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THE ROLE OF A DETAILED AQUEOUS PHASE SOURCE RELEASE MODEL IN THE LANL AREA G PERFORMANCE ASSESSMENT

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THE ROLE OF A DETAILED AQUEOUS PHASE SOURCE RELEASE MODEL IN THE LANL AREA G PERFORMANCE ASSESSMENT

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ABSTRACT

The Performance Asessment for the LANL Low-Level Radioactive Waste (LLRW) Disposal Facilty, Area G, is on-going. A detailed review of the inventory data base records and the existing models for source release led to the development of a new modeling capability to describe the liquid phase aqueous transport from the waste package volumes. Inventory is sorted into four release form categories and screened in a comparison of leachate concentrations to the drinking water limit. Percolation through the disposal unit is prescibed in an independent surface water balance model incorporating site rainfall statistics. Waste package types and the disposal unit matrix have independently specified solubility limits and solid-liquid phase partition coefficients, or Kd values. Analytic solutions for inventory limited release of each nuclide in each of the four different waste package release forms are computed. Isotopic contributions are summed over elements to limit the waste package liquid phase concentrations to the elemental solubility limits. Time dependent releases from the waste packages for each nuclide which may be inventory or solubility limited are specified as model output which is provided as the source term to the unsaturated transport model. The waste package efflux is distributed over the 2-D unsaturated zone model grid points corresponding to the cross-sections for 5 representative disposal units within the mesa top. Results show the Area G release is dominated by the inventory in the 'rapid release waste form' (Kd = 0), which percolates from the waste packages over 5-100 years and from the disposal unit over 50-1000 years. Nuclides in waste package categories with larger Kd values are released proportionately slower. Uranium and thorium are the main nuclides of concern released as solubility limited nuclides from the 'historical inventory' at Area G. The analytic models provide an efficient means to explore the sensitivity of the results to variations and uncertainty in the model parameter values.

INTRODUCTION

A preliminary draft of the site Performance Assessment (PA) for the Los Alamos National Laboratory (LANL) low level radioactive waste (LLRW) disposal facility at Area G has been completed[1] as required per DOE orders. A

detailed review of the inventory data base, records and existing models for source release led to the development of a new modeling capability[2] to describe the liquid phase aqueous transport from the waste package volumes. The code output, a time dependent efflux to the disposal unit volume (as detailed in App.3B to ref.[1]), is provided as input to a sophisticated 2-D unsaturated flow model[3] for subsequent transport within the disposal unit and throughout the vadoze zone.

The Area G disposal facility is located on the top of a narrow finger-like mesa composed of volcanic tuff (Bandelier Tuff), deposited in stratigraphic layers of ash and solidified volcanic flows. Waste disposed at Area G is placed into pits excavated in the volcanic tuff, crushed in place, and backfilled with the native crushed tuff to about 30% waste package and 70% tuff by volume. These disposal operations are evolving to minimize future disposal volume, and to assure stability of emplaced waste.

Disposal operations at Area G began in 1959 and disposal inventory is established in a detailed electronic data base dating back to 1972. (Disposal for operations prior to that date is recorded in log books currently being transcribed to a new data base.) Nuclide quantities are associated with 50 waste codes, which characterize the physical and chemical form of the waste.

ANALYSES

These waste codes were sorted into four waste package release categories, based on preliminary modeling efforts which indicated the release characteristics of concern. The four release categories include rapid release (with compartment release time of one year, and with solid-liquid phase partition coefficient, Kd, set equal to zero, taking no credit for solid phase reabsorption), soil (Kd for Bandelier tuff at neutral pH), concrete/sludge (Kd for concrete at pH=12, and corrosion associated with metal waste forms (with compartment release time of three hundred years, and with Kd=0).

Of the 60 inventory nuclides listed with half-life greater than 5 years, 30 were screened out in a 'leachate screening', and 30 were followed in detail by the source release model. The leachate screening compared the total inventory concentration in the disposal unit moisture to the drinking water concentrations that result in 4mrem/yr at the standard ingestion rate. Of the 30 nuclides followed in detail, 15 were decay chain parents, with 48 non-secular equilibrium decay chain products evolved self-consistently.

Geochemistry for the waste packages was evaluated in terms of the equilibration coefficients, Kds, and elemental solubility limits, Csl, interpolated from the literature by Longmire[4]. Yucca Mountain Kds[5,6] were used as best estimates for Kds in tuff and as conservative estimates in soil, justified by a favorable comparison of water chemistry between Bandelier and Yucca Mountain tuff samples[4]. Solubility limits for tuff[7] were applied to all waste packages except waste in concrete/sludge, where values taken from the literature[8] were applied. A program to evaluate site-specific Kds under field

conditions is underway at Los Alamos. This will include five key nuclides in several Bandelier Tuff stratigraphies and over a range of water chemistry.

Percolation calculations were run with a modified version of the SPUR code[9] which does a detailed water balance at the surface. Results for the base case closure cover, 90 cm of crushed tuff, 10cm soil and a thin gravel-mulch seeded surface, showed an average of 4mm/yr percolation from the disposal unit bottom[10]. Recent results with revised crushed tuff hydrologic properties show an infiltration rate of 5.5mm/yr. The statistical analysis shows the average percolation rate is for a highly skewed distribution composed of 80% values equal to zero. The implications on the source release model of the actual distribution rather than the average value are under investigation. A range of infiltration model parameter variations were evaluated to account for evapotranspiration including rooting depth and leaf area index, and for run-off including surface slope and other factors important in the surface water balance[10].

The waste release model is based on a compartment representation of the package efflux, and depends upon package size, percolation rate or Darcy flux, retardation coefficient, and moisture content. The physical and conceptual flow models are shown in Fig.1, where the concentration and flux symbols are defined. An analytic solution for waste package efflux under inventory-limited conditions (everything which is not solubility limited) is evaluated for each nuclide in each waste package type (or release category). This is described in detail elsewhere[2] with the result for the waste package liquid phase compartment concentration, C_w, written as

$$C_w(t) = \frac{C_{so}}{\theta_w R_w} \frac{\lambda_s}{(\lambda_w - \lambda_s)} \frac{V_s}{V_w} \left(\exp(-\lambda_s t) - \exp(-\lambda_w t) \right) \qquad (1)$$

with

$$\lambda_s \equiv \frac{A_s u_s}{V_s} = \tau_s^{-1}; \qquad \lambda_w \equiv \frac{A_w q_w}{V_w \theta_w R_w}. \qquad (2)$$

The compartment subscripts are shown in Fig.1, and the λ values are 'compartment clearance rates' derived by a control volume integration of the continuity equation defined in terms of the compartment area, A (horizontal planar area), volume, V, Darcy percolation rate, q, dissolution velocity, u, moisture volumetric content, θ, and retardation coefficient, R, related in the usual way to the solid-liquid phase partition coefficient, Kd, as $R = 1 + \rho K_d/\theta$.

Using this time dependent concentration, the waste compartment efflux, Γ_w, is

$$\Gamma_w = A_w q_w C_w. \qquad (3)$$

Nuclide contributions to the same element, including ingrowth from decay chains, are summed to compare to the elemental solubility limits in

each waste package type. Waste package concentrations for each nuclide of the solubility limited elements are partitioned to the contributing nuclides. The solubility limited waste package efflux for a nuclide is

$$\Gamma_w = A_w q_w f_{sli} C_{sl}, \tag{4}$$

where C_{sl} is the elemental solubility limit and f_{sli} is the fraction of the solubility limit which is contributed by that specific nuclide. This fraction is assumed equal to the nuclides contribution to the elemental solubility limit at the time when that limit is first exceeded.

Solubility limited effluxes continue until the solid phase waste package concentration is depleted to the point the water phase concentration can no longer exceed a solubility limit, and subsequently the waste package is depleted at the inventory-limited release rate[2]. An algorithm of how the solubility limits are tracked over nuclides, elements and waste package types is included as Table I.

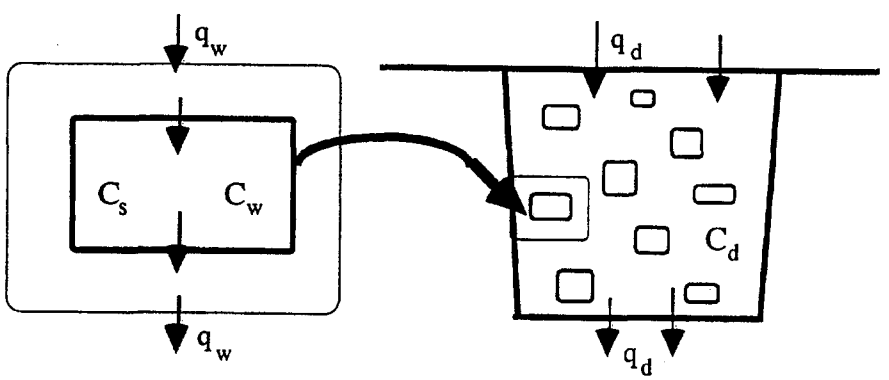

Fig. 1a. Conceptual model for subsurface liquid phase release from a disposal unit. A representative waste package is shown to the left, and a disposal unit is shown on the right.

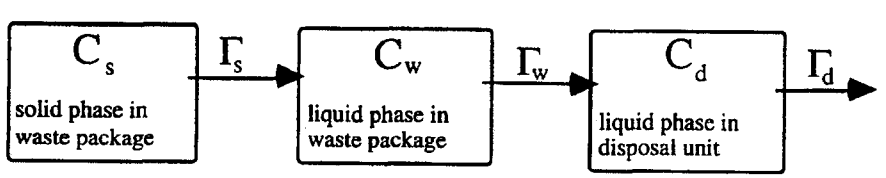

Fig. 1b. The three compartments in the release model are an equivalent representation of the conceptual model from Fig.1a, showing contaminant concentrations, C, and flux, Γ for the three compartments.

Table I

Algorithm for Model Implementation Over Nuclides and Waste Package Forms.

1. Calculate $C_s(t)$, $C_w(t)$, and $C_d(t)$ using inventory release limited equations for all time for each nuclide and for each waste form.
   A. inventory nuclides - analytic solutions
   B. decay chain daughters - numerical solutions

2. Sum all nuclide contributions (inventory and daughter products) to each element concentration for each waste form.

3. Determine time, tsl1, when solubility limit first applies for each waste form, when C_w(for element sum) > C_{sl}(element).

4. Interpolate (in time) to get C_w(tsl1) for each nuclide and each waste form which is solubility limited.

5. Calculate $C_s(t)$ and $C_d(t)$ under solubility limited equations
   A. inventory nuclides - analytic eqns
   B. daughter nuclides - numeric solution to eqns

6. Sum nuclide contributions to elemental concentrations per waste form.

7. Determine time, tsl2, when C_s(element) is depleted to point where solution is no longer solubility limited, when $C_s < C_{sl}\, q_w\, A_w/(u_s A_s)$.

8. Interpolate in time to get C_w(tsl2) for all nuclides for each waste form.

9. Continue from t = tsl2 to t_{end}, calculating $C_s(t)$, $C_w(t)$, $C_d(t)$ using the inventory limited equations.

The release model was tested extensively as described previously[1,2]. The analytic models described above were supplemented with numerical modeling in two areas. One, the ingrowth of progeny nuclides which can have transport properties different from the parent nuclides makes the analytic solutions to the compartment model impractical. Ingrowth of progeny is calculated numerically in a simple implicit scheme which implements the compartment governing equations with the added complexity of progeny ingrowth. This was used in the present modeling effort to account for 'daughter-product' ingrowth which could contribute to the solubility limit within a waste package. From the perspective of nuclide

transport in the site PA work, ingrowth within the packages was considered negligible, and ingrowth was modeled within the disposal unit and in the unsaturated zone within the 2-D unsaturated model[3] because that code has had more extensive QA and verification.

A second numerical modeling effort examined the compartment solutions where the waste package efflux feeds a one-dimensional (vertical axis) representation of the disposal unit. In this case, release from the disposal unit is controlled by the vertical gradient at the disposal unit bottom which evolves consistently along the vertical axis from waste packages homogeneously distributed throughout the disposal unit. This work confirmed that the global or compartment model results were reasonably accurate compared to results with 1-D profile effects in the disposal unit[2].

RESULTS

Figure 2 shows the concentrations verses time inside the waste package matrix. The solid concentration, C_s, is uncorrected for readsorption in the solid phase and corresponds to the case where Kd = 0. The actual solid phase concentration corrected for readsorption inside the waste package and assuming a nuclide with Kd = 0.5 is shown as 'Csmod', based on a governing equation derived elsewhere[2]. Csmod decays over the time scale for 'rapid release' to an equilibrium plateau at a fraction, $(\rho_m Kd/\theta/(1+\rho_m Kd/\theta))$, of the original solid phase concentration. It then decays to zero over the time scale for percolation from the waste package, along with the waste package liquid phase concentration, Cw. If this nuclide becomes solubility limited, the Csmod concentration is needed to accurately track the solubility limited release through the solubility limited release duration.

For the 'historical inventory' (1988 to the present) and site conditions at Area G, most nuclides are found to be inventory limited. An important result observed from the source release model is that for inventory limited nuclides, it is the component of inventory which is considered to be 'rapid release' which dominates the efflux function of time. All other release forms effect slower release rates which therefore reduce the efflux peak in time. The eventual peak in the aquifer concentration is determined by this release model modified by translation and dispersion during transport through the unsaturated and saturated zones.

These concepts are illustrated in Fig. 3, which shows the inventory release, Ci/yr, as a function of time for four of the plutonium isotopes in the Area G inventory. The peak release rate for each nuclide is dominated by the 'rapid release' (Kd = 0) waste package source term which occurs at about 3-5 years. Contributions to each nuclide from the other waste package release forms have relatively slower release rates due to effects of solid-liquid partitioning and readsorption (Kd > 0), and are seen in the Figure as the 'bumps' which occur later in time from 200-10,000 years. Because these

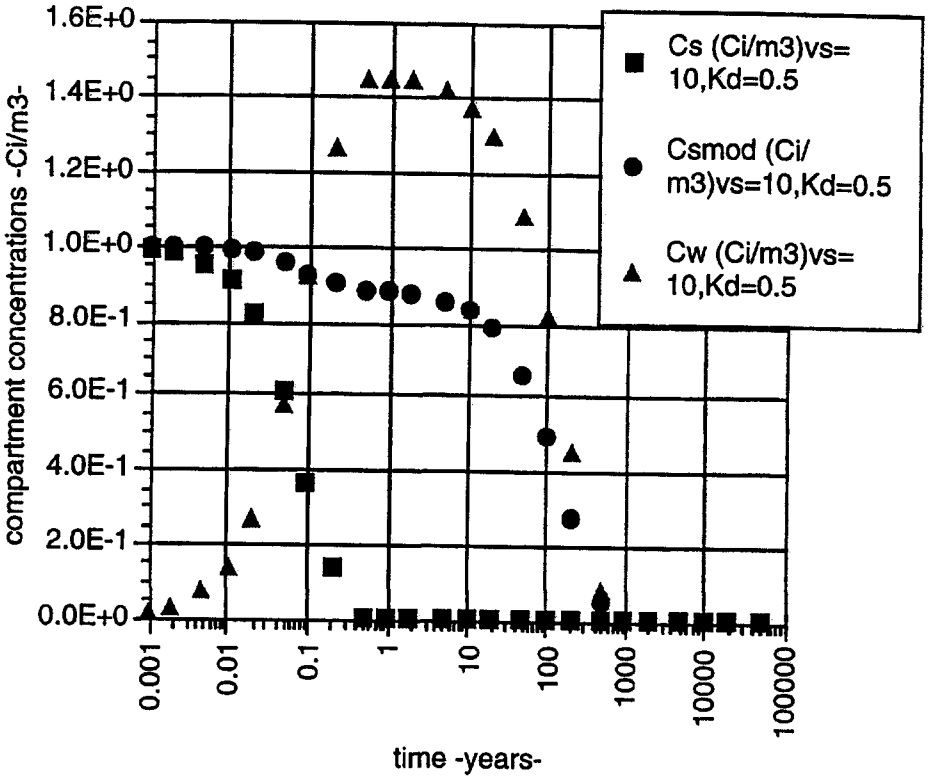

Fig.2 The solid phase concentration, Cs, is modified for reabsorption, Cs-mod, and decays to an equilibrium plateau over the time scale for 'rapid release'. It then decays to zero over the time scale for percolation from the waste package, along with the waste package liquid phase concentration, Cw.

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release rates are so small, the contribution from these waste package types to the peak release rate is small even if their inventory is comparable to that in the 'rapid release' category.

For the Area G inventory and site conditions, the only nuclides which contribute significantly to dose and are solubility limited are uranium and thorium. Uranium is depleted at a solubility limited rate which is sufficient to reduce the concentrations to an inventory-limited release at about 5000 years, after which the source decreases rapidly to negligible levels. This is illustrated in Fig.4 for three isotopes of uranium. As in the inventory limited case in Fig.3, there are contributions from multiple waste package types. The most inventory is in the 'rapid release' form which is solubility limited until about 5000 years. Imposed on top of this is a contribution from another waste form (concrete/sludge) which is also solubility limited but is depleted at about

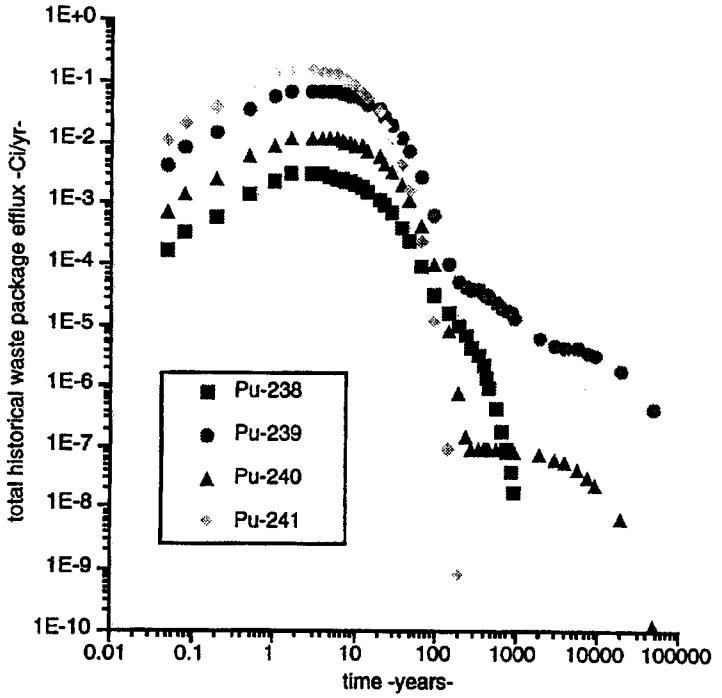

Fig. 3 Area G results for the historical inventory showing the total waste package efflux in Ci/yr for four inventory-limited plutonium isotopes. These curves correspond to the source term (moles/yr) input to the unsaturated zone 2-D transport model of Area G.

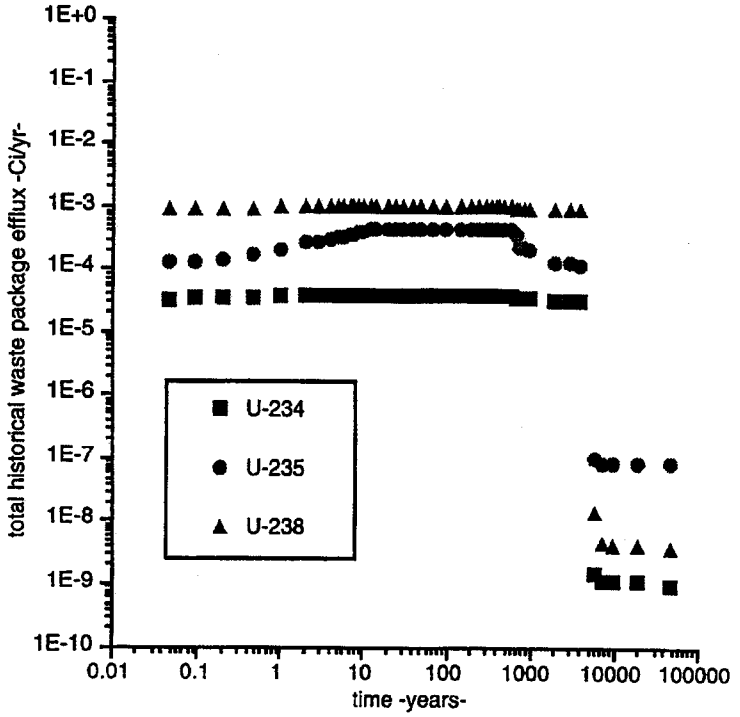

Fig. 4 Area G results for the historical inventory showing the total waste package efflux in Ci/yr for three solubility limited nuclides of uranium. These curves correspond to the source term (moles/yr) input to the unsaturated zone 2-D transport model of Area G.

500 years. Most of the uranium in this form is U-235, so only that isotope is noticably influenced by the second waste form.

The thorium solubility limit is so small that the release remains solubility limited effectively for all time. Plutonium is close to the solubility limit and would become solubility limited under slightly increased disposal concentrations.

Uncertainties in the results are examined via parametric examination of the input variables, and will be discussed in some detail[1,2]. Variability due to transient percolation effects is currently under investigation. Spatial variation in percolation rate or percolation differences within the disposal unit matrix verses within the waste package itself are being evaluated.

CONCLUSIONS

The results of a site Performance Assessment depend critically upon the source release model and its input data. The maximum concentrations in the time dependent efflux from the waste package will be integrated over the disposal unit volume and then translated through the vadoze zone and diluted in the aquifer while being modified slightly for dispersion and diffusion during the aqueous phase transport. Thus, it is a modification of the time dependent source release which will determine the receptor location concentrations and ingestion dose.

ACKNOWLEDGMENT

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HYDROGEOLOGIC ANALYSES IN SUPPORT OF THE CONCEPTUAL MODEL FOR THE LANL AREA G LLRW PERFORMANCE ASSESSMENT

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E.L. Vold, K. Birdsell, D. Rogers, E. Springer, D. Krier, H.J.Turin
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ABSTRACT

The Los Alamos National Laboratory (LANL) low level radioactive waste (LLRW) disposal facility at Area G is currently completing a draft of the site Performance Assessment (PA) as required per DOE orders. The site is located on a narrow mesa top of volcanic tuff where possible mechanisms of subsurface transport include vapor movement with vapor-liquid phase coupling, transport in fractures, and other as yet unresolved subsurface dynamics. The large depth to the water table (~250m) combined with uncertain hydrogeologic data below 100m and the complex stratigraphy of layered volcanic flows and ash provide for a challenging analysis. Results from previous field studies have estimated a range in recharge rate up to 1 cm/yr. Recent estimates of unsaturated hydraulic conductivity for each stratigraphic layer under a unit gradient assumption show a wide range in recharge rate of 10^{-4} to 1 cm/yr depending upon location. Numerical computations show that a single net infiltration rate at the mesa surface does not match the moisture profile in each stratigraphic layer simultaneously, suggesting local source or sink terms possibly due to surface connected porous regions. The best fit to field data at deeper stratigraphic layers occurs for a net infiltration of about 0.1 cm/yr. Surface moisture data and vertical moisture profiles suggest a wide range in infiltration at the near surface. Transients are hypothesized to play an important role in fracture infiltration and evaporation, but are seen in analyses to dampen rapidly within the mesa tuff matrix to a steady state condition.

A recent detailed analysis evaluated liquid phase vertical moisture flux, based on moisture profiles in several boreholes and van Genuchten fits to the hydraulic properties for each of the stratigraphic units. Results show a near surface infiltration region averages 8m deep, below which is a dry, low moisture content, and low flux region, where liquid phase recharge averages to zero. Analysis shows this low flux region is dominated by vapor movement. Field data from tritium diffusion studies, from pressure fluctuation attenuation studies, and from comparisons of in-situ and core sample permeabilities indicate that the vapor diffusion is enhanced above that expected in the matrix and is presumably due to enhanced flow through the fractures. Below this dry region within the mesa, near the interface of the vitrified and devitrified tuff units and also coincident with the elevation of the adjacent canyon floor, is a moisture spike which analyses show corresponds to a moisture source. This may indicate a narrow region with unique hydrologic properties, or horizontal moisture transport from the canyons. However, the likely physical explanation is seasonal transient infiltration through surface-connected fractures. This anomalous region is being investigated in current field studies, because it is critical in understanding the moisture flux which continues to deeper regions through the unsaturated zone.

INTRODUCTION

The Los Alamos National Laboratory (LANL) low-level radioactive waste (LLRW) disposal facility at Area G has completed a preliminary draft of the site Performance Assessment (PA)[1] as required per USDOE orders. The final draft PA becomes the technical basis for authorization and management of disposal operations at the active disposal site.

The Area G disposal facility is located on the top of a narrow finger-like mesa composed of volcanic tuff (Bandelier Tuff), deposited in stratigraphic layers of ash and solidified volcanic flows. Waste disposed at Area G is placed into pits excavated in the volcanic tuff, crushed in place, and backfilled with the native crushed tuff to about 30% waste package and 70% tuff by volume. These disposal operations are evolving to minimize future disposal volume, and to assure stability of emplaced waste.

The unresolved subsurface dynamics of the mesa top site location and the adjacent canyons with the large depth to the water table (~250m) provide for a challenging analysis. Uncertain hydrogeologic data below 100m have led to preliminary analyses focusing on the near surface hydrology in the mesa-canyon system. Preliminary review by the USDOE Peer Review Panel indicated that the proposed subsurface transport conceptual model[2] was not fully integrated with the numerical models[3] used to evaluate the site, especially in defining the transition from significant transient events to the steady state analysis and in defining the possible role of vapor transport. This report provides additional data review and analyses to address these issues.

DATA REVIEW

Area G site hydrology was summarized[4] as of 1987, based primarily on field studies by Bendix[5]. Several boreholes and core samples were used to characterize moisture verses depth and detailed hydrologic parameters. Moisture profiles by in-situ neutron probe measurements were seen to be independent of time below depths of about 2m. This was confirmed in recent measurements in a borehole into the crushed tuff backfill of an active Area G disposal unit[6]. These results suggest transients in moisture content play a negligible role at depth.

A recent review of the site geohydrologic data[7] including recent permeability work has been completed as part of the Area G PA work. The layered stratigraphy within and beneath the disposal site mesa is illustrated in Fig.1. Interpretation of the site geohydrologic data[2] identified several field observations which may be important in subsurface transport and postulated that transient infiltration into and long-term drying from fractures play a significant role.

In-situ air permeability measured by a borehole packer-isolation method was seen to be 4 to 20 times greater than the permeability of recovered core samples[4,5]. Similar results were obtained recently comparing core sample air permeability in borehole (G-5)[8] to in-situ permeability measured in the same borehole[9]. Attenuation with depth of atmospheric pressure oscillations[10] is consistent with permeabilities much larger than that measured on intact core samples. Subsurface diffusion of a tritium plume at Area G has been compared with numerical results to determine an in-situ diffusion coefficient.[11]

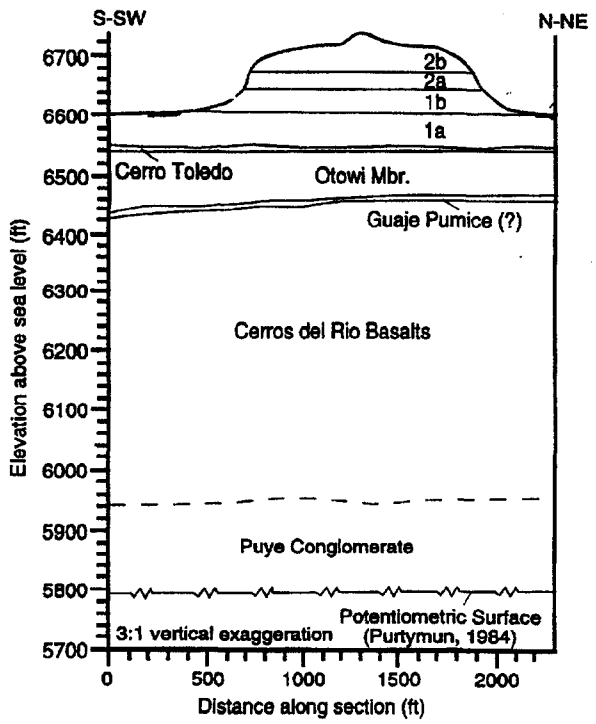

Fig.1 Stratigraphy beneath the Area G disposal facility, located on the top of a mesa (figure center). Disposal units are excavated to a depth of 70 feet or less into Unit 2b and can extend into Unit 2a.

Attributing the migration to vapor diffusion (liquid phase transport is negligible under the field conditions as will be discussed) gives a vapor diffusion coefficient which is more than one order of magnitude larger than that expected in the volcanic tuff matrix. These results indicate that fractures in the mesa top stratigraphic layers play a major role in determining the air permeability and effective vapor diffusion in the mesa.

ANALYSES

Unsaturated hydraulic conductivity and water characteristic curves were determined from van Genuchten fits[12] to matric potential data and from saturated conductivity data for several core samples in each of the near surface stratigraphic units at Area G disposal facility[13,14]. Detailed comparisons of unsaturated conductivities predicted in this manner with values measured by the unsaturated flow apparatus (UFA) centrifuge method[15] show good agreement in only about half the cases and indicate a large variability and uncertainty in the transport characterization[16,17].

The stratigraphic unit averages of van Genuchten fits were summarized for the Area G PA work[7] (Fig.2) over a range in moisture content up to 25% where the fits are accurately approximated by a straight line on a log scale. Assuming unit hydraulic gradients in the field, these curves correspond to recharge rates of 10^{-4} to 1. cm/yr in the

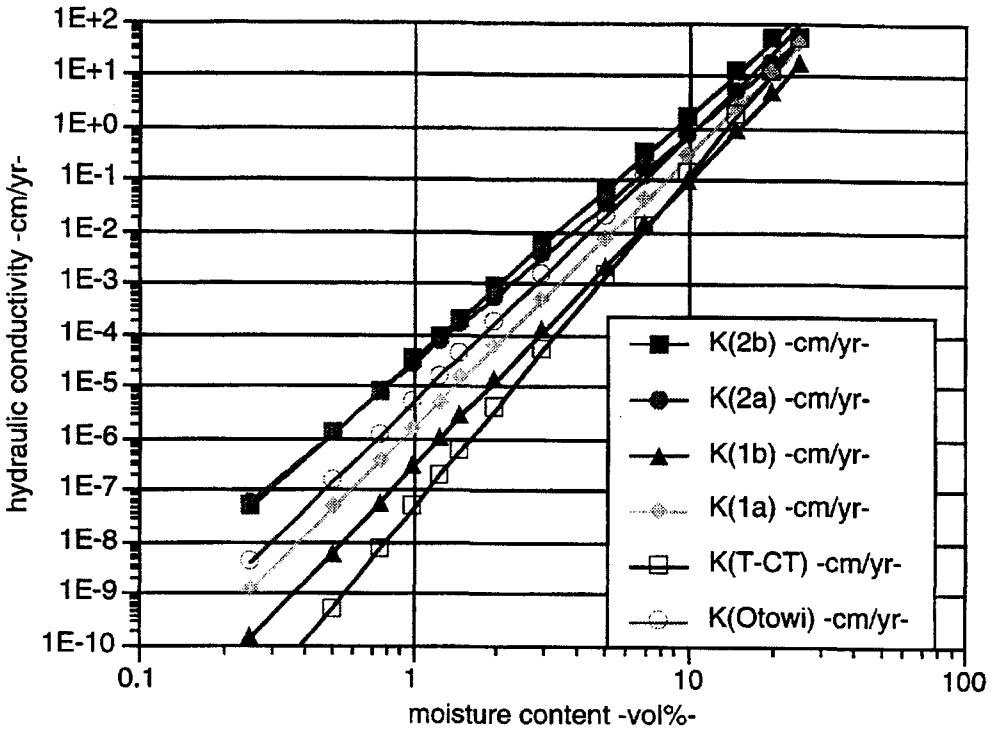

Fig. 2 Unsaturated hydraulic conductivity (van Genuchten fits) for the near surface stratigraphic units at Area G disposal facility. A preliminary estimate to the recharge rate is this function evaluated at the in-situ moisture content (in the range of 1-10%).

range of in-situ moisture content from about 1% to about 10%. If there are no local source or sink terms, then the recharge or flux through the strata must be constant, which would result in higher moisture contents in Units 1b and in the Cerro Toledo (see Fig.1, results labelled T-CT in Fig.2, and labelled C in Fig.3). This trend is consistent with field observations, but an accurate match of moisture content to field observations in each unit simultaneously is not possible as shown in the following.

This issue is examined in a comparison between numerical results and field data shown in Fig. 3. Steady state moisture contents are computed for several values of net infiltration at the mesa top, shown along a vertical line through the center of the mesa in a 2-D model with accurate stratigraphic cross-sections. The range of field data is shown as the darkened area. The canyon floors adjacent to the mesa coincide closely with the interface between Units 1b and 1a at Area G. The best agreement with field data occurs for downward vertical flux values of 0 to 0.001 cm/yr in the top two units, about 0.1 cm/yr through Units 1b and 1a, and as much as 1 cm/yr in the Otowi. Recent simulations show the higher moisture contents in the Otowi can be matched with lower flux values ($\leq$ 0.1 cm/yr) when hydraulic properties of deeper strata (subject to large uncertainty) are included in the numerical model.

The steady saturation profile calculated for any single infiltration rate does not match all the in-situ saturation data gathered at the site. Saturations within the mesa top

(Units 2b and 2a) are extremely low in field data. Saturations below the base of the mesa (Unit 1b and below) are higher. Several factors which may contribute to this discrepancy include: 1) infiltration from canyons or from deep surface-connected fractures resulting in higher saturations below the base of the mesa, 2) evaporation from the mesa sides and from fractures resulting in very low saturations within the mesa top, 3) uncertainty and heterogeneity in the hydrologic transport parameters.

For preliminary PA modeling work through all of the stratigraphic units, a rate of 0.1 cm/yr was chosen as the best 'fit' because there is no detailed knowledge of local source or sink terms to supply to the numerical effort. However, the comparison summarized in Fig.3 suggests a complex situation with local (elevation dependent) source and sink terms to the recharge rate.

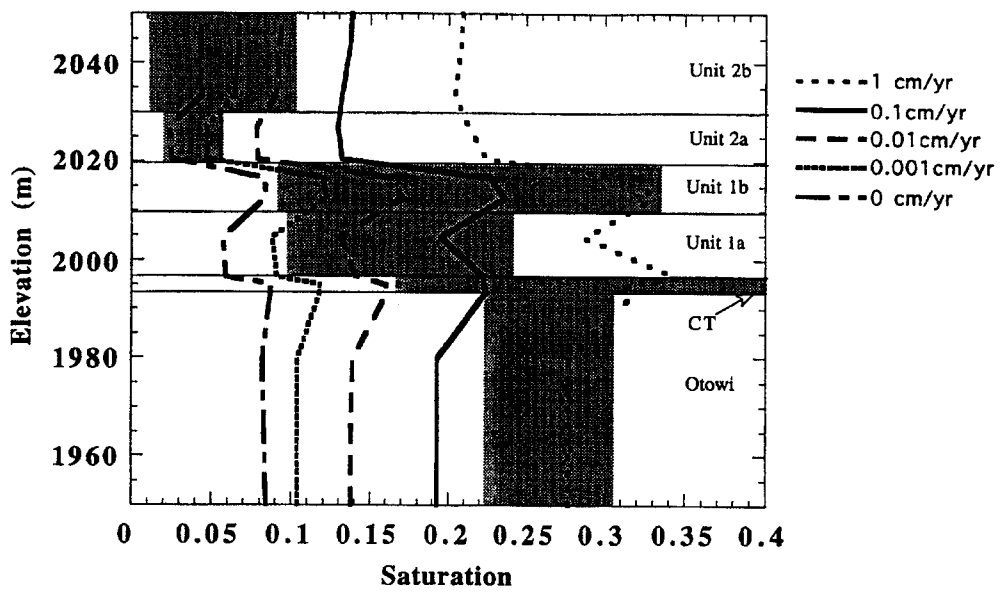

Fig.3 Numerical model prediction of saturation values verses depth at several net infiltration rates (from Ref.3). These are shown in comparison to field observations (the range in data within each stratigraphic unit is shown as a shaded area).

The source and sink terms in the underlying strata were examined in detail in a recent study[18]. The vertical flux and the vertical component of its divergence, interpreted as a local source or sink term, were evaluated directly from vertical moisture profiles using the stratigraphic unit-averaged van Genuchten fits to determine the hydraulic transport parameters for matric potential and for unsaturated hydraulic conductivity, and thus the vertical flux as a function of local moisture content. Results are discussed in detail for data from a dozen boreholes at Area G[18], for example, as shown for one 'typical' hole in Fig. 4.

Fig. 4A(top) shows the moisture profile in a borehole located near the center of the mesa top disposal facility. The inferred vertical flux and an effective source term to the local recharge rate labelled 'source' in figure 4B (middle), evaluated as the difference in vertical flux between adjacent points, show nearly zero flux or source throughout the range in depth from 7m to 25m. Near the surface there is a net downward flux (negative value of flux) of about 1 cm/yr, but there is also a net sink (negative source term) in this region, presumably corresponding to an evaporative loss to the surface or surface-connected fractures. At ~30m depth there is an apparent source of moisture with vertical moisture movement and evaporation (net sink) away from that source location. Similar results are seen in all the boreholes at this depth, however, quantitatively, there is a large variability with location[18].

The near surface infiltration region also has a large variability with location dependent upon the local disposal operations[18]. Modeling of deep percolation from the disposal pits is sensitive to assumed rooting depths, leaf area index, and other parameters which govern the detailed surface water balance[19]. The large variability in surface flux is consistent with the broad range measured in surface soil moisture content[20].

The source term indicated by the vertical difference in moisture flux can also be normalized per unit depth which allows the source to be expressed as an inverse time scale. This time scale can be interpreted as a characteristic time over which the moisture flux (labelled St-inv in Fig. 4C) is changing locally due to liquid phase movement, or, the time scale over which the moisture content (labelled St-inv*vol%) is changing if the entire vertical component of the divergence of flux is interpreted as a time dependent effect.

In Fig. 4C (bottom) the source term time scale for the moisture content (St-inv*vol%) is seen to be at least one year at all locations, and 100-1000 years over most of the 'low flux region' identified from Fig. 4B. In the near surface infiltration region, the observed characteristic time scale of 6-10 years implies deep penetration only once every several years, consistent with predictions by detailed surface water balence modeling studies[19]. Interestingly, the apparent source at the ~30m horizon has the minimum characteristic source time scale of about 1-3 years, consistent with significant infiltration once every few years. The short times near depth 30m may be an artifact of the unresolved stratigraphic hydrologic properties near the 1b-1a interface or may indicate a significant local source. Field studies are currently underway to resolve the matric properties within this region, which should allow a determination as to whether the apparent source term is real.

If the apparent source of moisture proves to be real, it may indicate moisture influx by horizontal transport from the adjacent canyons or from another fast flow path. A possible explanation is that this elevation is the bottom of a network of surface-connected fractures which allow transient infiltration during intense and infrequent storm events. The magnitude of the source term as a characteristic time scale of about 1-3 years is consistent with significant recharge once every few years.

DISCUSSION

Transients

Field studies [4,5] and analyses[18] indicate that transients in moisture content within the Bandelier Tuff matrix are dampened over short distances of 1-2m. Transients are

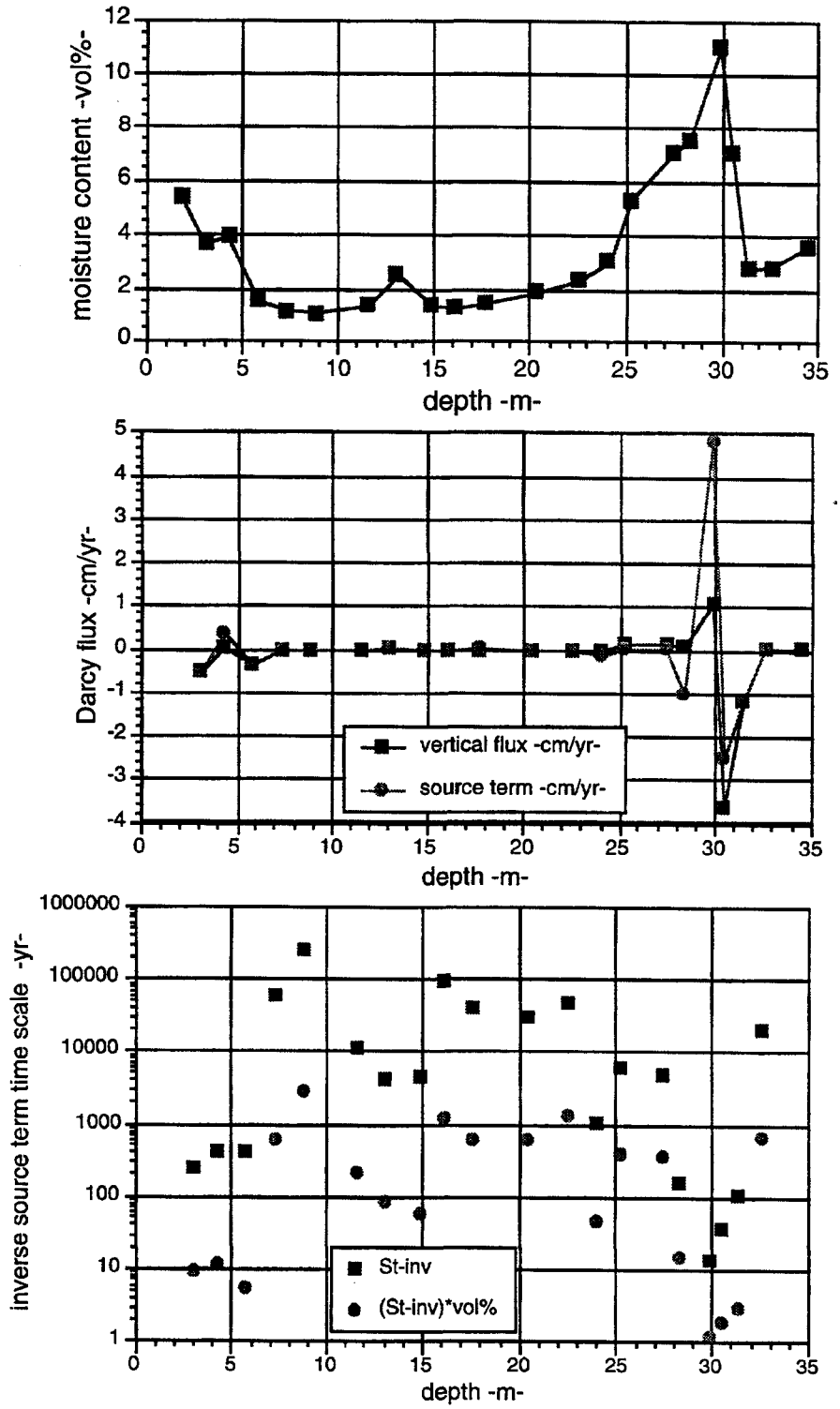

Fig. 4 Vertical profiles for hole **LGM-85-06** showing: A. volumetric moisture content, B. vertical flux and the source term expressed as a flux, and C. source terms expressed as a characteristic response time (from Ref .18).

expected to play a negligible role in moisture flux through the mesa tuff matrix compared to steady state flux. Transients can be significant if fast paths exist, e.g., near elements of surface-connected macro-porosity which can include surface-connected fractures, horizontal strata of high permeability (as proposed may occur at the surge beds between Units 2a and 2b) or other possible 'macro-pores'. An example of this may be the moist horizon observed near the vitrified-devitrified interface at ~30m depth. These transients average to an effective steady state moisture flux and moisture source term for long time scale numerical simulations.

Vapor Phase Flow

Vapor phase movement may be enhanced by diffusion due to barometric pumping or temperature fluctuations especially in strata of high permeability, however, the net convective movement of air through the tuff matrix is negligible. Evidence suggests convective movement through surface-connected features is significant and may influence the matrix flow. As such, diffusion of vapor phase contaminants is related to empirically determined effective diffusion coefficients which include possible barometric pumping effects.

Moisture profiles are expected to be driven by vapor diffusive movement when the moisture content falls below a specific value, which is about 2-5% for the hydrologic properties of the Bandelier Tuff under Area G. Here, in the low flux, dry region of the mesa interior, vapor phase loss through evaporation to surface connected fractures may contribute as a local sink term to drying regions below the moisture content expected to support liquid phase movement. It may be possible to model this as a sink term in the liquid phase matrix transport equations. To model transient dynamics between the fractures and the matrix, more sophisticated source/sink terms need to be considered such as that afforded by a dual porosity, dual permeability numerical model which can account for rapid fracture flow following infiltration events and evaporation along fractures during dry periods.

Upward and Horizontal Migration

Analysis of upward vertical moisture or contaminant flux from the disposal units to the surface is on-going. Preliminary results suggest near surface moisture profiles relax rapidly enough that long-term upward contaminant transport is negligible, leading to less surface contamination than the small amount attributed to translocation by biotic species as analyzed previously[1].

A preliminary parametric evaluation of horizontal moisture flux from the disposal unit to the mesa edges shows small contaminant concentrations can reach the mesa edge on the time scale of a thousand years. Numerical simulations currently in progress in accurate 2-D mesa geometry yield similar results and show radionuclide concentrations in the adjacent canyons comparable to that eventually reaching the ground water compliance point. Contamination at the mesa edge is assumed to be carried to the adjacent canyon floor by run-off and will be input to the off-site receptor dosimetry model and to a surface contamination source term for subsequent unsaturated transport to the deep aquifer under the saturation conditions existing beneath the canyon. The net contribution to canyon receptor dose and to deep ground water contamination via this canyon

contamination route is expected to be demonstrated to be small compared to the direct path beneath the mesa site.

Canyon Recharge

Recharge under the canyons adjacent to the mesa top disposal facility is likely to be much larger that that predicted within the mesa. Borehole data at nearby canyon locations shows volumetric moisture content values greater than 10% and profiles which do not vary greatly with depth in the Otowi layer. Assuming unit gradient conditions, valid under the reasonable assumptions that $\partial\theta/\partial z \sim 0$, and $\partial h_m/\partial z \sim 0$ (where θ is volumetric water content and h_m is matric potential expressed as a head) then the recharge rate is evaluated from the K_{unsat} curve (Fig.2) and corresponds with (1 to ~30) cm/yr over the observed moisture range in the Otowi from 10% to 22%. This large recharge rate may mix with the low recharge rate under the mesa at some depth, dependent upon the complex 3-D stratigraphy and stratigraphic properties. This is the subject of on-going investigation.

Toward a Conceptual Model

In this iteration of continuing analyses, the conceptual model which emerges considers first the unperturbed system and then the result of disposal operations. Transient events and vapor-liquid phase coupling can be modeled using local sources (increased local infiltration) and sinks (net evaporation from fractures or surge beds) or using more sophisticated dual porosity, dual permeability models.

Vertical contaminant flux through the unsaturated zone may be complicated by moisture source and sink terms for elevations equal to and above the adjacent canyon floors. The physical nature of these terms is uncertain, however, a picture consistent with data in the upper most strata assumes that infiltration and drying from the fractures in the mesa contribute to near surface (0-10m) moisture profiles and deeper (10-25m) drying relative to that expected for a constant vertical moisture flux. At the elevation of the adjacent canyons there is evidence of a moisture source which implies moisture migration from canyon alluvial aquifers with possible 2-D and 3-D effects, or deep infiltration through open fractures. The evidence is ambiguous and intended to be resolved in on-going field studies.

Disposal operations have been projected to increase the net moisture flux to the aquifer by over 200% of the unperturbed flux[1,3], through effectively increasing the moisture content and thus the hydraulic conductivity of the region beneath the disposal units and thus beneath the mesa as a whole. This perturbation is very sensitive to the assumed ground cover and rooting depth on the disposal unit post-closure covers[19]. The effect of disposal operations on moisture flux and therefore contaminant migration from the site is therefore likely to be significant, emphasizing that operations should be conducted to minimize the perturbations to a geologic system which is naturally well suited to minimize contaminant migration and maximize waste isolation.

Unsaturated transport from the disposal site through each of the strata to the main aquifer is being approximated using conservative estimates for the unknown transport properties in the deeper strata. The net result in concentrations and transit time to the aquifer is being revised from that determined in the original steady state analysis[1,3] to include transport through the deeper basalt layers and 3-D geometry effects.

CONCLUSIONS

As more data and analyses accumulate, the best estimate of the mean recharge rate in the upper stratigraphic layers within the mesa decreases while the variation between locations remains large. Our present best estimate is that the mesa interior is dominated by vapor phase movement and supports negligible liquid phase movement at least down to an elevation near the adjacent canyon floors. This holds under undisturbed conditions but is uncertain under the disturbance of disposal operations. A moisture source is apparent in analyses of the moisture profile peak observed near the depth of the interface between the vitrified and the devitrified tuff, also near the elevation of the adjacent canyon floors. Surface connected fractures to this depth could explain this source although the analysis method is inconclusive, subject to data uncertainties requiring additional field work for verification. At greater depths below the mesa the recharge rate is less well determined but results are consistent with a downward flux of 1 mm/yr or less directly beneath the mesa. Beneath the adjacent canyons the recharge rate is higher and remains to be quantified in near future studies. Preliminary results showed little mixing of infiltration originating beneath the canyon and mesa in the case of horizontal stratigraphy. Recent model results show different behaviour with mixing sensitive to the imposed dip of some units. The extent of mixing between canyon and mesa infiltration in more realistic 3-D geometry is currently under evaluation. The implications for PA modeling is the subject of on-going work.

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