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A SOURCE RELEASE MODEL WITH APPLICATION TO THE LANL LLRW DISPOSAL SITE PERFORMANCE ASSESSMENT

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ABSTRACT

A source release model was developed to quantify time dependent liquid phase releases of radioactive material to the vadose zone from a disposal site. The model has been implemented to evaluate the source terms in the Performance Assessment for the Los Alamos National Laboratory (LANL) Low Level Radioactive Waste (LLRW) Disposal Facility at Area G, an analysis of the long term post-closure impact of disposal operations required by the USDOE orders.

Analytic solutions describe transport through the model compartments: the solid phase waste package, the liquid phase within each waste package and the liquid phase within the disposal unit. The model accounts for elemental solubility limits and retardation coefficients (R_f) separately in the waste package and in the disposal unit. Several parameters define the site specific aspects of the disposal unit. In our case for example, the disposal unit is waste buried with crushed volcanic tuff, and a small net infiltration rate is determined from independent numerical studies. The analytic solution allows efficient explorations of the sensitivity to the input parameters. Numerical solutions extend the model to account for decay product ingrowth which may have different transport properties than the parent nuclides.

Results show source release rates versus package or disposal unit K_d , and effects of solubility limits. At the LANL LLRW disposal site, only thorium, uranium and some low-level nuclides are solubility limited. Otherwise, a 'rapid rinse' waste package category dominates the peak disposal unit efflux which occurs for the LANL disposal site at about 100 years for nuclides with $K_d=0$ ($R_f=1$) and for proportionately longer times as K_d or R_f increases. Additional simple analytic models or detailed numerical codes can be coupled to these results to predict groundwater concentrations.

INTRODUCTION

A source release model was developed to quantify time dependent releases of radioactive material in the liquid phase to the vadose zone from a disposal site. The waste package chemical and physical forms, the solid-liquid phase equilibrium coefficients, and the solubility limits all play decisive roles in determining a realistic release rate from the disposal site, which ultimately controls the maximum contaminant concentrations seen subsequently after transport in the environment. The model has been implemented and used to evaluate the source terms in the LANL Low Level Radioactive Waste (LLRW) disposal site preliminary Performance Assessment (PA) [1], an analysis of the long term post-closure impact of disposal operations required by the USDOE orders.

It was not clear how previous source term models such as the mixed cell cascade model [2] or numerical codes such as DUST [3] could integrate the LANL inventory waste forms and obtain an accurate assessment of concentrations in comparison with solubility limits. In order to assure this, a simple analytic inventory-limited release model was developed and modified for the solubility limited regime. Numerical treatment was found necessary to account for decay

ingrowth products with different transport properties than the decay chain parent. This paper focuses on the development of the model, with some results from the Area G PA.

ANALYSIS

Model Development - Inventory Limited Release

The conceptual model for subsurface liquid phase release from a disposal unit is shown in Fig. 1a, and the compartment or control volume formulation of the flow for the model is shown in Fig. 1b. The three compartments are designated with the subscripts: s, the solid phase in the waste package volume, w, the water-dissolved or liquid phase within the waste package volume, and, d, the water phase within the disposal unit.

Multiple similar waste packages with solid phase contaminant concentration, C_s , dissolve to feed the liquid phase contaminant concentration within the waste package, C_w , while accounting for reabsorption on the solid phase. The liquid phase concentration infiltrates from the waste package with a Darcy flux, q_w , to feed the liquid phase contaminant concentration in the disposal unit, C_d . Flux from the disposal unit is controlled by a specified Darcy flux, q_d . Darcy flux through the disposal unit was determined independently for the Area G assessment [4] and the same value was assumed to apply to flux through the waste package.

The equations for transport through the compartment model are derived by integrating the continuity equation over the control volume represented by each compartment. The result for liquid phase concentration, C_l , is

$$\int \left(\theta \left(1 + \frac{K \rho_m}{\theta} \right) \frac{\partial C_l}{\partial t} + \nabla \cdot \Gamma \right) dV = 0$$

where we have used the definitions,

$$\begin{aligned} \rho &\equiv C\theta \\ \Gamma &\equiv C\theta u \equiv Cq \end{aligned}$$

and a source term, S_p , where

$$S_p = - \frac{\partial C_m}{\partial t} = - \frac{\partial (K \rho_m C_l)}{\partial t} = - K \rho_m \frac{\partial (C_l)}{\partial t},$$

has been used to represent loss from the liquid phase concentration, C_l , to the matrix concentration, C_m , by absorption processes. This is modeled with a retardation coefficient, R , based on the equilibrium partition coefficient, K , such that

$$C_m = K \rho_m C_l,$$

consistent with the retardation factor,

$$R = 1 + K\rho_m/\theta,$$

which reduces the effective contaminant flux accounting for absorption and contaminant holdup on the solid phase of the matrix material.

Fig. 1a. Conceptual model for subsurface liquid phase release from a disposal unit. A representative waste package is shown to the left, and a disposal unit is shown on the right.

Fig. 1b. The three compartments in the present release model are an equivalent representation of the conceptual model from Fig. 1a, showing contaminant concentrations, C , and flux, Γ for the three compartments.

The governing control volume continuity equations for each of the model compartments are

$$V_s \frac{\partial C_s}{\partial t} = - A_s u_s C_s,$$

$$\theta_w V_w \frac{\partial C_w}{\partial t} = \frac{A_s u_s C_s}{R_w} - \frac{A_w q_w C_w}{R_w},$$

and

$$\theta_d V_d \frac{\partial C_d}{\partial t} = \frac{A_w q_w C_w}{R_d} - \frac{A_d q_d C_d}{R_d}.$$

Here, A and V are compartment areas (in the horizontal plane) and volumes, and u_s is a dissolution speed, characteristic for the particular physical and chemical form of the waste package.

The contaminant is assumed initially to be present only in the waste package (solid phase, C_s) so the initial conditions for the problem of interest are

$$C_s(t=0) = C_{so}; \quad C_w(t=0) = C_d(t=0) = 0.$$

One can define the following

$$\lambda_s \equiv \frac{A_s u_s}{V_s},$$

$$\lambda_w \equiv \frac{A_w q_w}{V_w \theta_w R_w},$$

and

$$\lambda_d \equiv \frac{A_d q_d}{V_d \theta_d R_d}.$$

Using these definitions, the solutions to the governing equations and these initial conditions are

$$C_s(t) = C_{so} \exp(-\lambda_s t)$$

$$C_w(t) = \frac{C_{so}}{\theta_w R_w} \frac{\lambda_s}{(\lambda_w - \lambda_s)} \frac{V_s}{V_w} \left(\exp(-\lambda_s t) - \exp(-\lambda_w t) \right)$$

$$C_d(t) = \frac{C_{so}}{\theta_w} \frac{\lambda_w \lambda_s}{(\lambda_w - \lambda_s)} \frac{V_s}{V_w} \frac{V_w \theta_w R_w}{V_d \theta_d R_d} \times$$

$$\left(\frac{(\exp(-\lambda_s t) - \exp(-\lambda_d t))}{(\lambda_d - \lambda_s)} - \frac{(\exp(-\lambda_w t) - \exp(-\lambda_d t))}{(\lambda_d - \lambda_w)} \right).$$

The solutions can be modified for species decaying with decay constant, λ_i , by multiplying each solution equation by $\exp(-\lambda_i t)$.

These equations are then evaluated for each waste type with unique release and transport properties. The integrated compartment efflux, for example, from the disposal unit (Ci/yr) is $\Phi_d = A_d q_d C_d(t)$, and a similar expression applies to the liquid phase efflux from the waste package volume.

Solid Phase Partitioning

The formulation above accounts for the solid-liquid partitioning only in the C_w equation. It does not account self-consistently for the time dependent exchange between the C_s and C_w compartments nor does it account for the solid-liquid phase partitioning in the C_s equation. Therefore, the C_s values given by the above equation must be modified for $K_d > 0$, to give a solid phase concentration, C_{s-mod} , which accounts for the reabsorption.

The solid phase dissolution speed, u_s , prescribes the characteristic time scale over which nuclides are available for dissolution into the waste package water phase compartment. The

nuclide concentration in the solid phase will then initially decrease on this time scale but asymptote to the equilibrium partitioning between the solid-liquid phases as given by the appropriate value of K_d . After this asymptotic behaviour the waste package solid phase will decrease towards zero on the time scale describing the water phase leaching from or percolation through the waste package. This time scale is approximately equal to the waste package height divided by the percolation velocity, $u = q/\theta$, and is about 3-5 years for the Area G parameter values.

Based on this description of the physical mechanisms, an analytic expression for the time dependence of solid phase waste package concentration, C_{s-mod} , corrected for reabsorption and radiologic decay, is

$$C_{s-mod}[t] = C_s[t_0](f_c + (1 - f_c)\exp(-\lambda_s t))\exp(-(\lambda_w + \lambda_i)t)$$

where $f_c = 1/(1 + \theta/(\rho_m K_d))$, and $C_s[t_0]$ is the initial waste inventory concentration for the nuclide in that waste category volume.

Solubility limits

Solubility may limit the maximum concentrations in the waste package liquid phase for some elements. This is accounted for by evaluating $C_w(t)$ for all nuclides of a particular element, (including decay chain daughter contributions) summing over these nuclides and comparing this total concentration to the elemental solubility limit, C_{SL} . The elemental concentration is then limited at $C_w = C_{SL}$, with the limit for each nuclide, i , determined as its fraction, f_{si} , of the total elemental concentration at the time the solubility limit was first exceeded based on inventory limited release. Each waste form is treated separately, since no more than one waste form is expected in the same waste package at Area G.

During the time that the solubility limit applies then $C_w \equiv C_{SL}$ and $(\partial C_w / \partial t) = 0$. The governing equations for the s and w compartments can be combined to yield an equation valid during the solubility limit for the change in source term compartment concentration, C_s . The solution for the s -compartment after the solubility limit applies is a linear decay

$$C_s(\text{time} > t_{sl}) = f_{si} \left(C_s(t_{sl}) - \frac{A_w q_w C_{SL}}{V_s} \Delta t \right),$$

with $\Delta t = \text{time} - t_{sl}$, where t_{sl} is the time when the solubility limit is first exceeded, $C_s(t_{sl})$ is evaluated using the equation above for C_{s-mod} .

The solid phase concentration is reduced by the above Eqn. until the flux from the solid phase equals the liquid efflux from the waste package during a solubility limited release, or

$$A_s u_s C_s = A_w q_w C_{SL}.$$

At times greater than when this condition first applies, the element is no longer solubility limited and the inventory limited equations apply again to the subsequent release.

Therefore, initially the concentration in the w -compartment builds up to reach the solubility limit at some time, t_{sl} . The waste package efflux is then constant until the inventory is depleted to the extent that the solubility limit no longer limits the flux release. The linear decay of the waste package concentration during the time the solubility limit applies provides a

constant source into the disposal unit compartment. Using the above equations the solution in the disposal unit during the solubility limited duration for a nuclide with decay constant, λ_i , becomes

$$C_d(\text{time} > \text{tsl}) = \frac{S_{SL}}{\lambda_d} + \left(C_d(\text{tsl}) - \frac{S_{SL}}{\lambda_d} \right) \exp(-(\lambda_d + \lambda_i)\Delta t),$$

with

$$S_{SL} \equiv A_w q_w f_{si} C_{SL} / (V_d \theta_d R_w).$$

Numerical solutions extend the source release model to account for ingrowth of decay products which may have different transport properties than the parent nuclides. A numerical model for the 1-D vertical profile throughout the disposal unit has also been used to validate the compartment model approach used here. These topics are covered in detail elsewhere [5].

Application to the Area G Disposal Facility

The model assumes waste packages have release rates which are specified as a function of four waste categories derived from an inventory data base, with the categories including rapid release waste, soil absorbed waste, concrete absorbed waste, and corrosion-metal waste. These packages are distributed throughout a disposal unit volume, which in the LANL case, is backfilled to about 30% waste and 70% crushed volcanic tuff from the area (Fig.1). Equilibration K_d values and solubility limits for all the elements of concern in the Bandelier Tuff and in the concrete waste forms has been summarized by Longmire in [6]. Infiltration rates through the Area G disposal units have been computed independently [4].

RESULTS

Results in Fig.2 show compartment concentrations and efflux rates for LANL site conditions and for a single nuclide with a small but non-zero retardation ($R_f \sim 1.5$, or $K_d \sim 0.1$) in the waste package, and no retardation ($R_f=0$) in the disposal unit compartment ($K_d(\text{tuff})=0$). This gives the minimum time for releases. The 'Csmod' curves show the actual solid phase concentration accounting for retardation while 'Cs' shows the 'rapid release' ($K_d=0$) case.

Fig.3 shows source release rates for varying package K_d , and illustrates the reduction in maximum release rate with increasing K_d . The analytic solution allows efficient explorations of the sensitivity to the other parameters, including waste characteristics such as solubility limits or site-specific parameters such as infiltration rate. Parameters which describe disposal operations can be explored to compare effectiveness of various disposal and waste minimization options.

Extending the computation from predicting behaviour of a single nuclide in a single waste package form to an entire site inventory has proven to be non-trivial. Solubility limits must consider contributions summed over several nuclides including decay chain daughter ingrowth, while retaining separate waste package forms, and finally integrating the contributions across waste package forms for input to the subsurface transport model used to evaluate ground water contamination per inventory nuclide. An example of the integrated waste package release for the 'historical inventory' at Area G is shown for several Plutonium isotopes in Fig.4(left). The contributions from multiple waste forms contribute to the characteristic curve shapes.

Fig.2 Compartment concentrations and compartment efflux from the waste package liquid phase compartment for a unit source concentration.

Fig.3 (left)

Waste package compartment efflux for varying values of waste package K_d , $dcs=1$ is one year 'rapid rinse'.

An example of a solubility limited release is shown in Fig.4(right) for uranium, where the 'rapid release' waste form contributes the most and is solubility limited until depleted at about 5000 years. Here again, different waste forms contribute with solubility limits of shorter duration in other waste forms. At the LANL LLRW disposal site, only thorium, uranium and some low-level nuclides are solubility limited. Otherwise, a 'rapid rinse' waste package form dominates the disposal unit peak efflux which occurs for the LANL disposal site at about 100 years for nuclides with $K_d \sim 0$ ($R_f \sim 1$) and for proportionately longer times as K_d or R_f increases. Additional simple analytic models or detailed numerical codes can be coupled to these results to predict groundwater concentrations.

CONCLUSIONS

A new code was developed to model liquid phase release from waste packages in a disposal unit to the unsaturated zone for the Performance Assessment of the LANL Area G

Fig. 4 Area G results for the historical inventory showing the total waste package efflux in Ci/yr for four inventory-limited plutonium isotopes (left) and for three solubility limited nuclides of uranium (right). These curves correspond to the source term (moles/yr) input to the unsaturated zone 2-D transport model of Area G.

LLRW disposal facility. It accounts for several site-specific parameters including solid-liquid phase equilibration coefficients and solubility limits in different waste forms. The basic analytic forms for inventory release allow sensitivity studies and a practical integration over the complex source terms at a real site.

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