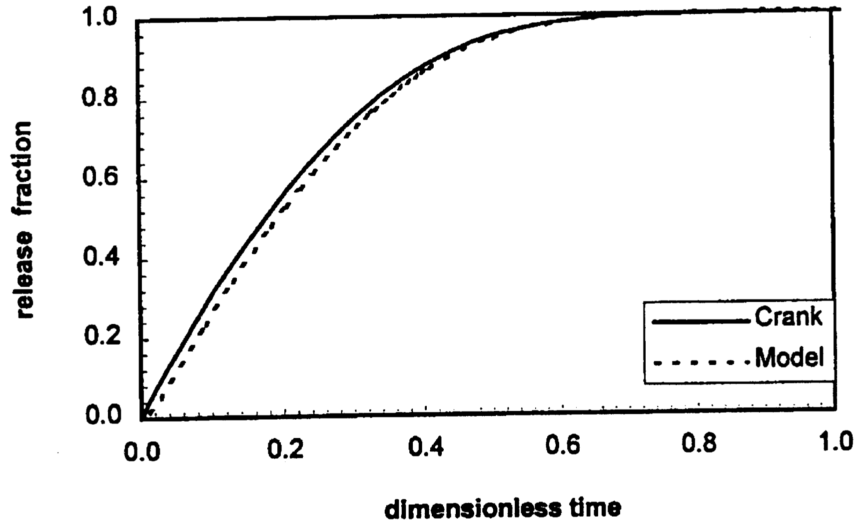


Figure C-6
Comparison between Model and Analytical Solution in Spherical Diffusion Equation

For diffusional release of radionuclide release from a cylinder, the analytical solution gives the fraction of diffusing substance leaving the side of a cylinder as follows [Crank, 1957]:

$$\text{Release } fraction = 1 - \sum_{n=1}^{x} \frac{4}{\alpha_n^2} e^{-\bar{D}_2 \alpha_n^2 t/\rho_c^2} \qquad \textbf{Eq. C-22}$$

Where, the α_n'n are roots of $J_0(\alpha_n) = 0$,

And $J_0(x)$ is the Bessel function of the first kind of order zero.

The other variables have been defined previously.

Again, uniform initial concentration and zero boundary conditions are assumed. Assuming that only diffusion in a radial direction is responsible for the release, the predicted release fraction from the model is compared with the analytical solution of Equation (C-22). Comparisons shown in Figure C-7 indicate close agreement between the model and the analytical solution of the cylindrical diffusion equation. And the scale of dimensionless time in Figure C-7 is defined as $(\overline{D_i}t)^{0.5} / \rho_c$ in cylindrical coordinate.

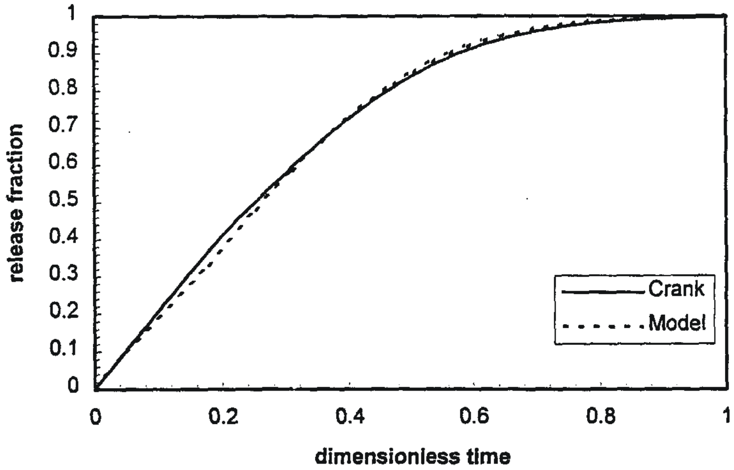


Figure C-7
Comparison between Model and Analytical Solution in Cylindrical Diffusion Equation

In order to validate the coupling of the model between steps 1 and 2, a hypothetical system where 0.025 cm-radius resins are stacked in a 25 cm-radius and 100 cm-height container is assumed. Again, it is assumed that diffusion is the only mechanism responsible for mass transfer. For a system with identical diffusion coefficients in both resins and liquid ($\overline{D_i}$ =6.1x10^{-8} cm^2/sec), the release fraction from resin beads (step 1) builds up quickly at the beginning of the leaching process. The release fraction from step 2, as expected, increases relatively slowly due to the time required for diffusion to make the release from a large container (refer to Figure C-8).

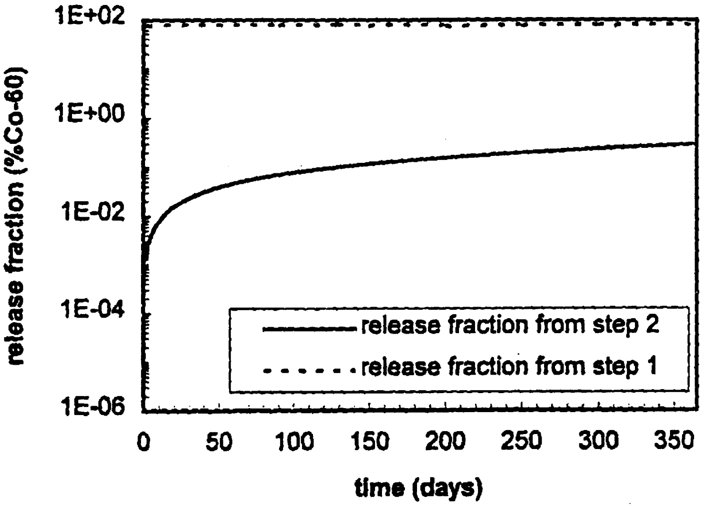


Figure C-8
Release Fractions in a Hypothetical Diffusion System with $D_2 = \overline{D_i}$

It is assumed that the diffusion coefficient in the liquid phase is much larger than that in the resins. When $D_2 = 10^3 \overline{D_i}$, the results show a trend (Figure C-9) similar to those in the system of Figure C-8. However, the increase of the release fraction from step 2 is much faster because of the larger liquid diffusion coefficient.

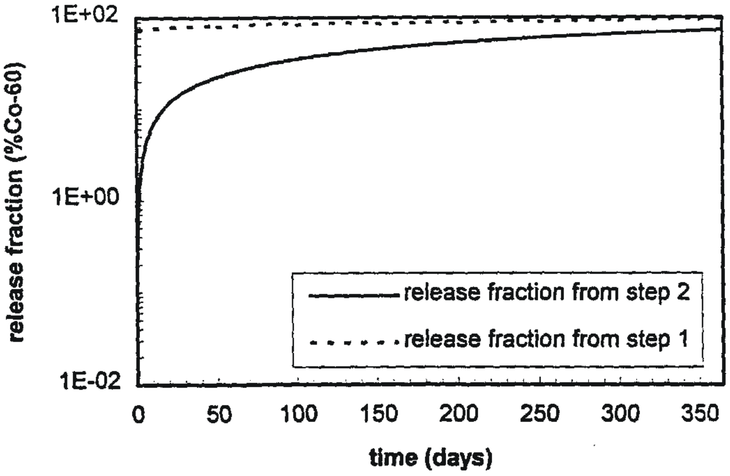


Figure C-9
Release Fractions in a Hypothetical Diffusion System with $D_2 = 10^3 \overline{D_i}$

In Figure C-10, the result of the predicted release fraction of step 2 is shown to be almost identical to the release fraction of step 1 when the liquid diffusion coefficient of the liquid phase is as much as 10^5 times the resin diffusion coefficient. These results correspond to the physical understanding of the processes and demonstrate the adequacy of the model coupling between phases I and II.

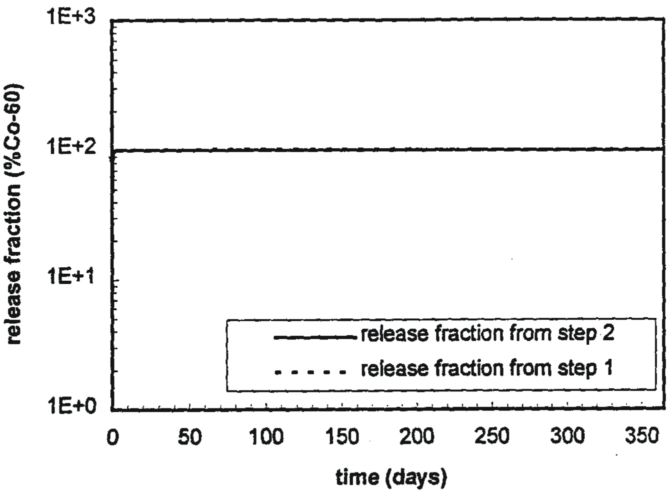


Figure C-10
Release Fractions in a Hypothetical Diffusion System with $D_2 = 10^5 \overline{D_i}$

C.1.5 Sensitivity Analysis

Sensitivity studies of the resin release model were performed [8] to examine the processes and components responsible for the release of radionuclides from IX resins. These parameters are listed in Table C-1. The corresponding ranges of parameters used in the sensitivity study of the resin system are also given in Table C-1.

Table C-1
Ranges of Parameters Used in Sensitivity Study of Resin System

Parameters	Range of Values
radius of resins	0.01 ~ 0.05 cm [9]
density of resins	1.1 ~ 1.3 g/cm^3 [9]
diffusion coefficient in resin	6.1x10^{-10} ~ 6.1x10^{-8} cm^2/sec [10]
water infiltration	1 ~ 100 cm/yr [11]
liquid diffusion coefficient in waste	6.1x10^{-8} ~ 6.1x10^{-5} cm^2/sec [11]
distribution coefficient of waste	0 ~ 54 ml/g [13]

Due to the difficulties in obtaining the experimental data for the effects of counter ions on leaching, it is assumed that the concentrations of the counter ions of the leachant water is at its maximum and the total inventory of activity in IX resins is available for release, subject to chemical reactions or transport processes. This would facilitate the examination of other parameters' effects on radionuclide release.

Effect of Resin Properties

The results of comparing the effects of different resin properties (such as resin bead radius, resin density, and interdiffusion coefficient for radionuclide migration within resin beads) are shown in Figure C-11. The results indicate the release of radionuclides from the bulk pore water phase is not affected by the examined properties within the given ranges (as given in Table C-1).

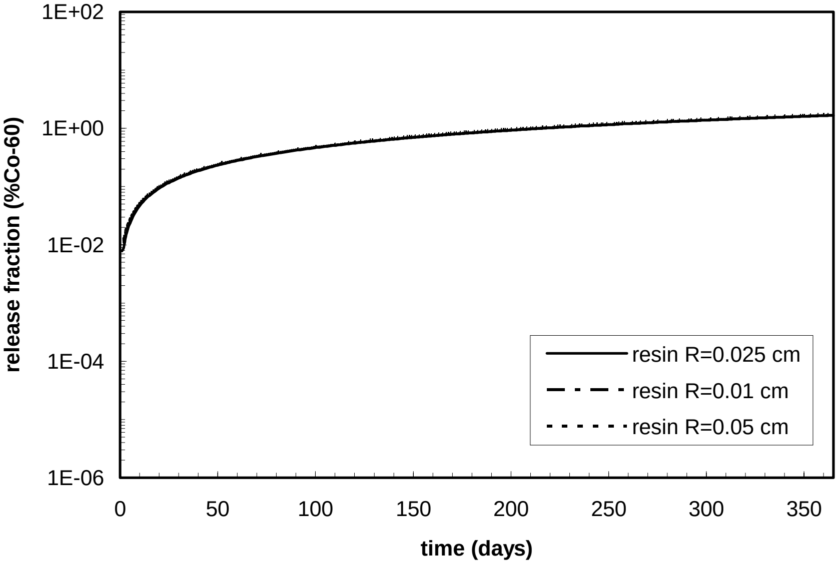


Figure C-11
Release from Ion-Exchange Resins with Different Resin Radius

In the diffusion process, the mean length, defined as the average distance traveled in the resins, is determined by multiplying the diffusion coefficient and the time interval and taking the square root. For example, consider radionuclide ^{60}Co with an interdiffusion coefficient of 6.1×10^{-8} cm^2/sec in resins of 0.025 cm radius. Radionuclide ^{60}Co can diffuse from the center of resin to the resin surface within one or two days. Since most radionuclides in LLW appear to have interdiffusion coefficients of similar magnitude within the resins, the time required for radionuclides to be released from IX resins is very short compared to the time frame of interest in performance assessment. This supports the predicted results that changes in physical properties of resins, such as interdiffusion coefficients, density and radius, do not affect the release of radionuclides from waste containers containing IX resins within the time frame of interest in performance assessment.

Effects of Water Infiltration, Diffusion Coefficient and Distribution Coefficient in the Bulk Pore Water Phase

Since the concentrations of radionuclides in the bulk pore water phase are controlled by the advective transport, diffusional transport, and sorption processes during the transport (Equation (C-4)), changes in the radionuclide concentrations in the bulk pore water phase are expected by the changes in rate of water infiltration into the resin matrix region, the diffusion coefficient of radionuclides in the bulk pore water, and the distribution coefficient of radionuclides. Again, the selected ranges of variation of these parameters expected in different disposal conditions are shown in Table C-1.

Figure C-12 shows the effects of the changes in the rate of infiltrating water flow on the predicted ^{60}Co release. The results indicate a strong dependence of the release on the rate of infiltrating water flow, particularly when the rate of infiltrating water flow is high.

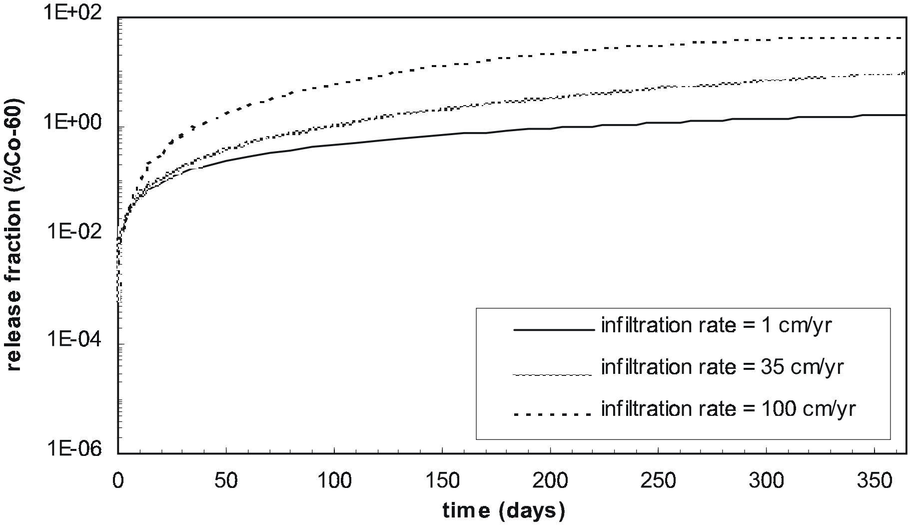


Figure C-12
Release from Ion-Exchange Resins with Different Flow Rate of Water Infiltration

When the rate of infiltrating water flow is low, the effect of diffusion becomes important and rate-limiting. This is shown in Figures C-13 and C-14. As shown in Figure C-13, when the rate of infiltration flow is low (at 1 cm/yr), the predicted release is largely dependent on the diffusion coefficient in the bulk pore liquid. But when the rate of water infiltration is much higher (at 100 cm/yr), the effect of diffusion coefficient variation on the release becomes much smaller (Figure C-14). This result demonstrates the importance of describing both diffusion and advection to predict radionuclide release at different site conditions.

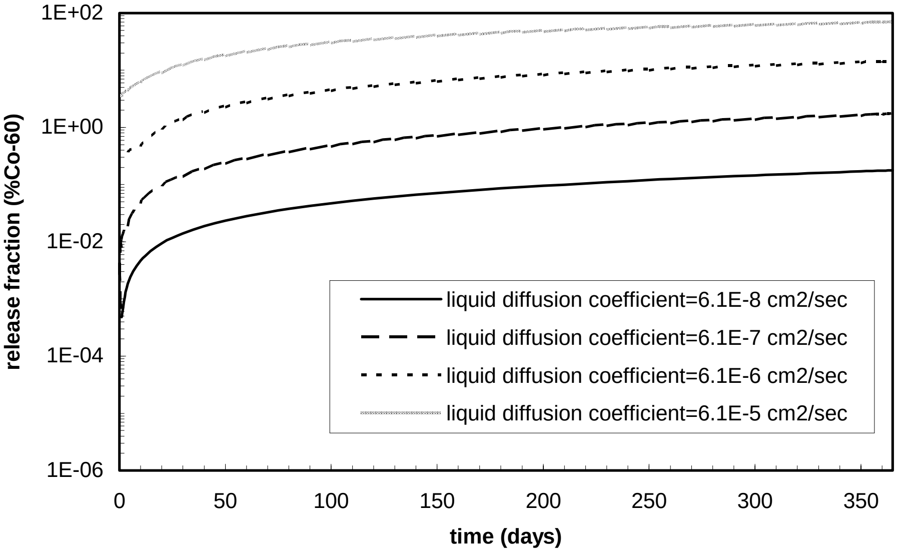


Figure C-13
Release from Ion-Exchange Resins with Different Liquid Diffusion Coefficients at the Infiltration Rate of 1 cm/yr

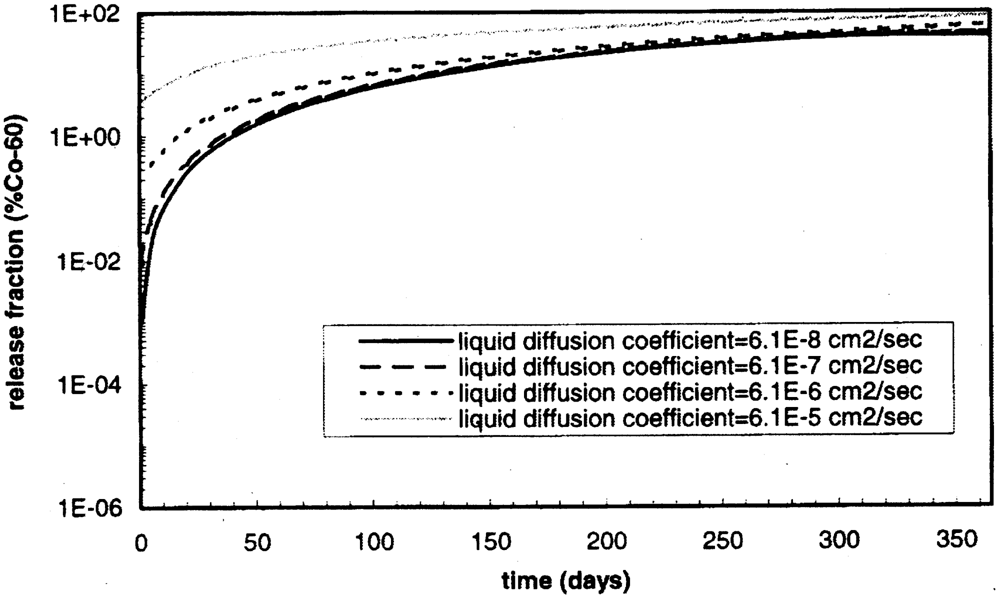


Figure C-14
Release from Ion-Exchange Resins with Different Liquid Diffusion Coefficients at the Infiltration Rate of 100 cm/yr

Effects of sorption, ion exchange, or any other chemical reactions during the transport of radionuclides in the bulk pore water phase on the release fraction are shown in Figure C-15. The effect is described by the use of different values of the distribution coefficient of ^{60}Co. As shown in the figure, the changes in the distribution coefficients (from 0 to 54) result in almost two orders of magnitude difference in the predicted release. When the distribution coefficient is larger, more radioactivity will be absorbed on the solid. With smaller distribution coefficients, radionuclides are easily released from the resin system.

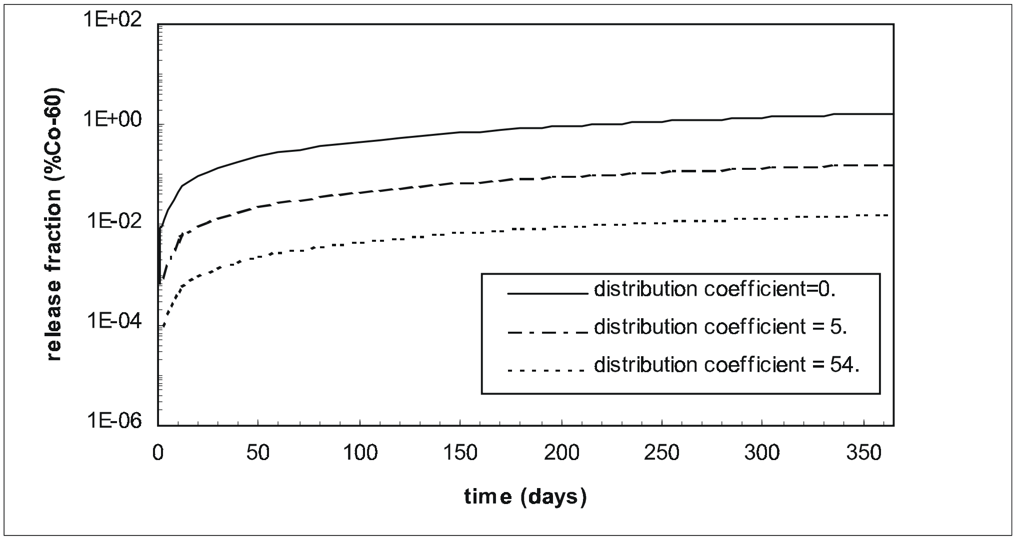


Figure C-15
Release from Ion-Exchange Resins under Different Liquid Distribution Coefficients

These findings indicate that important parameters affecting radionuclide release from IX resins are counterion concentrations of the contacting water, rate of infiltrating water flow, the diffusion coefficient and the distribution coefficient in the bulk pore liquid. Within the ranges of expected variation in radius and density of resin beads, the interdiffusion coefficient in resins appear to have little effect on radionuclide release for the time frame of interest in the performance assessment.

C.1.6 Comparisons with the Surface Rinse Model

In this section, the new resin release model developed in this study is compared with the surface rinse model. In this comparison, two different values of percolation flux of water are used to consider the effects of different site conditions on the release. The values of water percolation flux used are: 1 cm/yr to represent a dry site and 35 cm/yr to represent a humid site.

The surface rinse model is based on an assumption that all of the radionuclides in the waste forms are available for release through surface phenomena and that they are washed off by the contact of water which is controlled by equilibrium partitioning. The equilibrium partitioning is represented by the distribution coefficient (K_d). Therefore when the surface rinse model is adopted to describe the release of radionuclides from IX resins, it is implied that the whole inventory of radionuclides is available for release regardless of the chemical compositions of the infiltrating water or the manner in which the radionuclides are attached to the resin matrix. And the predicted release is virtually determined by the value of K_d used in the calculation. The K_d value is typically chosen from existing data base. For the radionuclides known for very low sorptive activity (such as ^{14}C or ^{129}I), small values of K_d (i.e., 0.01~1 ml/g) are used and the predicted release from resins is expected to be significant. For the radionuclides known for large sorptive activity (most of the cations), large values of K_d (i.e., greater than 10 ml/g) are used and the predicted release would be rather small. These K_d values however do not necessarily represent the actual conditions of nuclide leaching from the waste package containing spent resin beads.

The new resin release model assumes that the radionuclides are distributed uniformly within resin beads and released from them through ion exchange or surface rinse. Only the activity captured in the resin surfaces will be released through surface rinse. The remainder of the activity will be released through ion exchange controlled by the chemical compositions of the infiltrating water. And the model takes into account the fact that the activity released from each resin bead will go through diffusional, advective transport inside the waste container being subject to chemical reactions until it is released out of the container.

Key considerations in using the new resin release model in comparison with the surface rinse model are defining the fraction of activity in resins available for surface rinse, the concentration of counterions in the percolating water, and the diffusion coefficient in the bulk pore water phase. Based on these considerations, hypothetical bounding cases are developed for the resin release model for comparison with the surface rinse model.

The fraction of activity available for surface rinse is found to be much different between the radionuclides with low sorption (small K_d) and the radionuclides with high sorption (large K_d). Based on preliminary experimental studies [14], this fraction is estimated to range between

0.3% and 72% for the radionuclides with small K_d and to be less than 1% for the radionuclides with large K_d values. For the diffusion of radionuclides in the bulk pore water phase, it is expected that the actual diffusion coefficient lies in between the diffusion coefficient in water and the diffusion coefficient in resins. Conservative estimates of these two values - diffusion coefficient of the radionuclide in water (10^{-6} cm^2/sec) and diffusion coefficient in resin beads (10^{-8} cm^2/sec) - are used as bounding values in the analysis. The concentration of counterions in the percolating water can be assumed at their maximum for this comparison purposes making all of the activity in resins available for release through ion exchange. Comparison between the new resin release model and the surface rinse model is made separately for the radionuclides with relatively large sorption (high K_d) within the resins and the radionuclides with very low sorption (small K_d) within the resins.

For the radionuclides with low sorption, the hypothetical upper bound case is: (1) To use the surface rinse fraction of 90% and the liquid diffusion coefficient of 10^{-6} cm^2/sec for the new resin release model, and (2) To use the K_d value of 0.01 ml/g for the surface rinse model. For the hypothetical lower bound case with the radionuclides with low sorption: (1) The surface rinse fraction of 1% and the resin diffusion coefficient of 10^{-8} cm^2/sec are used for the new resin release model, and; (2) The K_d value of 1 ml/g is used for the surface rinse model.

For the radionuclides with high sorption, the hypothetical upper bound case is: (1) To use the surface rinse fraction of 10% and the liquid diffusion coefficient of 10^{-6} cm^2/sec for the new resin release model, and (2) To use the K_d value of 1 ml/g for the surface rinse model. For the hypothetical lower bound case with the radionuclides with high sorption: (1) The surface rinse fraction of 0.1% and the resin diffusion coefficient of 10^{-8} cm^2/sec are used for the new resin release model, and; (2) The K_d value of 100 ml/g is used for the surface rinse model.

Comparison for Radionuclides with Low Sorption

Figure C-16 shows the results from two models for the radionuclides with low sorption under high infiltration, where the upper and lower bound cases are indicated for both of the model.

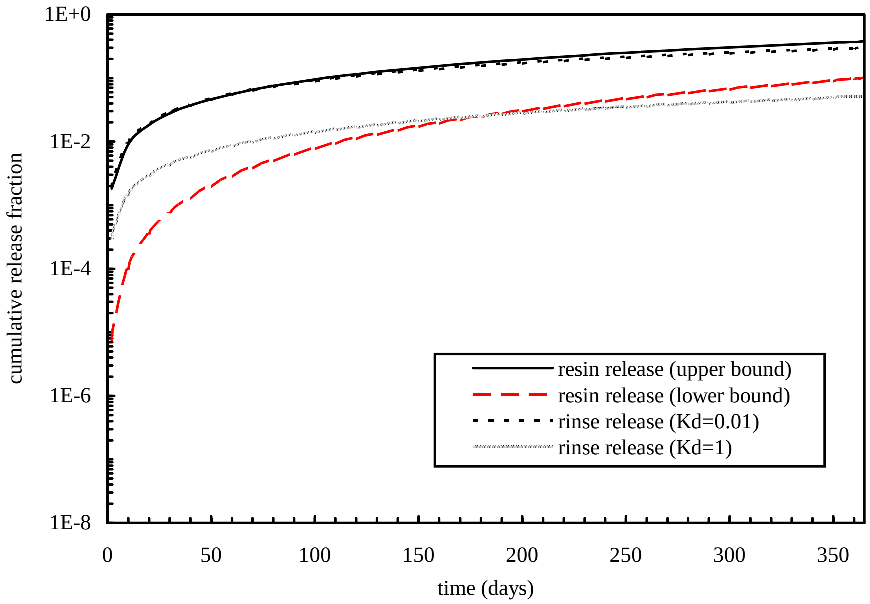


Figure C-16
Comparisons between Resin Release Model and Surface Rinse with Small K_d under High Infiltration Rate

The predicted radionuclide release from the resin release model is 37.6% per year in the upper bound case and 9.6% per year in the lower bound case, which are close to the predictions from the rinse release with K_d of both 0.01 and 1.

The comparisons of the results under low infiltration conditions for the radionuclides with low K_d are shown in Figure C-17. The predicted release of radionuclide is highest with the resin release model with 10^{-6} cm^2/sec of bulk pore water phase diffusion coefficient (3.55% per year) and lowest with the resin release model with 10^{-8} cm^2/sec of bulk pore water phase diffusion coefficient (0.038% per day). The results from the surface rinse model lie in between these two cases (with predicted release of 0.96% with $K_d = 0.01$ and 0.15% per year with $K_d = 1$). These results indicate that under low infiltration conditions, the resin release model is very sensitive to the bulk pore water phase diffusion coefficient and can predict higher result than the surface rinse model.

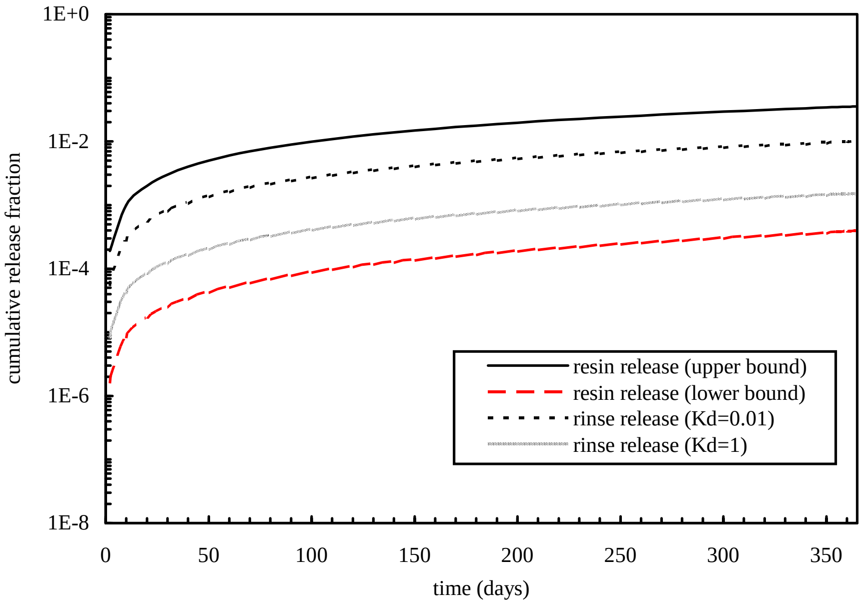


Figure C-17
Comparisons between Resin Release Model and Surface Rinse with Small K_d under Low Infiltration Rate

Comparison for Radionuclides with High K_d

For radionuclides with relatively high K_d, comparisons of the predicted release show (Figures C-18 and C-19) that the resin release model predicts higher release than the surface rinse model in general under both the high and low infiltration conditions, with the exception of the surface rinse model with $K_d = 1$ at the low infiltration case. As observed earlier, difference in the bulk pore water phase diffusion coefficient was not important under the high water filtration conditions for the resin release model, but was important under the low water infiltration conditions. In the low water infiltration case, the predicted release from the surface rinse model is 0.14% per year with K_d at 1 and 3.52×10^{-5}% per year with K_d at 100. With the resin release model, the predicted release is 2.53% per year with bulk pore water phase diffusion coefficient of 10^{-6} cm^2/sec (upper bound case) and 3.38×10^{2}% per year with the bulk pore water phase diffusion coefficient of 10^{-8} cm^2/sec (lower bound case).

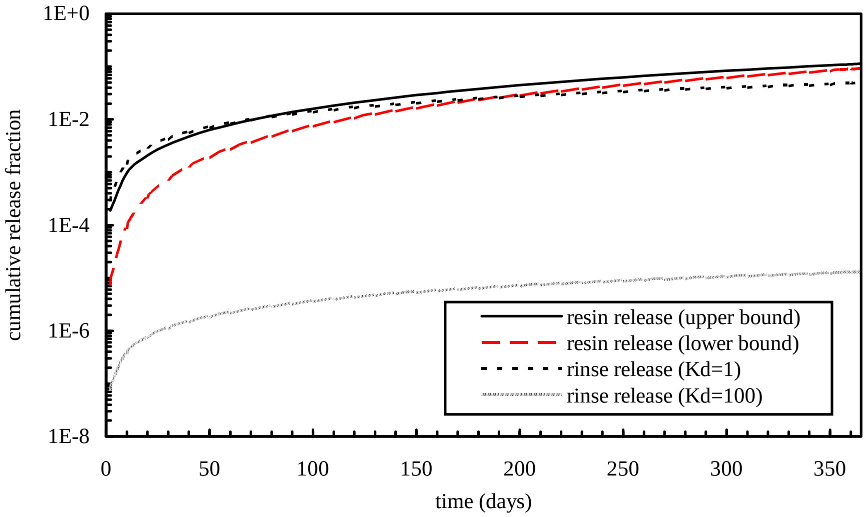


Figure C-18
Comparisons between Resin Release Model and Surface Rinse with Large K_d under High Infiltration Rate

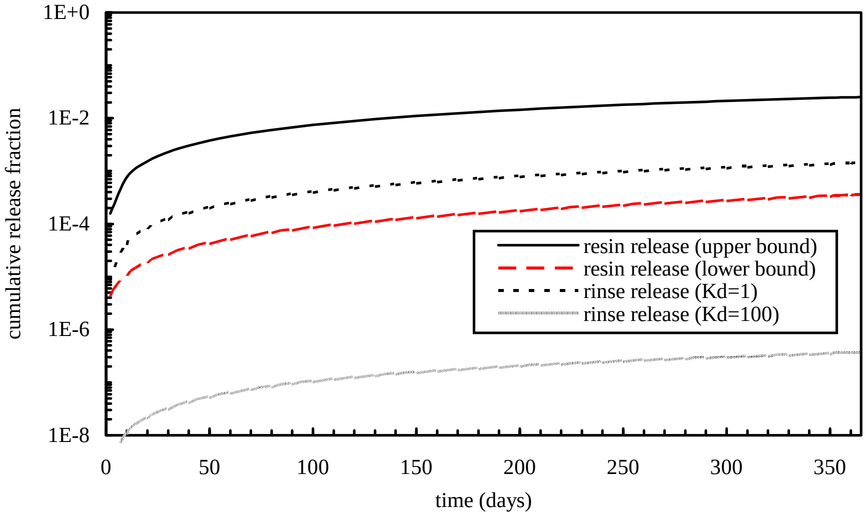


Figure C-19
Comparisons between Resin Release Model and Surface Rinse with Large K_d under Low Infiltration Rate

These results suggest that the use of surface rinse model may be acceptable with proper selection of the distribution coefficients. For the resin release model, accurate value of this diffusion coefficient is not currently available.

C.1.7 Summary and Conclusions

In this research, a new resin release model was developed to enhance the current state of knowledge to describe LLW disposal facility source term.

The new model developed is based on description of resin bead phase and the bulk pore water phase within waste containers separately. This modeling setup not only allows the prediction of radionuclide release from spent ion exchange resins but also provides capability to describe all the physical processes taking place for the release of radionuclides. Due to the difficulty in obtaining analytical solutions, a numerical solution approach was employed in the model.

By using this new resin release model, key processes and parameters in describing radionuclide release were examined. These are counterion concentrations, flow rate of infiltrating water, diffusion within the bulk pore water phase, and sorption during the transport in the bulk pore water phase. Some of the parameters were found to have little impact in describing the phenomena. These include interdiffusion coefficient within resin beads, density and radius of resin beads.

Insights gained from the use of this model may allow the use of more simplified modeling approaches. This study finds that the release is not only controlled by the partitioning between resins and the water but also limited by the diffusion in the bulk pore water phase. The use of rinse model ignores the bulk pore water phase diffusion. This study indicates that using simple models requires good understanding of the site-specific situation . Under certain conditions of water infiltration and sorption, these simple models are applicable.

Even though there has been a large amount of work done for the understanding of contaminant diffusion within IX resins, it has been found in this study that those studies have little relevance in modeling radionuclide release from IX resins. In contrast, work for the understanding of radionuclide release from IX resins is basically not existing. The research performed in this study lays basic understanding of the phenomena and provides a model for the analysis required in modeling IX resin leaching.

However, more experimental work is needed for the full utilization of the developed model. This includes the work on the effects of counterion concentrations in the leachant water on the leaching of radionuclides and the diffusion characteristics in the bulk pore water phase. Effects of long-term degradation of resins (i.e., irradiation effects, microbial degradation) on the leaching of radionuclides appear to be worthy of further investigation.

C.2 Cemented Waste

In the case of solidified waste, such as cemented waste, release of radionuclides takes place through diffusion of radionuclides through the pore water in the waste form. This process is known to be independent of the degree of water saturation of the porous matrix but is dependent on the hydraulics of the surroundings which determine the concentration gradient for diffusion. Typically currently available models for diffusion assumes zero nuclide concentration at the outer boundary of the waste form to facilitate the derivation of analytical solutions. However, with the trend of using cement backfills, the capability to address the effects of surrounding conditions on the diffusive release of radionuclides is desired. Approaches taken in this study to describe the release of radionuclides from cemented waste include both analytical solution model and numerical model. Analytical model, once developed, is easy to be implemented and does not involve any issues of numerical stability. However, analytical solution models are typically restricted to certain defined conditions. For more generalized application of the model, numerical model is also desired.

C.2.1 Analytical Solution Model

For the description of diffusion-controlled release, most of the existing models assume zero nuclide concentration at the waste form surface boundary to facilitate the derivation of analytical solutions. This boundary condition leads to release rates that are conservative and independent of the surrounding conditions. Effects of surrounding conditions (backfills) on the diffusive release of radionuclides have been investigated by using the numerical inverse Laplace transform technique [Kim, 1995] to calculate the changes in the release rate due to the variations in the diffusion coefficient in the backfill media. No analytical solution was given to describe the effects of backfill on the diffusion from waste.

The basic mathematical model used in this study to describe the backfill-dependent radionuclide diffusion is a two region cylindrical model,

$$\frac{fC_1(r,t)}{ft} = \frac{D_w}{R_w \quad r}\frac{f}{fr}(r\frac{fC_1(r,t)}{fr}) - \lambda C_1(r,t), 0 < r < r_w, t > 0 \qquad \textbf{Eq. C-23}$$

$$\frac{fC_2(r,t)}{ft} = \frac{D_s}{R_s \quad r}\frac{f}{fr}(r\frac{fC_2(r,t)}{fr}) - \lambda C_2(r,t), r \,?\, r_w, t > 0 \qquad \textbf{Eq. C-24}$$

$C_1(r,t)$ = concentration of radionuclide in porewater within the waste form

$C_2(r,t)$ = concentration of radionuclide in porewater of the surrounding medium

D_w = diffusion coefficient of radionuclide in porewater within the waste form

D_s = diffusion coefficient of radionuclide in porewater of the surrounding medium

R_w = retardation coefficient of radionuclide within the waste form

R_s = retardation coefficient of radionuclide in the surrounding medium

λ = decay constant

r_w = radius of the waste form

The initial and boundary conditions for these equations are:

$$C_1(r,t=0) = C_0, \quad 0 \le r \le r_w$$

$$C_2(r,\dagger = 0) = 0, \quad r > r$$

$$C_1(r_w,\dagger) = C_2(r_w,\dagger), \quad \dagger^w > 0$$

$$-\varepsilon_1 D_w \frac{fC_1(r_w,t)}{fr} = -\varepsilon_2 D_s \frac{fC_2(r_w,t)}{fr}, \quad t > 0$$

$$\frac{fC_1(0,t)}{fr} = 0, t > 0$$

$$C_2(\mathrm{x},t) = 0, t\,?\,0$$

where,

$\varepsilon_{1,2}$ = *porosity of waste/backfill*

C_0 = *initial nuclide concentration porewater within the waste form* = $\dfrac{S_{diff}}{\pi r_w^2 H(1 + K_d\ \rho\ /\ \theta)}$

S_{diff} = *source activity available for diffusion*

H = height of the waste form

Solutions of these equations can be sought by the Laplace transform approach. The rate of radionuclide release from the waste form in the Laplace transformed domain can be given by[1],

$$Q_{diff}(s) = dS\,?\,\overline{}\text{-}\varepsilon_1 D_w \frac{d\overline{C}_1(r,s)}{dr} \begin{matrix}\surd\\ \surd\\ \hookleftarrow_{r=r_w}\end{matrix} = 2\pi r_w H \quad F_c\,\overline{}{-}\varepsilon_1 D_w \frac{d\overline{C}_1(r_w,s)}{dr} \begin{matrix}\surd\\ \surd\\ \hookleftarrow\end{matrix}$$

$$= 2\pi r_w H \quad F_c \varepsilon_1 \sqrt{D_1} \quad C_0 \frac{1}{\sqrt{s}} \frac{1}{\dfrac{I_0(\sqrt{sr_w^2}\,/D_1)}{I_1(\sqrt{sr_w^2}\,/D_1)} + \alpha \dfrac{K_0(\sqrt{sr_w^2}\,/D_2)}{K_1(\sqrt{sr_w^2}\,/D_2)}}$$

Eq. C-25[2]

where,

Q_{diff}*(s) = total diffusive release rate in the Laplace transformed domain(s)*

F_c = *correction factor from an infinite to a finite cylinder* = $1 + r_w / H$

$D_1 = D_w / R_w$ = *effective diffusion coefficient within the waste form*

$D_2 = D_s / R_s$ = *effective diffusion coefficient in the surrounding medium*

$$\alpha = \frac{\varepsilon_1 R_w \sqrt{D_1}}{\varepsilon_2 R_s \sqrt{D_2}}$$

---

[1] In this equation, radioactive decay is ignored and is described in the final solution.

[2] I_0, I_1, K_0, K_1 are the modified Bessel functions of the first and second kind.

[3] In performance assessment, the peak dose usually comes from the initial release of mobile radionuclides.

I_0, I_1, K_0, K_1 are the modified Bessel functions of the first and second kind.

Due to the complexity in the inversion of the Laplace transform, it is difficult to obtain the exact solution for the entire time domain. However, for the period of early release which is important in performance assessment an asymptotic solution can be derived (In performance assessment, the peak dose usually comes from the initial release of mobile radionuclides). The solution is:

$$Q_{diff}(t) = \frac{2\pi r_w H \quad F_c \varepsilon_1 D_1^{1/2} C_0}{(1+\alpha)} \frac{\alpha D_2^{1/2} - D_1^{-1/2}}{2(1+\alpha) r_w} + \frac{t^{-1/2}}{\sqrt{\pi}} - \frac{D_1 + 3\alpha(D_1 + D_2) + \alpha^2 D_2 + 4\alpha D_1^{1/2} D_2^{1/2}}{4(1+\alpha)^2 r_w^2} \frac{t^{1/2}}{\sqrt{\pi}}$$

$$+ \frac{3(D_1^{3/2} + \alpha D_1^{1/2} D_2 - \alpha D_1 D_2^{1/2} - \alpha^2 D_2^{3/2}}{8(1+\alpha)^2 r_w^3} t \qquad ?e^{-\lambda t}$$ **Eq. C-26**

C.2.2 Numerical Solution Approach

The analytical solution model in the previous section relies on the assumption that the release is taking place in the early period. To relieve this restriction, numerical model is also implemented. The geometry of the waste is again considered cylindrical. The model considers the effects of advective transport to consider a situation where advective velocity of water in the waste form is comparable to diffusive velocity of contamiant. The model can describe the changes in radionuclide concentration in waste containers as a function of two-dimensional diffusion, one-dimensional advection and radioactive decay.

In order to describe diffusion more realistically, which is dependent upon the backfill condition, the diffusion model needs to incorporate the continuity laws at the boundary.

$$\frac{fC_2}{ft} = \frac{D_2}{R}\frac{1}{\rho}\frac{f}{f\rho}\ \overline{\ \ }\ \rho\frac{fC_2}{f\rho}\surd\hookleftarrow + \frac{D_2}{R}\frac{f^2 C_2}{fz^2} - \frac{v}{R}\frac{fC_2}{fz} - \lambda C_2 + S_2$$ **Eq. C-27**

for $0 \le r \le r_c$

$0 \le z \le h$

where C_2: radionuclide concentrations in the pore water in the solid (Ci/cm^3)

D_2: diffusion coefficients in the pore water in the solid (cm^2/yr)

ν: infiltration rate in container (cm/yr)

l: decay constant of radionuclide (yr^{-1})

R: retardation coefficient in the pore water in the solid

r_c: the radius of cylinder container (cm)

h: container height (cm)

The diffusion model of a homogeneous system assumes that the nuclide concentration is initially uniform throughout the entire container, shown as:

$$C_2(\rho, z, t = 0) = C_0 \qquad \textbf{Eq. C-28}$$

where C_0: initial concentration in the diffusion model of a homogeneous system (Ci/cm^3)

The boundary conditions in the diffuison model are:

a. The concentration distribution is symmetric at the center (r = 0) of the radial container plane.

$$-D_2 \frac{fC_2}{f\rho}\bigg|_{\rho=0} = 0 \qquad \textbf{Eq. C-29}$$

b. There is no concentration flux across the radial boundary (r = r$_c$).

$$-D_2 \frac{fC_2}{f\rho}\bigg|_{\rho=\rho_c} = 0 \qquad \textbf{Eq. C-30}$$

c. The continuity of radionuclide flux at the top of the container.

$$-\frac{D_2}{R}\frac{/C_2(x,z,\dagger)}{/z}\bigg|_{z=0^+} + \frac{vC_2}{R}\bigg|_{z=0^+} = -\frac{D_2}{R}\frac{/C_2(x,z,\dagger)}{/z}\bigg|_{z=0^-} + \frac{vC_2}{R}\bigg|_{z=0^-} \qquad \textbf{Eq. C-31}$$

d. The continuity of radionuclide flux at the bottom of the container.

$$-\frac{D_2}{R}\frac{/\,C_2(x,z,\dagger)}{/\,z}\bigg|_{z=h+0^+} + \frac{vC_2}{R}\bigg|_{z=h+0^+} = -\frac{D_2}{R}\frac{/\,C_2(x,z,\dagger)}{/\,z}\bigg|_{z=h+0^-} + \frac{vC_2}{R}\bigg|_{z=h+0^-} \qquad \textbf{Eq. C-32}$$

The transport equation (C-27) is numerically solved by the implicit Crank-Nicholson approach. Hence, the distribution of radionuclide concentration can be determined by the methodology in section 9.3 using zero S$_2$ and the corresponding initial condition of (C-28). The release of radionuclide out of the container Q_{diff} can then be computed by, $Q_{diff} = dS?J_S\big|_{r=r_s}$

where, J_s is the mass flux at the surface ($J_S = -D\frac{fC(r_s)}{fx}$). r_s denotes a surface of the waste form and dS is the element of the surface area.

C.3 Validation of the Model

Valiation aspects of the new computer model for the source term analysis is described in this section. Ideally the accuracy of the model needs to be examined against experimental data. Due to the lack of experimental information, comparisons with the results of DUST code were made for the model validation purpose. DUST is a code developed for NRC's LLW licensing purposes. And among the various source term codes available, DUST (see also section 6.1.1.3) is the most extensively examined code and the results are expected to be nonarbitrary.

For this comparison, the followings are used in both of the computer models:

1. The height of the waste container is 100 cm.
2. The radius of the waste container is 25 cm.
3. The waste container is carbon steel drum.
4. The waste container region is divided into 10 node regions for numerical solutions.
5. The waste container contains 1 Ci of C-14.
6. Density of waste is 1.331 g/cm^3.
7. Porosity of waste is 0.1.
8. Moisture content of the waste is 1.0
9. The Darcy velocity through the waste region is 1.0 cm/yr.
10. The K_d value of C-14 in the noncemented waste is 0.01 ml/gm.
11. The K_d value of C-14 in the cemented waste is 10 ml/gm.
12. Diffusion coefficient of C-14 in the cemented waste is $1x10^{-6}$ cm^2/sec
13. The fractional release rate from the congruent dissolution is $1x10^{-5}$ yr^{-1}.

The analysis is performed for each individual process of radionuclide release, i.e., the release of the total inventory in the waste container is controlled by only one process. These results as concentrations of C-14 in the immediate container outside region are shown in Figures C-20 through C-22. The comparison of rinse release between DUST and the new model is shown in Figure C-20. The comparison of diffusional release is shown in Figure C-21. The results of dissolution release are compared in Figure C-22.

The comparisons indicate that for all of the release mechanisms analyzed (rinse release, diffusion, and dissolution) the results from both codes are in good agreement in terms of the magnitude of the predicted release and the spatial changes in the concentration. The results of rinse release show that the predicted release from the new model is slightly lower that what is predicted by the DUST. This is also true for the case of dissolution release. The results of diffusional release from the new code was slightly higher that what is predicted by DUST.

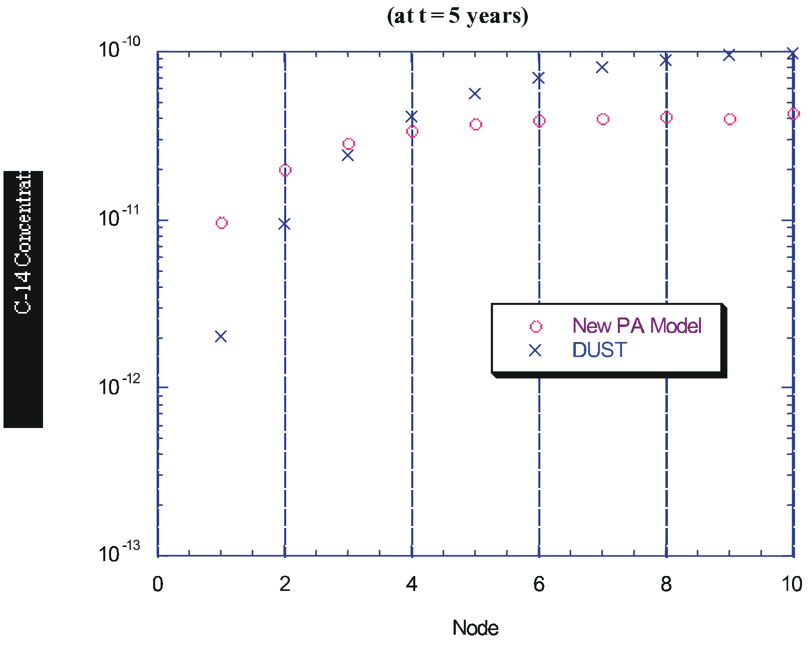


Figure C-20
Comparison of Rinse Release between DUST and the New PA Model

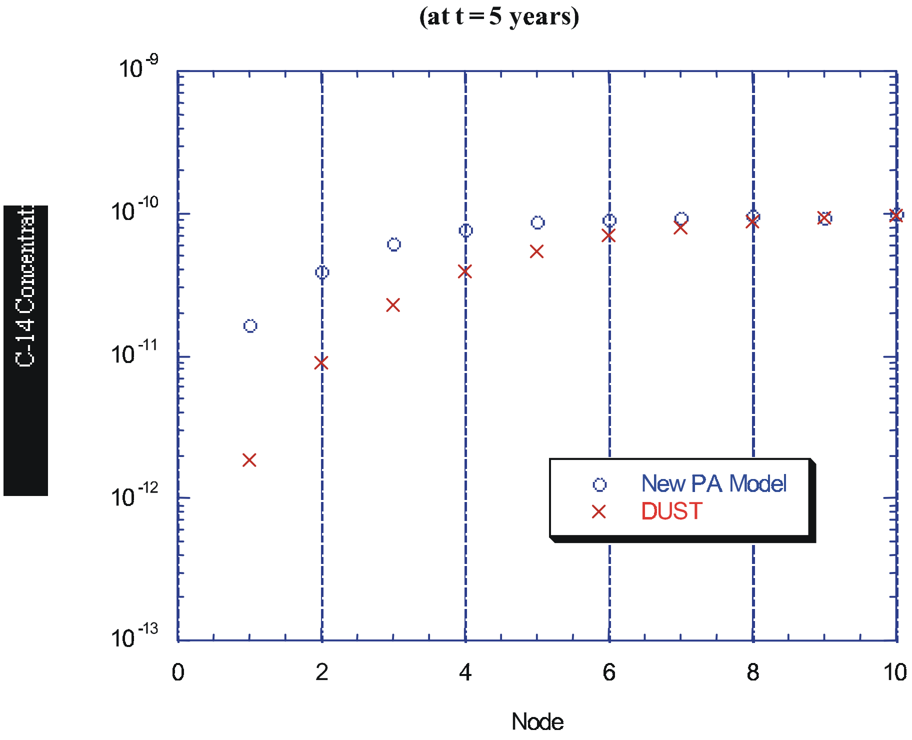
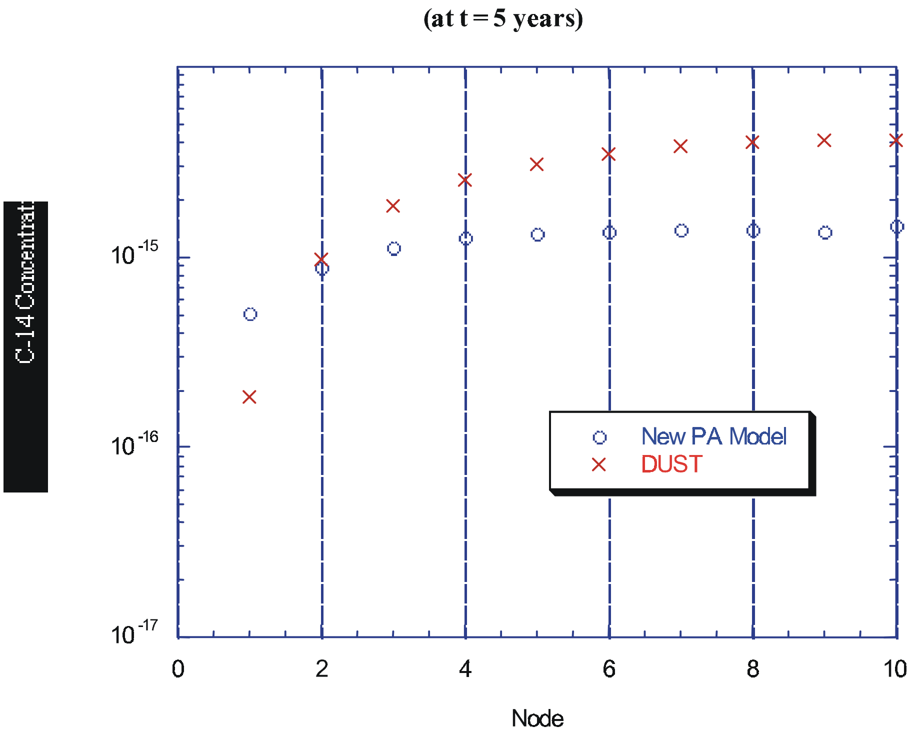


Figure C-21
Comparison of Diffusional Release between DUST and the New PA Model



(at t = 5 years)

Figure C-22
Comparison of Dissolution Release between DUST and the New PA Model

References

1. Dames and Moore, “Waste Inventory Revision 3, North Carolina Low-Level Radioactive Waste Disposal Facility Project,” DM/DR-002, Prepared for Chem-Nuclear Systems, Inc. by Dames and Moore, 1995.

2. Glasser, F. P., “Progress in the Immobilization of Radioactive Wastes in Cement,” *Cement and Concrete Research*, Vol. 22, pp. 201-216, 1992.

3. Helfferich, F., “Ion Exchange Kinetics,” in Marinsky J. A. (ed.), Ion Exchange, Marcel Dekker, 1966.

4. Levenspiel O., *Chemical Reaction Engineering*, 2nd edn., Chap. 12, Wiley, 1961.

5. Helfferich, F., *Ion Exchange*, New York, McGraw-Hill, 1962.

6. Schlogl, R. and Helfferich, F., “Comments on the Significance of Diffusion Potentials in Ion Exchange Kinetics,” *J. Chem. Phys.*, Vol. 26, pp. 5, 1957.

7. Guria, C. and Chanda, M., “Shell-Core Models for Ion-Exchanger Loading in Finite Bath: Sorption of Aqueous Sulphur Dioxide on Cross-Linked Poly(4-Vinyl Pyridine),” *Trans IChemE*, Vol. 72, Part A, pp. 503-512, July 1994.

8. Su, S.-I., “Development of a Mechanistic Resin Release Model for Low-Level Waste Source Term Analysis,” Ph.D. Thesis, North Carolina State University, 1997.

9. Rohm and Haas Company, Material Safety Data Sheet, Product Code: 698555, 1991.

10. Schwarz, A., Marinsky, J. A., and Spiegler, K. S., “Self-Exchange Measurements in a Chelating Ion-Exchange Resin,” J. Phys. Chem., Vol. 68, pp. 918, 1964.

11. U.S. Nuclear Regulatory Commission, “Source Term Implementation for Initial Performance Assessment Calculation,” Technical Review of Performance Assessment Modeling Strategy, Rockville, MD, July 12-13, 1993.

12. U.S. Nuclear Regulatory Commission, Low-Level Waste Performance Assessment Workshop, Rockville, MD, Nov. 16-17, 1994.

13. Chem Nuclear Systems, Inc, “Wake/Chathem County Site Safety Analysis Report,” November 1994.

14. Yim, M.-S., unpublished internal report, Department of Nuclear Engineering, North Carolina State University, 1998.