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Pacific Northwest
National Laboratory
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U.S. Department of Energy

Data Catalog for Models Simulating Release of Contaminants from Hanford Site Waste Sources

R. G. Riley
C. A. Lo Presti

September 2001

Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RL01830

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Richland, Washington 99352

Foreword

The Characterization of Systems (CoS) Task under the Groundwater/Vadose Zone Integration Project is responsible for establishing a consistent set of data, parameters, and conceptual models to support efforts at the Hanford Site to estimate contaminant migration and impacts. As part of these efforts CoS is assembling a series of catalogs to identify the depth and breadth of existing data and to facilitate access to those data. The preparation of these catalogs is aimed at facilitating the development of comprehensive, useable, and scientifically defensible database(s). However, it is also envisioned that these catalogs will be "living documents" that will continue to evolve as other existing data is found and new data collected.

This catalog summarizes information on models that have been used to simulate release of chemical and radioactive contaminants from waste sources on the Hanford Site over the past 14 years.

Summary

Models have been used to simulate the transport and fate of contaminants at the Hanford Site to assess the effectiveness of various environmental restoration and waste management activities and to estimate impacts to environmental and human receptors. Part of the process includes the ability to simulate the release to the vadose zone and groundwater of a wide range of contaminants from a wide range of waste sources that have been generated at the Hanford Site as a result of over 40 years of defense production activities.

Mathematical formulas that express the release of contaminants from various waste sources and their application in past assessments are documented in many reports published over the past 14 years. The sheer number of reports in which such information is reported makes it difficult for individuals to access this information in a timely manner. To provide users carrying out Hanford assessments with improved access to the information and data on release model capability, researchers at the Pacific Northwest National Laboratory (PNNL) consolidated these sources of information and data into a single location. This work was conducted under the Vadose Zone Groundwater Integration Project managed by Bechtel Hanford Incorporated for the U.S. Department of Energy – Richland Operations (DOE-RL).

This report provides summaries of release models used in Hanford Site assessments published over the past 14 years (1987 to 2001). Mathematical formulations that commonly have been used in recent years (i.e., salt-cake, cement, soil-debris, reactor block, glass, and corrosion) are described, along with associated parameter definitions and their units. Tables in this report provide links to data sources needed to implement the models. These links enable users to quickly locate the specific release model information and data sources they need for applying the models to future to site assessments.

During the compilation of the catalog, the authors became aware of significant differences in the level of complexity of release models applied over the years to meet Hanford Site assessment objectives. For example, release model capability to support the current 2001 ILAW performance assessment is far more advanced than the simpler mathematical formulations for glass release used in prior assessments and depicted in this report. We, therefore, encourage readers of this report to contact the individuals listed in Table 1.1 of this report for possible additional up-to-date information in their area of interest.

Acronyms and Abbreviations

DOE-RL	U.S. Department of Energy, Richland Operations
ERDF	Environmental Restoration Disposal Facility
HDWEIS	Hanford Defense Waste Environmental Impact Statement
HIC	high-integrity containers
ILAW	immobilized low-activity waste
LLW	low-level waste
MEPAS	Multimedia Environmental Pollutant Assessment System
NRC	U.S. Nuclear Regulatory Commission
PCB	polychlorinated biphenyls
PNNL	Pacific Northwest National Laboratory
RCRA	Resource Conservation Recovery Act
RH	remote handled
SAC	System Assessment Capability

Contents

Tables

1.0 Introduction

Models have been used to simulate the transport and fate of contaminants at the Hanford Site to assess the effectiveness of various environmental restoration and waste management activities and to estimate impacts to environmental and human receptors. Part of the process includes the ability to simulate the release to the vadose zone and groundwater of a wide range of contaminants from a wide range of waste sources that have been generated at the Hanford Site as a result of over 40 years of defense production activities.

Mathematical formulas that express the release of contaminants from various waste sources, the parameters important to those formulations and associated parameter data, and their application in past assessments are documented in many reports published over the past 14 years. The sheer number of reports in which such information is reported make it difficult for individuals to access this information in a timely manner. To provide users carrying out Hanford assessments with improved access to the information and data on release model capability, researchers at the Pacific Northwest National Laboratory (PNNL) consolidated these sources of information and data into a single location. This work was conducted under the Vadose Zone Groundwater Integration Project managed by Bechtel Hanford Incorporated for the U.S. Department of Energy – Richland Operations (DOE-RL). In addition to providing easier access to past information, consolidation also encourages the standardization of release model capabilities, information, and data, on future site assessments with the potential benefit of improving the intercomparibility of future assessment results.

The purpose of this report is to 1) provide a summary of descriptions and uses of release models used in assessments over the past 14 years, 2) describe the structure of selected release model mathematical formulas and assess their commonality, 3) link release models to data on various waste sources found on the Hanford Site, and 4) provide listings of sources of parameter information and parameter data used in the models. For purpose four, this report provides links to specific pages, figures, and tables for locating specific information and data within documents.

We recognize that release model capabilities continue to advance. In recognition of this dynamic, we have identified several individuals among the Hanford contractors who can be contacted for information that may be more recent than is cited in this report (see Table 1.1).

The report excludes information on release by way of the atmospheric pathway. Information on release to the atmospheric pathway from near-surface waste sources can be found in a number of previous assessments (Kincaid et al. 1998; Wood et al. 1995; Wood et al. 1996; Kincaid et al. 1993; DOE 1989; DOE 1996a, Vol. 5; Streile et al. 1996).

Table 1.1. Hanford Contacts for Information on Release Model Capability (as of 8-01)

Name, Phone, E-Mail	Affiliation	Technical Area
Steve Clark 509-372-9531 SWClark@mail.bhi-erc.com	CH2M Hill	Application of modeling (e.g., RESRAD) to Hanford cleanup activities
Charles Kincaid 509-372-6102 Charley.Kincaid@pnl.gov	PNNL	Release model applications to Hanford assessments (general knowledge of applications)
Charles Lo Presti 509-375-3923 Charles.lopresti@pnl.gov	PNNL	Development of release model capability within the Hanford Site System Assessment Capability
B. Peter McGrail 509-376-9193 Pete.McGrail@pnl.gov	PNNL	Release models for ILAW glass and high-level waste tank applications
Alex Nazarali 509-375-9432 amnazara@mail.bhi-erc.com	Bechtel Hanford Company	Application of modeling (e.g., RESRAD) to Hanford cleanup activities
Robert Riley 509-376-1935 Robert.riley@pnl.gov	PNNL	Release model applications in the Hanford Site System Assessment Capability (general knowledge of applications)
Frederick V. Roeck 509-372-9565 FVRoeck@mail.bhi-erc.com	Bechtel Hanford Company	Release model applications to Environmental Restoration Disposal Facility assessments
R. Jeff Serne 509-376-8429 jeff.serne@pnl.gov	PNNL	Release model applications to Hanford assessments (cement waste forms and general knowledge of applications)
Marcus I. Wood 509-373-3308 Marcus_I_Wood@rl.gov	Fluor Daniel Hanford	Release model applications to Hanford assessments (e.g., low-level waste performance assessments and general knowledge of applications)

2.0 Release Models Described in Previous Assessments and Reviews

Table 2.1 provides links to specific pages, tables, and figures in publications on release model capabilities relevant to or that have been applied in specific Hanford assessments over the past 14 years. The table includes only primary references to available information. The user can search for additional data by referring to publications cited in these documents. Brief summaries of information found in these sources are provided below.

Table 2.1. Sources of Information on Release Models and Data Relevant to Hanford Assessments

Subject	Specific Location Within a Reference
Release Models for Subsurface Sources	DOE 1987, Appendix P
Release Models for Specific Waste Forms	DOE 1987, Appendix P, p. P.10
Salt-Cake and Sludge in Single-Shell Tanks	DOE 1987, Appendix P, p. P.11
Liquid Release Scenario for Double-Shell Tanks	DOE 1987, Appendix P, p. P.15
Release from Grout in Vaults	DOE 1987, Appendix P, p. P.17
TRU-Contaminated Unsaturated Zone Soils in the 200 Areas	DOE 1987, Appendix P, p. P.19
Source-Release Data for Specific Waste Forms	DOE 1987, Appendix P, pp. P.37-P.41
Discussion of Source-Release Models Used on 200 Area Plateau Composite Analysis	Kincaid et al. 1998, pp. 4.2-4.6
Hanford Composite Analysis Source-Term Release Models	Kincaid et al. 1998, Appendix D
Contaminant Release Models	Kincaid et al. 1998, Appendix D, pp. D.3-D.26
Equations Used for Soil-Debris Waste Form Type	Kincaid et al. 1998, Appendix D, pp. D.10-D.13
Equations Used for the Cake Waste Form Type	Kincaid et al. 1998, Appendix D, pp. D.13-D.15
Equations Used for Glass Waste Form Type	Kincaid et al. 1998, Appendix D, pp. D.15-D.17
Equations Used for Cement Waste Form Type	Kincaid et al. 1998, Appendix D, pp. D.17-D.18
Equations Used for the Reactor Block Waste Form Type	Kincaid et al. 1998, Appendix D, pp. D.18-D.19
Rationale for Choosing Values for Radionuclide-Related Parameters in the Release Model Equations	Kincaid et al. 1998, Appendix D, pp. D.19-D.22
Composite Analysis Release Model Parameter Data	Kincaid et al. 1998, Appendix D, pp. D.27-D.29; Appendix E, pp. E.1-E.28
Conceptual Model for Source-Term Release from Low-Level Waste Disposal System	Krupka and Serne 1998, pp 1.2-1.3
Radionuclide Solubility Data Based on Speciation Modeling	Krupka and Serne 1998, pp 4.1-4.31
Distribution Coefficient Data for Selected Radionuclides on Cementitious Materials	Krupka and Serne 1998, p. 5.8

Table 2.1. (contd)

Subject	Specific Location Within a Reference
Extraction K_d Values from Diffusion and Leaching Data	Krupka and Serne 1998, Appendix E, pp E.1-E.6
Source-Term Release (congruent salt-cake dissolution release model) from High-Level Waste Tanks	DOE 1996b, Volume 4, Appendix F, pp. F-38-59
Source-Term Release (congruent glass dissolution model) from Low-Activity Waste Disposal Facility	DOE 1996b, Volume 4, Appendix F, pp. F-57-58
Residual Waste Source-Term Conceptual Model for High-Level Waste Tanks	DOE/RL 1999, Appendix A, p. A-27
Conceptual Model for Release of Contaminants from ILAW Disposal Facility	Mann 2000, pp. 25-26
ILAW Waste Form Release Rate	Mann 2000, pp. 41-43
ILAW Waste Form Release and Near-Field Contaminant Transport	Mann 2000, pp. 50-52
Conceptual Model for Dissolution of Silicate Glasses	Bacon and McGrail 2001, pp. 1-2
Dissolution Rate Equation for Glass	Bacon and McGrail 2001, p. 13
Flux Rate Equation for Release of ILAW Glass Radionuclide Constituents to the Vadose Zone	Bacon and McGrail 2001, pp. 16-17.
Release of Contaminants from Low-Activity Waste Disposal Facility (glass)	DOE 1996b, Volume 4, Appendix F, p F-57.
Overview of Modeling Approach (200 W and 200 East LLWPA)	Wood et al. 1995, pp. 3-8-3-10 Wood et al. 1996, p. 3-10 Wood et al. 1996, p. 3-12-3-14
Source-Term Analysis (release)	Wood et al. 1995, pp. 3-10-3-17 Wood et al. 1996, pp. 3-14-3-18
Advection-Dominated Release Model	Wood et al. 1995, pp. 3-12-3-13 Wood et al. 1996, pp. 3-16-3-17
Diffusion-Dominated Release Model	Wood et al. 1995, pp. 3-13-3-15 Wood et al. 1996, pp. 3-17-3-18
Solubility-Limited (constant concentration) Release Model	Wood et al. 1995, p. 3-15 Wood et al. 1996, p. 3-17
Release Rate Discussion and Curves	Wood et al. 1995, pp. 3-18-3-27 Wood et al. 1996, pp. 4-10-4-12
Release Rate (advection-dominated)	Wood et al. 1995, p. 4-22 Wood et al. 1996, p. F4-5
Release Rate (advection dominated)	Wood et al. 1995, p. 4-25 Wood et al. 1996, p. F4-6
Release Rate (diffusion-dominated)	Wood et al. 1995, p. 4-27 Wood et al. 1996, p. F4-7
Release Summary Results	Wood et al. 1995, Appendix D, pp. D-11-D-17

Table 2.1. (contd)

Subject	Specific Location Within a Reference
Selected Solubility and Sorption Data Useful to Release Models	Wood et al. 1995, Appendix E, pp. E-3-E-10
Waste Form Release Analysis and Data Collection	Wood et al. 1995, Appendix F, pp. F-3-F-10
Description of Release of Carbon-14 in Activated Metal	Wood et al. 1995, Appendix F, pp. F-3-F-4
Iodine Release Characteristics from Grout Waste Forms Under Partially Saturated Conditions	Wood et al. 1995, Appendix F, pp. F-4-F-10
Waste Configuration and Contaminant Release Scenarios (stored transuranic wastes)	Buck et al. 1996, pp. 6.1-6.8
Contaminant Release Scenarios	Buck et al. 1996, pp. 6.5-6.8
Release from Soil-Debris Wastes	Buck et al. 1996, pp. 6.5-6.8
Release from Cemented Waste Forms	Buck et al. 1996, p. 6.8
Geochemical Controls on Waste Form Leaching	Buck et al. 1996, p. 6.8
Example Contaminant-Release Calculations	Buck et al. 1996, pp. 6.12-6.15
Contaminant Property Data Useful to Release Models	Buck et al. 1996, Appendix B, pp. B.1-B.18
Conceptual Model for Contaminant Release from ERDF	DOE 1994, pp. 4-1-4-2
Constituent-Specific Parameters Important to Release Modeling (K_d, solubility and decay)	DOE 1994, pp. 4.4-4.6
Parameter Data to Release Modeling for ERDF	DOE 1994, Tables 4-1 to 4-8, pp. 4T-1-4T-8b
Release Models Used for ERDF Assessment (untreated waste)	DOE 1994, Appendix A, pp A-2-A-3
Release Models Used for ERDF Assessment (grouted or vitrified waste)	WHC 1993
Model Formulations for Contaminant Release from ERDF Untreated and Treated Waste	WHC 1993a, pp. 2-3-2-7
Model Formulation for Contaminant Travel Time through Liner	WHC 1993a, p. 2-7
Waste Release Parameters	WHC 1993a, pp. 3-5-3-9
Contaminant Specific Parameters	WHC 1993a, p. 3T-4
Probability Distributions for Input Parameters	WHC 1993a, Appendix B
Release of Contaminants from Remediated Waste Sites (RESRAD model)	Yu et al. 1993, Appendix E
Sorption-Desorption, Ion Exchange Leaching Model	Yu et al. 1993, Appendix E, pp. 197-201
Parameters/Data (K_ds) to Support Leach Model Component of RESRAD	Yu et al. 1993, Appendix E, pp. 201-206
Estimation of the Distribution Coefficient on the Basis of the Solubility Constant	Yu et al. 1993, Appendix J

Table 2.1. (contd)

Subject	Specific Location Within a Reference
Conceptual Model for Release of Contaminants from Grout Disposal Facility	Kincaid et al. 1993, Figure 3.1, p. 3.2, p. 3.6, p. 3.12, p. 3.16
Release from Grout Source-Term	Kincaid et al. 1993, p. 3.28
Initial Release Model for Grout	Kincaid et al. 1993, p. 3.33-3.38
Flow and Transport Properties of the Concrete Vault	Kincaid et al. 1993, p. 3.39-3.42
Degradation of Source-Term and Vault Materials	Kincaid et al. 1993, p. 3.41-3.53
Waste Forms (graphite and shielding) and Release Rates from Surplus Production Reactors	DOE 1989, Appendix C, pp. C.2-C.5
Release Model Options for Surplus Production Reactor Source-Term	DOE 1989, Appendix C, p. C.13
Release Rates of Radionuclides from Surplus Production Reactor Source-Term Materials	DOE 1989, Appendix D
Leaching of Irradiated Graphite and Fractional Release Rates	White et al. 1984, pp. 42-61
Corrosion Performance of Nuclear Reactor Compartment Steels	DOE 1996c, pp. 4-12-4-16
Release of Lead from Reactor Compartments	DOE 1996c, p. 4-28
Release of PCB from Reactor Compartments	DOE 1996c, pp. 4-31-4-32
Release and Migration of Lead through Soils	Rhoads et al. 1992, p. vi, 4.18, 4.28
Estimation of Nickel Release at Reactor Compartment Burial Ground	Rhoads et al. 1994, pp. iii, vi, vii, 4.33
Estimation of Nickel Corrosion Products in Hanford Groundwater	Rhoads et al. 1994, pp. iii, vi, vii and 3.20
Review of Conceptual Release Models	Serne and Wood 1990, pp. 2.1-2.9
Release Models Used in Hanford Performance Assessments	Serne and Wood 1990, pp. 2.9-2.26
Release rate from ILAW waste form	Mann et al. 1998, p. 3-8.
Contaminant Release Scenario for ILAW Waste	Mann et al. 1998, pp.3-20-3-21
Waste Form Radionuclide Release Rate for ILAW Waste	Mann et al. 1998, pp 3.38-3-41

Models were evaluated in the Hanford High-Level Defense Waste Environmental Impact Statement (HDWEIS) (DOE 1987) for use in simulating the release of contaminants from waste forms that might be disposed of in the Hanford 200 Areas Plateau. Release models consisted of four types: adsorption-controlled release, solubility-controlled release, linear release, and diffusion controlled release. For adsorption-controlled release, release was governed by the retardation factor and concentration of individual radionuclides in the solid phase. For solubility controlled release, radionuclides were carried away from their source at their maximum solution concentration (i.e., the application of radionuclide-specific solubilities based on the highest radionuclide concentrations found in tank supernate). This release mechanism is an alternative to nitrate salt dissolution and congruent release of radionuclides from the salt (Kincaid et al. 1998, Appendix D). The linear release model was viewed as a model for dissolution-controlled release of a non-decaying chemical (e.g., nitrate). A diffusion-controlled release model was considered applicable when a protective barrier was present, replacing advective transport of contaminants to the containment boundary with diffusion transport of contaminants to the containment boundary. This assumes that the surface barrier (or cover) can be shown to be so effective as to cause diffusion to dominate over advection in the region below the barrier; thus, the release is represented as a diffusion-dominated phenomena. A congruent release of radionuclides and other chemicals is assumed from the waste.

For the HDWEIS, model selection was made for specific waste forms, considering both the presence and absence of protective barriers (DOE 1987). For salt cake and sludge in single-shell tanks, without a protective barrier, the solubility model was used for radionuclides and the linear release model was used for non-decaying chemicals. For release of liquid from single-shell tanks, an adsorption model was used. For release of transuranic wastes (e.g., in low-level waste burial grounds), adsorption and solubility models were used. In the presence of a protective barrier, a diffusion-controlled model was recommended for two of the above scenarios (salt/sludge, liquid release from tanks), with a decaying source for the radionuclides and a non-decaying source for chemicals.

Serne and Wood (1990) reviewed conceptual release models (solubility-controlled, diffusion-controlled, desorption controlled, or fractionally released) as well as models applied to distinct Hanford waste forms and their associated data requirements.

Constant concentration (i.e., empirical solubility-controlled) models have been used in numerous performance assessments. Such models are necessary when identification of the likely controlling solid(s) is difficult. The solubility of a constituent is not a constant value in a chemically dynamic system. However, in empirical solubility models, a controlling solid is assumed and the chemistry of all constituents is fixed to derive a fixed value for the concentration of specific contaminants.

Diffusion-controlled release models are applied to porous solid waste forms (e.g., cemented or grouted wastes). The effective diffusion coefficient is calculated from laboratory leach test data and reflects a constituent's retardation in the matrix (i.e., by reaction with the cementitious matrix or adsorption onto matrix additives), as well as the physical hindrance in pores and the tortuosity of the matrix. Release models that use effective diffusion coefficients have been able to effectively predict the results of laboratory leach tests.

Serne and Wood (1990) also describe a desorption–controlled model that contains the attributes of the sorption component of the soil-debris model described in Kincaid et al. 1998. Inherent in the model is the reliance on a linear adsorption isotherm, the requirement of rapid desorption kinetics, the existence of only one type of species for each constituent and one type of sorption site solid matrix.

Empirical release models were described as one of the following: 1) instantaneous release of the entire inventory, 2) constant fraction release (e.g., 1% of total inventory released per year), 3) constant dissolution or corrosion rate, or 4) congruent release. For congruent release, the major constituent in the waste (e.g., salt-cake) controls the release of all other constituents. As the major constituent dissolves, all other constituents within the effected volume are released in their like proportion.

For the *200 Area Composite Analysis* (Kincaid et al. 1998), five idealized generic types of contaminant source terms (i.e., generic waste form types) were considered for release of radionuclides: soil debris, saltcake/sludge waste, glass waste, cement waste, and reactor block waste. Release was conceptualized to occur as a result of water percolating through a well-mixed waste form, with radionuclides only being lost from the source term via radioactive decay. Releases of contaminants from a soil-debris waste form were proposed to be controlled primarily by partitioning between the aqueous and solid phases. If inventory levels in the waste form type were high enough, release was considered to be solubility controlled. For the cake type waste form, release of contaminants was at a constant rate in step with the dissolving of a major structural component of the waste (i.e., nitrate salt in a high-level waste tank). For the glass waste form, releases of contaminants occurred congruent with the surface dissolution of the glass. The model took into account changes in dissolution rate as the size of the waste form shrank with time. More advanced models for simulating release of constituents from glass are described in Section 2.2. For the cement waste form type, contaminants inside the waste form were assumed to diffuse toward the outer surface, where they were released into the infiltrating water flowing past the waste form. Therefore, release from this source term was controlled by the contaminants' effective diffusion coefficient in the waste form. For the reactor block waste form, release of contaminants was described by rates calculated from experimental leach test data.

A recent literature review and calculations have provided technical support to the Nuclear Regulatory Commission's (NRC) development of performance assessment methodology for low-level radioactive waste disposal facilities (Krupka and Serne 1998). The NRC's source-term model for contaminant release from a low-level waste (LLW) disposal system requires input from water infiltration and engineered barrier calculations to determine the flux of water that may pass through the disposal unit, contact the waste forms and containers, and possibly lead to release of radionuclides. The source-term model addresses the mechanisms and rates of failure of the waste containers, if the containers are believed to delay the release of any radionuclides. Once the waste containers are breached, it is envisioned that radionuclides are released as a result of chemical reactions that occur when the infiltrating water contacts the waste forms. These reactions are affected by the composition of the infiltrating water as modified by the chemical environment associated with the waste disposal facility. This environment is envisioned as including cement, metal, and other materials present in the engineered system as well as in the waste forms, containers, and any backfill materials used in the facility.

The source-term model for LLW considers radionuclide release by either rinse release, diffusion, or dissolution mechanisms. The rinse release model assumes quantitative transfer of radionuclides can occur from the waste to the aqueous phase and, therefore, it is the most conservative of the models. It is used when radionuclides are not modeled either with diffusion release (cement solidified waste) or dissolution release (e.g., metallic waste forms). The amount of radionuclide released into the aqueous phase from any of the three source-term models, however, is limited by the thermodynamic equilibrium solubility of the radionuclide. In the real disposal system, it is understood that both chemical factors (e.g., solubility limits, sorption, and dissolution kinetics) and physical factors (e.g., matrix diffusion and limited water contact with the waste) provide constraints on the total amount of radionuclide that can actually be released to water in a facility. The chemistry of the waste form as it ages and the contact of infiltrating water over time with cementitious materials in the engineered system do result in solubility controlling the release of some radionuclides from LLW.

The Multimedia Environmental Pollutant Assessment System (MEPAS) source-term release formulations include models for estimating contaminant loss from three different types of source zones: contaminated aquifer, contaminated pond/surface impoundment, and contaminated vadose zone (Streile et al. 1996). The models have the capability of partitioning contaminants among multiple phases (e.g., aqueous solution, sorbed to solid particles, in vapor-filled pore space, or in separate non-aqueous phase liquid). The processes considered by the source-term release module are first order decay/degradation, leaching to the vadose zone or groundwater, wind suspension of contaminated surface soil particles, water erosion of contaminated surface soil particles, and volatilization from the source into the atmosphere.

Other more specific applications of release models are described below.

2.1 Release from High-Level Waste Tanks

Alternatives were evaluated for the management and disposal of waste in 177 underground storage tanks at the Hanford Site. Release of contaminant inventory from the tanks was determined based on a congruent dissolution model (salt-cake) applied following loss of institutional control (100 years). In the model, all constituents in the waste inventory were assumed to be released in proportion to the most abundant material in the waste inventory, nitrate, and at the rate of nitrate dissolution (DOE 1996b). The solubility of nitrate was assumed to be 360 g/L (Serne and Wood 1990).

The release of contaminants from tank residuals following retrieval of the high-level waste has been evaluated assuming that the structural integrity of the tanks degrades over time, allowing recharge water to enter the tank, dissolve contaminants from the residuals, and drain out into the surrounding vadose zone through cracks in the tank (DOE/RL 1999). It was assumed that essentially all the drainable liquid waste would be recovered from the tank, eliminating the potential for contaminant release for a period of time following waste retrieval and tank closure. Subsequently, infiltrating water would enter the tank. An enhanced Resource Conservation Recovery Act (RCRA) Subtitle C barrier was assumed to be constructed over the tank farm following waste retrieval and tank stabilization.

The radiological and chemical source term consisted of the inventory of a 1% residual waste remaining in the tanks after sluicing. The approach to estimating the source term parameters for the residual tank waste assumed that, over time (following closure), the liquid containment integrity of the tanks degraded and the release of contaminants occurred from dissolution by infiltrating water that migrates into and out of the tanks through cracks. This approach was in agreement with the assumed mechanism for contaminant releases from single-shell tanks following closure (Serne and Wood 1990).

A constant concentration release model (analogous to solubility) was used to develop contaminant flux rates based on post-retrieval inventory data (COGEMA 1998). Specific waste type wash factors (Colton 1995, 1996) were assumed to provide representative concentration values for how contaminants would be dissolved in infiltrating water and released over approximately 30% of the tank base area. This assumption was based on engineering judgment and available data on potential leak mechanisms (WHC 1994). For tanks in general, the best estimate values for contaminant release rates from the tank residuals were based on the empirical solubility constraints using data that were most directly applicable to the waste type contained in the individual tanks.

2.2 Release from Immobilized Low Activity Waste (ILAW)

Past modeling of vitrified waste (from a low-activity waste disposal facility) associated with the tank waste remediation system final environmental impact statement used a glass release model assuming a constant corrosion rate for the glass waste form of 3E-06 cm/yr (DOE 1996b). The applicable release model used in this assessment was likely the one described in Kincaid et al. 1998, Appendix D. The release concentration of the contaminants was assumed to be proportional to their concentration in the low-activity glass. Because the total mass loss rate is constant, the composition of the released solution is unaffected by the recharge rate. It was noted that the source term release is considered overly conservative for many contaminants because solubility controls in groundwater of neutral pH and relatively high oxidizing conditions would cause contaminants to leach at a rate less than that of nitrate or because the contaminants would be insoluble under these conditions.

The baseline concept for disposal of ILAW at Hanford has been identified as the remote handled (RH) trench (Taylor 1999). The existing vaults designed for disposal of grout waste forms may also be used for disposal of ILAW. The RH trench is a RCRA-compliant landfill (i.e., double-lined trench with leachate collection system) with a surface barrier installed at the time of closure. The RH trench and vault conceptual designs have been described in detail (Mann et al. 2000). The total ILAW waste volume is estimated to be 1.581 x 10^5 m^3 contained in 68,741 waste packages. The composition of the waste form is unspecified at this time and will likely change as wastes are retrieved from different tanks for vitrification. Data obtained from the testing of glass composition LAWABP1 is being used in current assessments of ILAW wastes.

The conceptual model for release of contaminants from the ILAW glass waste form and their transfer from the vault to the vadose zone was depicted as follows. Infiltration of moisture from precipitation enters the engineered system. The water moves toward the waste form, but most of it is diverted by the disposal system barrier. The water that is not diverted is chemically modified by the local environment and interacts with the metal canisters containing the waste form. Corrosion of the containers occurs over time. Subsequently, the canisters are breached. Water containing corrosion products from the canisters as well as constituents from the surrounding soil interacts with the waste form. The waste form (silicate glass) corrodes in the following three phases, releasing radionuclides: 1) the glass reacts with water under dilute conditions to release components of the glass into solution; 2) corrosion rates approach a very low constant value as saturated fluid conditions are approached; and 3) secondary mineral phases may form from the saturated fluid resulting (with time) in an acceleration of the forward rate of release. The moisture containing the released contaminants travels downward through the vadose zone until the contaminants reach the unconfined aquifer (Mann et al. 1998).

Dissolution of the waste form (glass) along with local chemical conditions is assumed to control the release rate of the radionuclide contaminants. The waste form release rate is evaluated (based on theoretical considerations) by modeling the basic physical and chemical processes known to control dissolution behavior instead of using empirical extrapolations from laboratory leaching experiments commonly used in other performance assessments. There is no physical constant such as a leach rate or a radionuclide release parameter that can be assigned to a glass waste form in such a dynamic system. This is because both the pH and composition of the fluid contacting the glass are affected by the flow rate, reactions with other engineered materials, gas-water equilibria, secondary phase precipitation, alkali ion exchange, and dissolution of the glass itself. A general rate equation has been formulated that describes the dissolution of glass as a function of a number of these different parameters (e.g., the amount of moisture, amount of silicic acid, pH, amount of secondary phases) (Mann et al. 2000). The normalized flux to the vadose zone for radionuclides released from the waste packages for the 2001 ILAW performance assessment that incorporates this dissolution concept has recently been described (Bacon and McGrail 2001).

2.3 Release from Solid Waste Burial Sites

2.3.1 Release of Contaminants from Low-Level Waste Burial Grounds

Release of contaminants from low-level waste cannot be modeled precisely because of the variability of the chemical and physical reactions that occur in the waste material. In the real system, radionuclides and chemicals are distributed in a heterogeneous fashion among different waste materials. Waste package containers fail at different rates because of the variability in waste material, and variable types and quantities of radionuclides and chemicals are dissolved into the infiltrating water over time depending on which waste material contacts a particular volume of water. Therefore, averaging concepts are used in modeling that simplify the mathematical representation of the real system. These concepts must be justified as being a conservative representation of the real system.

Past performance assessments of low-level waste burial grounds (Wood et al. 1995, 1996) have used three release processes (advection dominated, diffusion dominated, and solubility limited) to address contaminant release from these waste systems. The advection-dominated release model (mixing-cell cascade model, Kovak et al. 1990) was used to simulate the processes of release from unstabilized (not contained) waste. In this case, the entire inventory was immediately available for release. Neither sorption effects nor decay were factored into these calculations. For unstabilized waste, the radionuclides exited the facility at a rate determined by the flow of water and the amount of dispersion (i.e., mixing in the disposal unit, e.g., by near-field transport processes). The diffusion-dominated release model was used to simulate the release of contaminants from stabilized, contained wastes. In the absence of convection through the waste container, the release was modeled as a diffusion-limited process. Release from the waste form was represented by a diffusion coefficient. In addition to the diffusion-dominated release of radionuclides from the burial trench, an alternative approach was to specify a solubility or corrosion control limit in the waste form. Infiltration rates of 5 cm/yr were chosen for the category 1 facility and 0.5 cm/yr for the category 3 facility for different modeling scenarios. Category 1 and category 3 wastes are distinguished by their radionuclide content, as indicated in DOE (1997).

In the assessment of the 200 West Area burial grounds (Wood et al. 1995), descriptions were provided for releases of radionuclides (i.e., carbon-14 and iodine-129) from waste forms often found in low-level waste burial grounds (highly shielded containerized waste [activated metal] and grout). Information was also provided from the results of numerical release analyses (Wood et al. 1995, Appendix D) selected solubility, and sorption data (Wood et al. 1995, Appendix E), and descriptions of release of carbon-14 from activated metal and iodine from grouted waste forms under partially saturated conditions (Wood et al. 1995, Appendix F).

Buck et al. 1996 made an assessment of the consequences of the stabilization of low-level wastes at DOE sites (including Hanford) as opposed to their disposal at the Waste Isolation Pilot Plant. Two options were considered: above-ground disposal on an asphalt slab and below-ground disposal on an asphalt slab. In consideration of release of contaminants, metal buildings that would contain the waste in the surface disposal scenarios were assumed to degrade quickly (i.e., relative to the 10,000-yr evaluation period). For both scenarios, assumptions were also made that the plywood that surrounded and segregated the individual waste packages (metal drums) would degrade rapidly (i.e., relative to the 10,000 year time-frame), along with the metal drums, acting as containment for the waste packages (<100 yrs). The cement waste blocks were assumed to remain intact for the first 500 yrs and then to catastrophically fail. After the failure, the waste was assumed to act as a porous material.

Five processes were considered for release of the contaminants from these two scenarios: decay, leaching, wind erosion, water erosion, and volatilization. All of these processes were considered in the surface disposal scenario, but leaching and decay were the only processes considered for the buried-waste scenario. Both scenarios were performed without consideration of the presence of a cover and no credit was taken for the presence of a concrete or asphalt pad.

The release model formulation for the buried waste scenario was that previously applied in the 200 Area Plateau Composite Analysis (Soil-Debris model, Kincaid et al. 1998). The formulation was also modified to accommodate wind and water erosion for application to the surface waste disposal scenario. Release of contaminants from the cement waste blocks was according to the model formulation for release from cement previously applied in the 200 Area Plateau Composite Analysis (Cement model, Kincaid et al. 1998), which also accounted for radioactive decay. Infiltrating water percolating through the waste zone was assumed to not penetrate the waste forms. Rather, leaching loss is caused by this water picking up contaminants as they diffuse through the water-filled pores of the cement from the interior to the waste form surface. The source-term-release module compares the leaching mass flux calculated by the cement model with the leaching mass flux calculated by the soil-debris model where it is assumed that the waste zone was composed of soil. If the release predicted by soil-desorption control or by solubility control is lower, this value is used for the leaching mass flux.

2.3.2 Release of Contaminants from the Proposed Environmental Restoration Disposal Facility (ERDF)

Previous modeling for comparing performance assessment/risk assessment of alternative ERDF designs considered waste release mechanisms such as contaminant solubility and solid-liquid partitioning that were applied to untreated waste (i.e., contaminated soil) and dissolution and diffusion processes for vitrified waste and grouted waste. In the modeling of treated waste in ERDF (i.e., grout waste) (WHC 1993b), total release of contaminants from the waste was assumed to be the sum of releases derived from 1) dissolution or alteration of the waste matrix with subsequent release of the bound contaminants and 2) diffusion through the pore water to the waste surface where the contaminant subsequently is leached into infiltrating water. It was assumed that advective transport out of the grout waste was negligible. In the case of vitrified waste, release due to diffusional processes was considered zero (virtually no pore water in vitrified waste). Advective transport of contaminants out of vitrified and grouted waste was also considered negligible. Travel time through various liners was also evaluated. It was determined that, given the greater importance of vadose zone travel time, the advantage of accounting for diffusion through the liner was not warranted. Additional reasons given to ignore this mechanism included the computational difficulties in simulating diffusion as a plug flow process and the lack of information regarding constituent-specific diffusion coefficients (WHC 1993; WHC 1993a; DOE 1994).

2.3.3 Release from Remediated High Volume Liquid and Solid Waste Disposal Sites

The RESRAD model (Gilbert 1989) is used at Hanford to derive cleanup criteria and dose calculations for excavated high-volume liquid and solid wasted sites (BHI 1999). The model uses a time-dependent leach rate constant calculated from a retardation factor for selected constituents in the contaminated zone to determine a contaminant release rate. Typical average distribution coefficients for various elements in various subsurface materials along with representative values for the physical properties of soils used with this model have been reported (Yu et al. 1993).

2.3.4 Release from Grout Vaults

Kincaid et al. (1993) conducted an assessment of the disposal of double-shell tank waste in grout vaults on the Hanford Site. Initial release of contaminants from the grout considered simultaneous, advection, diffusion and sorption processes. Diffusion was used for those constituents that are controlled by molecular diffusion in the pore solution. Constituents with low solubility or that experience sorption at low concentrations in the pore solution were modeled using sorption/solubility constraints. Diffusion coefficients for specific species were obtained from laboratory leach tests (Serne and Wood 1990; Serne et al. 1992; Martin and Lokken 1992; Lokken 1992; Lokken, Martin, and Shade 1992) and sorption coefficients were calculated from the effective diffusion coefficients. Advection of contaminants from the grout was also assumed as a result of degradation of the engineered system over time.

2.3.5 Release from Reactor Cores of Production Reactors

In the reactor release model, irradiated solids were assumed to release contaminants (via leaching of graphite and corrosion of activated metal) into infiltrating water over time. Release of lead (a shielding component in the reactor core system) was based on a solubility-controlled release into the infiltrating water. The reactor block release model was used to simulate release from each of the surplus reactors (DOE 1989). No credit was taken in the analyses for liner or leachate collection systems. For the dosimetric analysis, it was assumed that half of the released carbon-14 and other constituents were transported by the groundwater and the remainder was assumed to be transported directly out of the burial site into the atmosphere. For the various scenarios, a post-disposal assessment period of 10,000 years was evaluated.

2.3.5.1 Release Rates for Graphite

A release function was developed for carbon-14 release from graphite. (The key reaction was assumed to be carbon reacting with oxygen in the air dissolved in the water.) A rate of 2.2×10^{-12} g/cm^2/day was determined (wet storage conditions) (Gray 1982). The resulting release would extend over 23,000 years. White et al. (1984) studied the leaching of carbon-14 from demineralized water and calculated a release rate of 5.5 x 10^{-7} cm/day at 22 °C, which is in good agreement with the results of Gray (1982). White et al. (1984) was able to develop an equation to describe the release rates from irradiated graphite in saturated groundwater as a function of time and temperature. Release rates under dry storage conditions were related to release rates under wet storage conditions, based on the relative humidity of the surrounding air. Carbon-14 release rates from irradiated metals are unknown. For activation products in metal components in the reactor block, release rates can be equated with corrosion rates (see below).

2.3.5.2 Release Rates of Radionuclides (other than C-14) from Graphite

Release rates of radionuclides other than carbon-14 have been reported by White et al. (1984) for tritium, cesium-134, cobalt-60, barium-133, iron-55, europium-154, and chlorine-36.

2.3.5.3 Release Rates from Shielding and Metal Components

The iron shielding and aluminum components of the reactor contain activation products that are subject to release as corrosion occurs. To provide a release rate for these radionuclides, iron was assumed to corrode at a rate of 5 mils (0.005 in./yr) or 3.5 x 10^{-5} cm/day. Based on this corrosion rate, the radionuclides would release over a period of 390 years (DOE 1989). Aluminum components were assumed to corrode at a rate of 0.1 mil/yr, yielding a release rate of 7 x 10^{-7} cm/day. Based on this corrosion rate, the radionuclides in the aluminum would release over a period of 1,250 years (DOE 1989).

2.3.5.4 Release Rates of Lead from Shielding

Water was assumed to reach a solubility limited lead concentration of 0.29 mg/L. Lead migration would be very slow. Predicted peak concentrations would not occur for between 4.5 million and 45 million years for disposal in the 200 West Area and between 200,000 and 10 million years for disposal in the 100 Area (DOE 1989).

2.3.5.5 Releases of Contaminants from Naval Reactor Compartments

Radioactivity in the compartments of decommissioned Naval reactors is primarily in the form of corrosion-resistant activated metals (i.e., nickel-63, carbon-14, niobium-94, nickel-59, selenium-79, and technetium-99) that make up the hull and internal structure of the reactor pressure vessel. The Navy estimated that more than 99% of the inventory is found in these metals (Wood et al. 1996). Also present within the reactor vessel are polychlorinated biphenyls (PCBs) used for thermal shielding and lead (used for radioactivity shielding). After 500 years, only nickel-63 remains. High-strength carbon steels and very-high-tensile strength alloyed steels form the exterior of reactor compartment disposal packages (DOE 1996c).

A previous study (Rhoads et al. 1994) considered the disposal of a group of 120 reactor compartments at a 200 East Burial Ground as a potential nickel radionuclide source due to the presence of metal alloys inside the compartments that contain activated nickel (nickel-59 and nickel-63). The compartments were modeled with average quantities of nickel alloy and activated nickel, based on total inventories found in reactor compartments. Nickel radionuclides were modeled as activated constituents of corrosion resistant steel and steel alloys. Recharge (0.1, 0.5 and 6 cm/yr) passing through this area was assumed to contact the reactor compartment and exit saturated with nickel. Corrosion rates used were 0.0001 mg/cm^2/yr and 0.0002 mg/cm^2/yr, respectively, for the different corrosion resistant steels. It was found that nickel-63 would decay to negligible levels (1 x 10^{-10} pico Curies per liter [pCi/L]) prior to reaching the aquifer, even under the postulated wetter condition (0.5 cm/yr versus 6 cm/yr infiltration rate).

In a similar study (Rhoads et al. 1992), the release and migration of lead from the reactor compartments was also estimated. As with nickel, average lead quantities were used. Lead was very conservatively assumed to be immediately available for dissolution, so that all groundwater contacting the 15.2 x 15.2 m^2 reactor compartments would exit the area being fully saturated with dissolved lead. Lead solubility was set at roughly double experimental results.

A solubility-limited concentration of 15 ppb was used to simulate the release of PCBs from reactor compartments and to assess impacts to the Columbia River. Downstream concentrations of PCBs in the aquifer would be less than 0.5 ppb for the postulated wetter condition and less than 0.1 ppb for the current climate (DOE 1996c).

2.3.6 Releases of Contaminants from Process Facilities (canyons and tunnels)

Documented work on release of contaminants from process and storage facilities is unknown. Kincaid et al. (1998) chose to exclude such source terms from the 200 Area Composite Analysis based on 1) the absence of data on radionuclide inventories for these facilities and 2) the fact that these facilities appear to retain excellent waste performance characteristics for stabilization of radionuclide contamination. For the latter point, it was assumed that it was unlikely that such facilities would be a significant source of groundwater contamination, especially in the next 1,000 years. One exception to this exclusion was the modeling of the release of cesium-137 and strontium-90 inventories from B-Plant and B-Plant filters which, under a most conservative release scenario, showed no release of these constituents to the groundwater within 1,500 years. Decommissioned process facilities would consist of waste, both internal and external to entombed concrete structures. Future modeling of contaminant release from such waste sources might consider employing a combination of cement and soil-debris models.

3.0 Release Model Mathematical Formulations and Data Sources

An assessment of the publications identified in Table 3.1 indicated that model formulations used over the past 15 years to simulate contaminant release from some of the more common waste forms have been relatively constant in their depiction. Formulations used in the recent 200 Area Plateau Composite Analysis (Kincaid et al.1998) appear to have been commonly used. General descriptions of these models are summarized in the subsections below, along with associated parameter definitions and their units. Analytical solutions for each model are limited to those that describe contaminant release as a function of time and do not include a term for decay ($e^{-t\lambda}$).

Table 3.1. Relationship Between Selected Release Formulations Found in Kincaid et al. (1998) and Those Found or Assumed Used in Other Referenced Sources. A blank cell means no model available for comparison.

Other Sources	Kincaid Listed Release Model Formulations			
	Soil-Debris	Salt Cake	Cement	Glass
DOE 1987, Equation P.6, p. P.3; Equation P.17, p. P.4	Same			
DOE 1987, Equations p.21 and p.22, p. p.5		Same		
WHC 1993, Equation 2-1, p. 2-3 (untreated waste)	Same			
WHC 1993, Equation 2-5 (treated waste)			Same	
Wood et al. 1995, Equation 3.5 and 3.7, pp. 3-12-3-13	Same		Same(a)	
Wood et al. 1996, Equation 3,5 and 3.7, pp. 3-16-3-17	Same		Same(a)	
Buck et al. 1996, Equation 6.3 and 6.5, p. 6.6	Same		Same(b)	
Serne and Wood 1990, Equations 1 and 6, p. 2.4	Same(c)	Same	Same	
DOE 1996b, Appendix F, p. F-39		Same		
Mann et al. 1998, Equation 3.9, p. 3-40				Same as Kincaid et al. 1998, App D, Eq. D.5
Bacon and McGrail 2001, pp. 16-17				Not the same

(a) The model is called a mixing-cell cascade model and has the same formulation as the soil-debris model with the exception of the addition of a dispersion term.
(b) Equation 2.5 of Buck et al. 1995 factors in the effects of soil erosion.
(c) Serne and Wood 1990, p. 2.13, report the use of a congruent release model for release of radionuclides.

Possible applications of the release models to various Hanford waste source terms are summarized in Table 3.2. Assignment of a sub-set of these models to some of these source terms was made in performance of the initial run of the System Assessment Capability (Riley and Lo Presti 2001).

Table 3.2. Summary of Possible Source-Term Release Model Assignments to Waste Source Types and Associated Assumptions

Release Model	Waste Source Type
Liquid	Single shell tank past leaks and future losses,(a) unplanned releases,(b) trenches,(c) cribs,(c) drain/tile fields,(c) radioactive process sewers, french drains, retention basins, ponds, ditches, sumps, sand filters, injection/reverse wells, storage tanks, diversion boxes, catch tanks, valve pits, settling tanks, receiving vaults, neutralization tanks
Soil-debris	Unplanned releases,(b) burial grounds, laboratories, storage, stacks, landfills, decommissioned reactor compartments(d)
Cement	Process units or plants, control structures, storage tunnels
Salt-cake	Single-shell tank(a) and double-shell tank(e) residuals
Reactor block(f)	Surplus production reactor graphite cores
Glass	Vitrified immobilized low-activity waste
Corrosion	Decommissioned Naval reactor compartments(g)

(a) Releases from single-shell tanks have been modeled using a combination of liquid and salt cake models. Releases include past tank leaks, liquid released during retrieval and contaminant release from dissolution of residual solids following waste retrieval completion.
(b) Modeled as initial liquid release, release from a surface contaminated soil or a combination of both.
(c) Radionuclides from 216-Z-1A drain/tile field, 216-Z-9 trench and 216-Z-18 crib are modeled as liquid release. Carbon tetrachloride from these sites has been modeled as release from entire vadose zone profile using soil-debris model.
(d) Several chemicals (Cr^{+6}, lead and PCB) are found in reactor compartments. Following corrosion of the reactor hull, such constituents would be release based on solubility or sorption controls.
(e) Double shell tanks are assumed not to leak prior to and during retrieval. Release of contaminants from residual solids modeled using salt cake model.
(f) B reactor release occurs entirely in the 100 Area. Following a specified period of time (75 years) remaining inventories for all other reactors moved to 200 West Area burial ground (218-W-5) where release continues using the reactor block model.
(g) Activation products (e.g., nickel-63) are contained within the hull steel and would be released to the infiltrating water as corrosion of the steel occurred.

Table 3.3 summarizes the key parameters for the release models and refers the user to tables appearing below in this report that link the user to the relevant source documents for each model and parameter. In the discussion that follows, the user is provided a brief description of information associated with each table along with any important points that need to be noted regarding parameter or data applications to specific release models.

Table 3.3. Parameters and Data Sources for Release Models[(a)]

Model Parameter	Release Model Type Liquid	 Soil-Debris	 Salt-Cake	 Cement	 Reactor Block	 Glass	 Corrosion[(b)]
Cross sectional area of source zone	©	Table 3.7	Table 3.7				
Depth of waste		Table 3.7					
Distribution coefficient		Table 3.4					
Bulk density		Table 3.5					
Volumetric moisture content		Table 3.5					
Solubility		Table 3.6	Table 3.10				
Fractional release					Table 3.12	Table 3.13	Table 3.14
Recharge rate	Table 3.8	Table 3.8	Table 3.8				
Tank solid waste density			Table 3.10				
Waste surface area				Table 3.7		Table 3.7	Table 3.7
Waste volume				Table 3.7		Table 3.7	
Diffusion coefficient				Table 3.11			
Dissolution rate				Table 3.11		Table 3.13	
Corrosion rate							Table 3.14
Mass of structural component in source zone			X				X
Mass or activity of contaminant	X	X	X	X	X	X	X
Temperature					Table 3.12		
Distance from soil surface to bottom of contaminant source zone		Table 3.7					
Water erosion rate		Table 3.9					
Wind suspension rate		Table 3.9					
Dispersion coefficient		Table 3.9					

(a) Fields marked with an X indicate information that is required by the specified release model but is not documented in this report. A key source of such information would be the System Assessment Capability (SAC) inventory database.
(b) Model developed from information in Rhoads et al. (1994), pp. vi and 4.2.
© Blank fields indicate this data are not required by the specified release model.

3.1 Soil Debris Model

3.1.1 Mathematical Formulations for Soil Debris Model

The rate of loss of contaminant for a given contaminant by the soil-debris model is given by:

$$dM\ /\ dt = -\,Q_w A C_w \qquad \text{(Equation D.35, p. D.11, Kincaid et al. 1998)}$$

where $C_w = C_{sol}$ in Equation D.35 when the release process is solubility-controlled
$Cw = M/(\theta RAh)$ in Equation D.35 when the release process is desorption-controlled where:

$$R = 1 + (\beta K_d)\ /\ \theta$$

Switching régimes is controlled by comparing the remaining mass with the maximum mass M_{max} consistent with an aqueous phase saturated with the contaminant. If M, the mass remaining in the waste form is larger than the quantity M_{max} where:

$$M_{max} = \theta R C_{sol} A h$$

the release process is considered to be solubility controlled; otherwise it is desorption controlled

Definitions:

- M_{max} is the maximum amount of contaminant possible in the source zone (in Ci or kg) without a precipitated phase.
- M = M(t) is current quantity of contaminant contained in the source zone (Ci or kg)
- Q_w is recharge rate for the site in cm/yr. Q_w can be considered constant, or it can be time-dependent based on site climate and remediation activities.
- A is surface area of the soil waste form exposed to the release mechanism (cm^2)
- h is depth of the waste form in the site (cm).
- C_w is a coefficient expressing the effective release of the contaminant (Ci/cm^3 or kg/cm^3)
- C_{sol} expresses aqueous solubility of the contaminant in Ci/cm^3 or kg/cm^3
- R is either a retardation factor or a soil apportionment factor (unitless) which depends on several factors:
  - ß Soil bulk density in g/cm^3
  - K_d Sorption factor (cm^3/g)
  - θ Soil volumetric content of water in soil (unitless fraction)
- dM/dt is the rate of loss of contaminant from the source zone (the rate contaminant crosses the soil waste form boundary and enters the environment)
- t is the elapsed time (years) from the beginning of release from containment.

Buck et al. (1996) took into account the effects of water erosion and wind suspension on contaminant release to the vadose zone from burial ground wastes. For this case, the soil debris equation takes the form:

$$dM / dt = - Q_w M / \theta_w R\left[h_o - (E + S)t\right]$$

where h_o = the initial distance from the soil surface to the bottom of the contaminant-source zone (cm)
E = the volumetric rate of soil removal by water erosion, per area (cm/yr)
S = the volumetric rate of soil removal by wind suspension, per area (cm/yr).

3.1.2 Sources of Data for Soil Debris Model

Sources of data for the Soil-Debris Model are provided in the text and tables below.

3.1.2.1 Distribution Coefficients for Soil-Debris Model

Table 3.4 summarizes distribution coefficient data potentially applicable to the soil-debris model. Since application of the model is to the near field, recent efforts have focused on using values in the model that reflect a specific waste type and waste zone (high impact) (Riley and Lopresti 2001) as initially developed for the 200 Area plateau composite analysis (Kincaid et al. 1998). In many cases, particularly with organic constituents, such specificity is not available. In such cases, distribution coefficient values applied to vadose zone or ground water modeling may also be applied to the release model.

Table 3.4. Sources of Distribution Coefficient (K_d) Data for Soil Debris Model

Information Source Summary	Reference
Distribution coefficients for the soil debris model as applied to the System Assessment Capability initial run (SAC Rev. 0) are found here. Those applied were those for the high impact zone for six source categories as defined in Kincaid et al. (1998), Appendix E, Table E.4.	Riley and Lo Presti (2001) http://www.bhi-erc.com/vadose/sac.htm#info
Definitions of and the rationale for development of zone categories and source categories for K_d values are provided. Conservative, best estimate, and range estimates of K_d's for different radionuclides are provided that were used in the 200 Area plateau composite analysis. Relevant K_d's for release are the high impact zone category for the six source categories.	Kincaid et al. (1998), Appendix E, Tables E.2 through E.17
Partition coefficients used in a screening performance/risk assessment of the Environmental Restoration Disposal Facility (ERDF)	WHC (1993a), pp 3-7-3.8; Table 3-4
Probability distributions for values of K_d for constituents of concern in the ERDF screening performance/risk assessment	WHC 1993a, Appendix B, p B-8
K_d values used in a fate and transport model to predict groundwater concentrations at the ERDF boundary. K_d values were calculated to be K_{oc} X 0.001 organic content for organic constituents.	DOE/RL 1993,Tables 4-2, 4-3, and 4-4
K_d values used in the release source term for the Waste Isolation Pilot Plant no-action alternative	Buck et al. 1996, Appendix B, Table B.3
Retardation factor for advection-dominated release model set to 1 for disposal units for the 200 West and East area performance assessments	Wood et al. 1995, p. 3-12 Wood et al. 1996, p. 3-16
Preferred distribution coefficients for selected radionuclides for cement/concrete environments	Krupka and Serne 1998, p. 5.8
Desorption (R_d) values for contaminant release from soils and solid wastes	Serne and Wood 1990, Appendix A, Table A.3
Distribution coefficients for desorption-controlled release from contaminated soils	Serne and Wood 1990, Table 6, p. 2.25
Distribution coefficients for selected metals and radionuclides used in the RESRAD model	Yu et al. 1993, pp. 202-205

3.1.2.2 Bulk Density and Moisture Content Data for Soil Debris Model

Table 3.5 summarizes sources of bulk density and moisture content data for the soil-debris model. The model is generally applied to source zones at Hanford that are within 20 feet of the ground surface. The greatest amount of data for bulk density and moisture content within 20 feet of the surface is found for 200 East Area soils (Fayer et al. 1999). Considerably less data is found for soils in the 100 and 300 Areas (Peterson et al. 1996; Schalla et al. 1988). No source of such data has been found for 200 West Area soils. Most recently, data from these sources were used to calculate bulk density and volumetric moisture content values as a function of depth for the 100, 200 West, 200 East, and 300 Areas for application to waste sources using the soil-debris model for the initial run of the system assessment capability (Riley and Lo Presti 2001). Some of the data in Table 3.5 is documented as percent moisture. The parameter in the model requires volumetric moisture content, which can be calculated knowing the percent moisture and the bulk density of the soil (volumetric moisture content = vol. of water in sample divided by ([dry wt of soil/bulk density] + vol. of water).

Table 3.5. Sources of Bulk Density and Moisture Content Data for Soil Debris Model

Information Source Summary	Reference
Bulk density and volumetric moisture content data for the soil debris model as applied to the System Assessment Capability initial run (SAC Rev. 0) are found here. Data were calculated from some of the sources described below	Riley and Lo Presti 2001 http.//www.bhi-erc.com/vadose/sac.htm#info
Volumetric moisture content and bulk density distributions for Hanford formation soils are presented.	Engleman et al. 1995, pp. 3-5.
Bulk density and moisture content data with depth presented for boreholes in the 200 East Area	Fayer et al. 1999, Tables B.3-B.9
Bulk density and moisture content data with depth for the Hanford Site's 100 B/C, 100 K, 100 D/DR, 100 H, and 100 F areas	Peterson et al. 1996, Tables 3-8, 4-9, 6-9, 7-9, and 8-8
Moisture content data with depth are provided for soils from the 300 Area	Schalla et al. 1988, Appendix B, Page B.2, Table B.2

3.1.2.3 Aqueous Solubility Data for Soil-Debris Model

Table 3.6 summarizes sources of solubility data that can be used in a constant concentration release of a contaminant using the soil-debris model.

Table 3.6. Sources of Solubility Data for Soil Debris Model

Information Source Summary	Reference
Solubility data for the soil debris model as applied to the System Assessment Capability initial run (SAC Rev. 0) are summarized	Riley and Lo Presti 2001 http.//www.bhi-erc.com/vadose/sac.htm#info
Aqueous solubility data for selected radionuclides used in 200 Area plateau composite analysis	Kincaid et al. 1998, Appendix D, Table D.2
Aqueous solubility data for selected metals in cement leachates and Hanford groundwater	Wood et al. 1995, Appendix E
Calculated aqueous solubilities for selected radionuclides and metals for Hanford's 200 West and East Areas as applied to the Waste Isolation Pilot Plant No-Action Alternative	Buck et al. 1996, Appendix B, Table B.2
Aqueous solubilities for organic, inorganic and radionuclide constituents used in Environmental Restoration Disposal Facility remedial investigation feasibility studies	DOE 1993a, Tables, 4.6, 4.7, and 4.8
Aqueous solubilities for organic, inorganic and radionuclide constituents used in Environmental Restoration Disposal Facility screening performance assessment/risk assessment	WHC 1993a, p. 3-9, Table 3-4
Radionuclide solubility data for Nuclear Regulatory Commission performance assessment test case	Krupka and Serne 1998, pp. 4.1-4.31
Radionuclide solubility data for TRU waste no disposal action scenario	DOE 1987, Volume 3, Appendix P, Table P.26
Solubility of PCB in water for reactor compartment assessment	DOE 1996c, p. 4-31
Aqueous solubility of lead for assessment of reactor compartment source-term	Rhoads et al. 1992, Table 3.7, p. 3.19
Solubility values for solubility-controlled release from contaminated soils	Serne and Wood 1990, p. 2.25

Values of aqueous solubility are derived from experimental measurements or estimated based on geochemical calculations (e.g., using the MINTEQA2 computer code). In cases where the solubility of a constituent is unknown, the aqueous solubility can be fixed at an arbitrarily high default value forcing the soil-debris model to operate in the desorption-controlled mode. In many cases, constituents without solubility values are not solubility-limited in aqueous solution. The soil-debris model may require solubility values in units of Ci/cm^3. In this case, solubility values measured in mg/L are converted to Ci/cm^3 by multiplying by the specific activity of each radionuclide (along with appropriate unit conversion factors). The specific activity was calculated from the decay half-life and the atomic mass according to the formula:

$$A = 3.578 \text{ X } 10^5 / t_{1/2} M \text{ (DOHEW 1970)}$$

where A = the specific activity of the contaminant (Ci/g)
$T_{1/2}$ = the decay half-life (yrs) of the contaminant
M = the atomic mass of the contaminant (g/mol).

3.1.2.4 Other Data for the Soil Debris-Model

The soil-debris model requires cross sectional area and height (depth) information for the waste source zone. This data can vary significantly depending on the dimensional scale to which the modeling is being performed. For example, the model may be applied to a specific waste site footprint (e.g., a low-level waste burial ground) in which case the data would consist of the dimensions of the waste site). In another case, low-level waste burial grounds within a given Hanford area (e.g., 200 West Area) may be aggregated and a cross sectional area footprint assigned that is the sum of the cross-sectional areas for all of the low-level waste burial grounds in the 200 West Area. This type of scaling is most recently observed in the application of the soil-debris model to the System Assessment Capability initial assessment (Riley and Lo Presti 2001). Feature data (i.e., dimensions) for specific source zones and model source zones for the soil-debris model and the other release models (Sections 3.2, 3.3, and 3.4) can be found in Table 3.7.

Table 3.7. Sources of Waste Zone or Waste Form Feature Data for Release Models

Information Source Summary	Reference
Cross-sectional areas (and/or length/width) and height (depth) data for the soil debris model as applied to the System Assessment Capability initial run (SAC Rev. 0) is summarized	Riley and Lo Presti 2001, http://www.bhi-erc.com/vadose/sac.htm#info
Cross-sectional areas (and/or length/width) and height (depth) data for Hanford Site waste source zones	WIDS database
Cross-sectional areas (and/or length/width) and height (depth) data for Hanford Site waste source zones	Stenner et al. 1988, Volumes 2 and 3 (HISS database)
Waste site volumes (as a substitute for cross-sectional area X height) for application of release models to 200 Area Plateau Composite Analysis	Kincaid et al. 1998, Table 4.3
Model waste configurations (height, cross-sectional areas, volumes) for buried TRU waste at Hanford as applied to Waste Isolation Pilot Plant No-Action Alternative	Buck et al. 1996, pp. 6.1-6.4
Environmental Restoration Disposal Facility and trench dimensions	DOE 1994, p. 4-2, Figure 4-1; WHC 1993a, p. 3-3, Figure 3-3
Geometry data for TRU sites at Hanford	DOE 1987, Volume 3, Appendix P, pp. p.20 to p.21; Table P.26, p.40
Treatment of disposal facility features in low-level waste performance assessment analysis (200 West Area)	Wood et al. 1995, pp. 2-32-2-36, Figure 2-15; p. 3-27
Treatment of disposal facility features in low-level waste performance assessment analysis (200 East Area)	Wood et al. 1996, pp. 2-21-2-22, Figures 2-12, 2-13, 2-14, 2-15, 2-16, and 2-17
High-level waste tank cross-sectional areas for the salt cake model as applied to the System Assessment Capability initial run (SAC Rev. 0)	Last et al. 2001 http://www.bhi-erc.com/vadose/sac.htm#info

Table 3.7. (contd)

Information Source Summary	Reference
High-level waste tank cross-sectional areas (calculated from diameter data)	WHC 1994b, Appendix D, p. D-3
Reactor compartment (decommissioned nuclear submarine and cruiser compartments) footprint information	DOE 1996c, Figure 2.1, p. 2-7
Reactor compartment (decommissioned nuclear submarine and cruiser compartments) burial ground configuration information	DOE 1996c, Figures 2.10 and 2.12, pp. 2-19 and 2-21
Model footprint for release of lead from reactor compartments	Rhoads et al. 1992, p 4.1-4.2
221 U facility footprint (also model footprint for B-Plant and T-Plant)	DOE/RL 1998, Figures 1-3 and 1-5
Grout disposal vault features	Kincaid et al 1993, p. 2.70-2.78
High integrity container (HIC) designs and features	Josephson 1996, p. 2-3
Low-level waste burial ground waste packages	Wood et al. 1995, p. 2-30; Wood et al. 1996, p. 2-20
Cross sectional areas for fuel storage basins and ground disposal sites associated with surplus production reactors	DOE 1989, Appendix C, p. C.4
Design of disposal of surplus production reactors	DOE 1989, Appendix H, p. C.4
Surplus production reactor design data (graphite stack, process tubes, thermal shield, biological shield)	Miller and Steffes 1987, p. 11, Table 2
Dimensions of decommissioned nuclear reactor compartment waste packages	DOE 1996c, Figure 2.1, p. 2-7
Dimensions of Trench 94 in 218-E-12B low-level waste burial ground	DOE 1996c, Figures 2.10 and 2.12, p. 2-19 and 2-21
Dimensions used to model release of constituents from Trench 94 in 218-E-12B burial ground	DOE 1996c, Figure 4.2, p. 4-18 and Figure 4.3, p. 4-21
Waste package arrays for decommissioned nuclear reactor compartment waste packages	Rhoads et al. 1994, p. 4.1; Rhoads et al. 1992, p. 4.16
Surface areas associated with decommissioned nuclear reactor compartment arrays	Rhoads et al. 1994, p. 4.2
Remote handled trench and concrete vault conceptual designs for ILAW waste	Mann et al. 2000, pp. 8-12, Figures 2.3, 2.4, 2.5, 2.6, and 2.7
ILAW waste package geometry	Mann et al. 2000, pp. 40-41
Material zones for remote handled trench and concrete vault	Mann et al. 2000, pp. 53-55
Estimate number of waste packages for ILAW waste	Mann et al. 2000, p. 20
Area-to-volume ratio for 55-gallon drum	Wood et al. 1996, p. 3-18
ILAW disposal designs	Mann et al. 1998, pp. 2-51-2-57, p. 2-61
ILAW vault dimensions	Mann et al. 1998, pp 3-43-3-44
ILAW vault and trench dimensions for simulations	Bacon and McGrail 2001, p 17

The soil-debris model has a recharge rate term. In general, recharge rates applied are those used in vadose zone modeling and vary based on site conditions (e.g., soil type, presence or absence of a cover). Recharge rate data sources are summarized in Table 3.8.

Table 3.8. Sources of Recharge Rate Data for Release Models

Information Source Summary	Reference
Estimated recharge rates as applied to the System Assessment Capability initial run (SAC Rev. 0) are summarized	Last et al. (2001) http://www.bhi-erc.com/vadose/sac.htm#info
Estimated recharge rates for ILAW performance assessment	Fayer et al. (1999), pp. iii-iv; p 2.2
Recharge rates for low-level waste performance assessments	Rockhold et al. (1995)
Variations in recharge at the Hanford Site	Gee at al. (1992)
Recharge rates for low-level waste performance assessment in 200 West Area	Wood et al. (1995), pp 3-16-3-17
Recharge rates for low-level waste performance assessment in 200 East Area	Wood et al. (1996), pp 3-19-3-20
Infiltration rates for Environmental Restoration Disposal Facility RIFS assessments	DOE (1994), pp. 4-2-4-3; WHC (1993a), p. 3-1
Infiltration rates for HDWEIS	DOE (1987), Appendix Q, pp Q.1 to Q.2
Infiltration rates for TWRS EIS remediation scenarios	DOE (1996b), Appendix F, pp. F-39-F-63
Estimated recharge rates as applied to the System Assessment Capability initial run (SAC Rev. 0)	See Appendix I, Table I.2
Recharge for Hanford grout performance assessment	Kincaid et al.(1993), p. 4.56
Recharge rate for RESRAD modeling of Hanford Site 116-C-1	BHI (1999), pp. 35 and 37
Recharge rates for modeling release of contaminants from decommissioned nuclear reactor compartments	Rhoads et al. (1994),p. ix; p. 4.3; p. 4.34-4.35
Recharge rate estimates for ILAW waste	Mann et al. (2000), p. 45
Recharge rates for single-shell tanks	Serne and Wood (1990), p. 2.13
Natural recharge rates and infiltration rates for ILAW assessment	Mann et al. (1998), pp. 2-37-2-38; pp. 3-48
Recharge rate for ILAW simulations	Bacon and McGrail (2001), p. iii
Recharge data for S-SX Field Investigation	Khaleel et al. (2000), pp. 5-6

Some previous assessments have modified the soil-debris model to take into account other factors that effect contaminant release from a source zone. Such factors include soil erosion due to wind and water (Buck et al. 1996) and hydrodynamic dispersion (Wood et al. 1995). Table 3.9 summarizes sources of such data and how they are incorporated into the conventional soil-debris model formulation.

Table 3.9. Other Sources of Data for Soil Debris Model

Information Source Summary	Reference
Wind and water erosion rates for Hanford Site soils	Buck et al. 1996, p. 7.2
Dispersion coefficient for advective dominated release model	Wood et al 1995, pp. 3-12-3-13

3.2 Salt Cake Model

The salt-cake model consists of a very simple mathematical formulation containing a recharge rate term, a term for waste solid solubility, and a term for the cross-sectional area of the waste source (i.e., single and double-shell tank footprint).

The contaminant release mechanism of the salt-cake model is the dissolution of the structural matrix. As the matrix dissolves, all the contaminants are assumed to leach congruently at the same rate. When applied to the Hanford high-level waste tanks, the term “salt-cake” applies to the salt-cake, sludge, and hard heel residual in the tanks, which compose the “structural matrix.” The release rate for a given contaminant is given by:

$$dM\ /\ dt = M_o\ A\,Q_w\ C_{wo}^{sol}\ /\ M_{wo} \qquad \text{(after Equation D.48, p. D.14, Kincaid et al. 1998)}$$

where M_{wo} = the original mass of salt cake (kg). M_{wo} may also be derived by the product of tank waste volume and waste density.
M_o = the original quantity of contaminant in Ci or kg embedded in the salt-cake.
M = M(t) is the current quantity of the contaminant contained in the salt-cake (Ci or kg) at time t
A = the surface area of salt-cake exposed to the release mechanism (cm^2)
C_{wo}^{sol} = the aqueous solubility of the salt-cake simulated as a nitrate salt (g/cm^3)
Q_w = the site recharge rate in cm/yr, also termed “infiltration rate.”
dM/dt = the rate of loss of contaminant from the salt-cake waste form per unit time t (the rate at which the contaminant enters the environment).

Recharge rates for the salt-cake model are handled in a similar fashion to the soil-debris model. Sources of data on recharge and cross-sectional area for this model can also be found in Tables 3.7 and 3.8, respectively. Cross-sectional footprints for the salt-cake model can consist, for example, of an individual tank, a tank farm, or a cluster of tank farms in a specific Hanford area.

In many cases, the dissolving solid is considered to be a nitrate salt and contaminants imbedded in the solid dissolve congruently with the nitrate. Nitrate concentrations measured in tank high-level waste pore fluids and supernate and used in Hanford assessments have varied. The concentration most commonly used today is 360 mg/L. A density value is required to convert tank waste volumes to equivalent masses.

(see Table 3.10). An alternative to congruent release depicted by the salt-cake model is solubility-controlled release where radionuclides are carried away from the source at their maximum solution concentration (Serne and Wood 1990, Appendix A, Table A.1).

Table 3.10. Sources of Data for Salt Cake Model

Information Source Summary	Reference
Nitrate salt solubility as applied to the System Assessment Capability initial run (SAC Rev. 0) is summarized	Riley and Lo Presti 2001 http://www.bhi-erc.com/vadose/sac.htm#info
Nitrate salt solubility concentrations found in high-level waste tank drainable liquors	Serne and Wood 1990, pp. 2.27 and 2.33
Nitrate salt solubility concentration used in TWRS EIS release model simulations	DOE 1996, Appendix F, pp. F-39-F-63
Tank solid waste density	Chen et al. 1998, Table 3.6, p. 3.18

3.3 Cement Model

The cement model is generally applied to cementitious waste forms. A knowledge of the total external surface area and the volume of the waste form are required. The ratio of area to volume is assumed to be constant, that is, the waste form is assumed not to degrade in terms of shape over the duration of the contaminant release process.

The contaminant release mechanism of the cement model is diffusion in the pore water of the solidified waste material to the outer surface of the waste form. The rate of loss of contaminant for a given contaminant is given by:

$$dM / dt = (A / V)\sqrt{D / \pi t}$$

(Equation D.61, p. D.17, Kincaid et al. 1998)

where M_0 = the original quantity of the contaminant contained in the cement (Ci or kg)
This can be seen as a function of concentration (kg/cm^3 or Ci/cm^3) and volume (cm^3)
M = current quantity of the contaminant contained in the cement (Ci or kg)
A = the surface area of the cement structure (cm^2)
V = the volume of the cement structure (cm^3)
D = the diffusion coefficient of the contaminant (cm^2/yr)
t = the elapsed time (years) from the beginning of release from containment
dM/dt = the rate of loss of contaminant from the cement waste form.

Data for parameters for the cement model can be found in Table 3.7. The most important term in the model is the effective diffusion coefficient, which governs the migration of contaminants from inside the waste form to the waste form surface where they are removed by infiltrating water. It is assumed that cementitious waste forms have sufficient permeability to allow the diffusion process to occur in the waste form pore water. Most effective diffusion coefficients are derived from experiments performed under saturated moisture conditions. Application of such coefficients in release models would result in faster contaminant releases to the vadose zone than would be anticipated at the Hanford Site. Most recently, diffusion coefficients for selected radionuclides have been determined for unsaturated conditions more representative of the Hanford Site (Mattigod et al. 2001).

Competing with diffusion of contaminants out of the waste form is dissolution or corrosion of the cementitious material with subsequent release of contaminants to the surrounding environment. Geochemical modeling of the dissolution of cementitious materials suggests slow degradation of cement materials in a radioactive waste disposal system environment. Pore fluid composition of pH above 10.5 is predicted for several hundred thousand years during dissolution of the calcium silicate hydrogel (C-S-H) and Portlandite phases of cementitious materials. These phases constitute up to 75 wt% of the cement. A study of natural analog systems indicates the stability of cementitious materials on time frames of thousands of years (Krupka and Serne 1998). Sources of diffusion coefficient and dissolution rate data can be found in Table 3.11.

Table 3.11. Sources of Data for Cement Model

Information Source Summary	Reference
Diffusion data for the cement model as applied to the System Assessment Capability initial run (SAC Rev. 0) are summarized	Riley and Lo Presti 2001 http://www.bhi-erc.com/vadose/sac.htm#info
Diffusion coefficients for grouted low-level waste forms	Serne et al. 1992, Table 6, p. 277; Table 8, p. 279; Table 9, p. 280; Table 11, p. 281; Tables 12 and 13, p. 282
Diffusion coefficients for encasement cement concrete and soil fill materials	Mattigod et al. 2001, Tables 4.8 and 4.9, p. 4-23
Diffusion coefficients as applied to the 200 Area Plateau composite analysis	Kincaid et al. 1998, Appendix D, Table D.2
Diffusion coefficients in support of WIPP no-action alternative 2	Buck et al. 1996, Appendix B, Table B.4.
Effective diffusion coefficients for Environmental Restoration Disposal facility assessments	WHC 1993b, p. 3-6; Table 3-4, p. 3T-4
Diffusion coefficients in support of grout low-level waste performance assessment	Kincaid et al. 1993, Table 3.3, p. 3.37
Diffusion coefficients in support of 200 West Area low-level waste performance assessment	Wood et al. 1995, p. 3-16
Diffusion coefficients in support of 200 East Area low-level waste performance assessment	Wood et al. 1996, p. 4-12
Diffusion coefficients for grout performance assessment	Serne and Wood 1990, p. 2.19, 2.22-2.23; Table 18, pp. 2.52-2.53; Appendix A, Table A.2
Dissolution rates for fixated wastes (cementitious)	WHC 1993, p. 3-6
Time frame for dissolution of cements	Krupka and Serne 1998, p. 2.2
Natural analogs of cement and concrete materials	Krupka and Serne 1998, pp. 244-2.5; Appendix C

The cement model can also be used to simulate the release of contaminants stabilized in cementitious encasements (e.g., high-integrity containers, or entombed underground portions of decommissioned buildings). In this case, the release of contaminants is additionally controlled by the need for the contaminants to pass through the encasement material. Diffusion, dissolution, and corrosion properties of the encasement materials control such migration. Lastly, cementitious materials (i.e., both waste forms and encasements) can be chemically converted in the environment to waste forms of greater stability and more restrictive migration properties (i.e., lower diffusivity). The conversion of cement to carbonate is an example (Mattigod et al. 2001).

3.4 Reactor Block Model

The reactor block model is used to simulate release of contaminants from decommissioned surplus production reactors on the Hanford Site. The model formulation is simple, containing only a mass and fractional release term. With the exception of carbon-14, release modeling of selected other contaminants of surplus production has been based on fractional release rates. These release rates have been calculated from experimental leach rates (White et al. 1984) and Hanford reactor configurations.

The contaminant release mechanism of the reactor-block model is leaching contaminants from the graphite blocks of the production reactors. The rate of loss of contaminant for a given contaminant is given by:

$$dM / dt = - M_o F_{rrr} \qquad \text{(Equation D.65, p. D.19, Kincaid et al. 1998)}$$

where M_o = the initial quantity in Ci or kg of contaminant in the graphite core

F_{rrr} = the fractional release rate in yr^{-1}. F_{rrr} is analyte specific and its value ranges from 0 to 1.

This model, described originally in the surplus production reactor EIS (DOE 1989), generates a family of curves such that the smaller the value of F_{rrr}, the more elapsed time is required until a specific contaminant inventory is completely depleted from the graphite block. M_o serves as a multiplier or scaler.

The surplus production EIS (DOE 1989) used a temperature dependent fractional release for calculating the release of carbon 14 from the reactor block. The fractional rate was defined as

$$\frac{dM_i}{dt} = M_{oi}\ (365) \left[565 \left(1 + 100\ e^{-(0.08)(365)t} \right) e^{-6440/T} \right] \qquad \text{(Kincaid et al. 1998, p. D.19, Equation D.64.}$$

where M_{oi} = the initial quantity in Ci or kg of contaminant in the graphite core

T = the absolute temperature of the reactor block (K).

Sources of data for the reactor block model can be found in Table 3.12.

Table 3.12. Sources of Data for Reactor Block Model

Information Source Summary	Reference
Fractional release data for reactor block model as applied to the System Assessment Capability initial run (SAC Rev. 0) are summarized	Riley and Lo Presti 2001 http://www.bhi-erc.com/vadose/sac.htm#info
Release rates from surplus reactor graphite cores	DOE 1989, Appendix C, Table C.1
Corrosion rates of shielding and metal components of surplus reactors	DOE 1989, Appendix C, p. C.4
Release rates of carbon-14 from surplus production reactors	DOE 1989, Appendix D, pp. D.1-D.3
Other sources of radionuclides and their releases from surplus production sources	DOE 1989, Appendix D., p. D.6
Solubility of lead in Hanford groundwater	DOE 1989, p. 5.21
Leach rates used to derive fractional release rates for surplus production reactors	White et al. 1984, pp. 42-61.
Release rates of carbon-14 from surplus production reactors	Kincaid et al. 1998, Appendix D, pp. D.18-D.19

3.5 Glass Model

The glass model is used to simulate contaminant release from vitrified waste. Three model configurations are described. In the first configuration, it is like the cement model, requiring data on the total surface area and volume of the waste form. Unlike cement waste forms, however, vitrified waste is considered impermeable and therefore diffusion of contaminants out of the waste form to the waste form surface is not considered a relevant mechanism. Instead, contaminant release is governed by slow dissolution of the glass waste form (Kincaid et al. 1998). In the second configuration, contaminant release is represented as a function of a fractional release rate of a specific contaminant from the waste form (Kincaid et al. 1998; Mann et al. 1998). In the third configuration, a mechanistic approach is taken where dissolution of the glass, influenced by local chemical conditions, is assumed to control the release rate of radionuclide contaminants. A general rate equation was formulated that describes the dissolution of glass as a function of key parameters (e.g., pH). This rate equation was subsequently reflected within an equation that expressed the normalized flux of radionuclides to the vadose zone for glass waste packages (Bacon McGrail 2001). The third configuration is considerably more advanced and representative of contaminant release from glass than the other two configurations.

For the first configuration, the rate of release of contaminant is given by:

$$dM / dt = - M_{oi} \left(A / V \right) r$$

(Equation D.52, p. D.15, Kincaid et al. 1998)

where M_{oi} = the original quantity of the contaminant contained in the glass (Ci or kg)
A = the total surface area of the glass waste form (cm^2)
V = the total initial volume of the glass waste form (cm^3)
r = the volumetric dissolution rate of glass per area of surface ($cm\ yr^{-1}$).

In the second configuration, contaminant release from the glass waste form is expressed as a function of a fractional release rate of a contaminant from the waste form. The equation that applies in this case is:

$$dM / dt = F^3 (3 / F - t)^2 M / 9$$

(Equation D.58, p. D.16, Kincaid et al. 1998)

where F = the fractional release rate of a specific contaminant from the glass waste form (yr^{-1})
M = the initial total activity of the specific contaminant in the source zone (Ci).

Mathematical expressions depicting the third configuration (i.e., the mechanistic approach) can be found in Bacon and McGrail, 2001 (see Table 3.13)

Sources of data for dissolution (corrosion) and fractional release rates and the mechanistic approach to contaminant release from glass waste forms can be found in Table 3.13.

Table 3.13. Sources of Data for Glass Model

Information Source Summary	Reference
Glass dissolution and fractional release rates for vitrified waste as applied to 200 Area plateau composite analysis	Mann et al. 1997
Fractional release rates for glass waste form in 200 Area plateau composite analysis	Kincaid et al. 1998, Appendix D, p. D.21 and Table D.2
Constant corrosion rate for glass waste in support of TWRS EIS	DOE 1996, pp. F-57-F58
Corrosion rate for ILAW waste package containers	Mann et al. 2000, pp. 51-52
Glass corrosion reaction for ILAW waste	Mann et al. 2000, pp. 42-43
Dissolution reactions for selected mineral phases associated with ILAW waste	McGrail et al. 1999
Dissolution rates for vitrified waste constituents	WHC 1993b, p. 3-6
Corrosion reactions for ILAW glasses	Bacon and McGrail 2001, p. 12
Dissolution equation for ILAW glasses	Bacon and McGrail 2001, p. 13
Secondary phase equilibrium constants for ILAW glasses	Bacon and McGrail 2001, p. 14-16
Flux equation for release of ILAW glass constituents to the vadose zone	Bacon and McGrail 2001, p. 16-17
Kinetic rate parameters for ILAW glasses	Bacon and McGrail 2001, p. 14

3.6 Reactor Compartment Model

Surplus production reactors contain contamination imbedded in the stainless steel hulls and reactor vessel steels. Sources of contamination also exist within the contained portions of the hulls and reactor vessels. Contaminant release is envisioned as requiring a corrosion model to simulate release of contaminants from the reactor compartment steels plus a soil-debris model is to simulate release of contaminants from sources within the reactor compartments.

The contaminant release mechanism for materials and contaminants constituting decommissioned reactor compartment steels is corrosion of the steels. The rate of loss of contaminant for a given contaminant is given by:

$$dM / dt = - M_o \mathrm{F}_{\mathrm{rrr}}$$

(Developed from information in Rhoads et al. 1994, pp. vi and 4.2)

where M_o = the initial quantity in Ci or Kg of contaminant in the steel
F_{rrr} = the fractional release rate in yr^{-1}.

The fractional release rate is determined by multiplying the corrosion rate of the specific steel (kg/dm^2-yr) by the total surface area of the steel (dm^2) and dividing by the total amount of steel containing the contaminant of concern (kg).

Sources of corrosion, solubility, and distribution coefficient data can be found in Tables 3.4 and 3.14. Other data in support of soil-debris model applications to reactor compartment contaminants can be found in Tables 3.4 through 3.8.

Table 3.14. Sources of Data for Reactor Compartment Model

Information Source Summary	Reference
Corrosion rates of decommissioned nuclear reactor compartment steels	DOE 1996c, pp. 4-12-4-16
Corrosion rates of decommissioned nuclear reactor compartment steels	Rhoads et al. 1994, pp. vi-vii; p. 3.2
Total PCB solubility for decommissioned nuclear reactor compartment assessments	DOE 1996c, p. 4-32
Lead solubility for decommissioned nuclear reactor compartment assessments	Rhoads et al. 1992, p. vi, p. 3.10; p. 3.19
Solubility of nickel in Hanford groundwater	Rhoads et al 1994, p. 3.10; pp 3.14-3.16
Corrosion rates of decommissioned nuclear reactor compartment steels in Hanford soils	NFESC 1993

3.7 Containment as a Factor Influencing Release of Contaminants from Source Zones

Containment is a term that recognizes the presence of barriers that contaminants must pass through in order to be free of the engineered system containing the waste. These barriers include waste form containment (e.g., steel canisters, drums, reactor compartment hulls, wooden boxes, etc.), repository containment (e.g., high-level waste tank structures containing concrete and steel, concrete vaults and high integrity containers (HICs), underground steel storage tanks, etc.) decommissioned building foundations containing waste, and engineered system bottom liners. Processes influencing the stability of these containment systems include resistance to corrosion, dissolution, and biodegradation. Contaminants in waste have to await the corrosion of steel before they are released from such containment. On the other hand, diffusion through porous materials (e.g., wood and cementitious materials, plastic liners) may compete with containment degradation for release of contaminants to the subsurface. It also should be noted that some source zones contain uncontained and contained (stabilized) wastes. For example, low-level waste burial grounds contain low-level waste debris and category 3 waste contained in high-integrity waste concrete containers suggesting the need to apply more than one release model to some waste source zones. Table 3.15 lists sources of data important to the issue of containment.

Table 3.15. Sources of Data on the Issue Containment

Information Source Summary	Reference
Corrosion rates of decommissioned nuclear reactor compartment steels	See Table 3.14
Corrosion rates of decommissioned nuclear reactor compartment steels in Hanford soils	See Table 3.14
Recommended corrosion rates for reactor vessels	See Table 3.14
Diffusion coefficients for encasement cement concrete and soil fill materials	See Table 3.11
Dissolution rates for fixated wastes (cementitious)	See Table 3.11
Time frame for dissolution of cements	See Table 3.11
Natural analogs of cement and concrete materials	See Table 3.11
Corrosion rates of shielding and metal components of surplus reactors	See Table 3.12
Diffusion of contaminants through engineered system liner	WHC 1993b, p. 2-7
Diffusion through and biodegradation of asphalt barrier	Kincaid et al. 1993, pp. 3.49-3.59, p. 3.100, pp. 3.113-3.115
Diffusion through concrete and grout	Kincaid et al. 1993, p. 3.115
Corrosion rates of ILAW waste package containers	Mann et al. 2000, pp. 51-52
Corrosion of carbon steel liners of Hanford HLW tanks	Anantatmula et al. 1994, pp. 2-1, 2-3, and 2-4.

4.0 References

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