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Probabilistic Modeling of One-Dimensional Water Movement and Leaching from Highway Embankments Containing Secondary Materials

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ABSTRACT

Predictive methods for contaminant release from virgin and secondary road construction materials are important for evaluating potential long-term soil and groundwater contamination from highways. The objective of this research was to describe the field hydrology in a highway embankment and to investigate leaching under unsaturated conditions by use of a contaminant fate and transport model. The HYDRUS2D code was used to solve the Richards equation and the advection–dispersion equation with retardation. Water flow in a Minnesota highway embankment was successfully modeled in one dimension for several rain events after Bayesian calibration of the hydraulic parameters against water content data at a point 0.32 m from the surface of the embankment. The hypothetical leaching of Cadmium from coal fly ash was probabilistically simulated in a scenario where the top 0.50 m of the embankment was replaced by coal fly ash. Simulation results were compared to the percolation equation method where the solubility is multiplied by the liquid-to-solid ratio to estimate total release. If a low solubility value is used for Cadmium, the release estimates obtained using the percolation/equilibrium model are close to those predicted from HYDRUS2D simulations ($\sim 10^{-4}$–10^{-2} mg Cd/kg ash). If high solubility is used, the percolation equa-

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tion over predicts the actual release (0.1–1.0 mg Cd/kg ash). At the 90th percentile of uncertainty, the 10-year liquid-to-solid ratio for the coal fly ash embankment was 9.48 L/kg, and the fraction of precipitation that infiltrated the coal fly ash embankment was 92%. Probabilistic modeling with HYDRUS2D appears to be a promising realistic approach to predicting field hydrology and subsequent leaching in embankments.

Key words: probabilistic; calibration; leaching, unsaturated; coal fly ash; Bayesian; Cadmium; fate and transport

INTRODUCTION

Highway embankments and pavement structural layers such as base/subbase layers, shoulders, asphalt concrete, and Portland cement concrete provide suitable settings to utilize large volumes of secondary materials such as coal fly ash, steel slag, reclaimed asphalt pavement, and recycled concrete (Apul *et al.*, 2003). A major environmental concern for use of secondary materials in the highway environment is the potential long-term leaching of contaminants, which may result in widespread soil and groundwater contamination. Leaching from embankments may pose an even greater problem than leaching from structural components of the highway considering that much larger volumes of material are used in uncovered embankments. If predictive methods for contaminant release are available, more informed decisions can be made about the use of secondary materials in the highway environment.

Physical and chemical factors dictating leaching from secondary materials are complex, and many studies have focused on various aspects of leaching. For example, a significant portion of the leaching literature discusses laboratory experiments under varying liquid-to-solid ratios and pHs (Kosson *et al.*, 1996, 2002). Some researchers have modeled the pH-dependent leaching behavior by equilibrium dissolution/precipitation reactions (Kida *et al.*, 1996; Fallman, 2000), and more recently by sorption reactions (surface complexation and surface precipitation) (Meima and Comans, 1998; Dijkstra *et al.*, 2002). Others have coupled diffusion with chemical equilibrium to model leaching behavior in the laboratory (Ganguly *et al.*, 1998; Gardner *et al.*, 2002; Park and Batchelor, 2002; Kosson *et al.,* 1996, 2002) suggested use of a percolation and a diffusion equation to extend laboratory results to field leaching conditions. Most of these studies have assumed that the water flow through the secondary material was uniform and constant, and did not consider the unsaturated flow in their analyses.

Modeling and field studies show that the highway environment remains unsaturated most of the time (Birgisson and Robertson, 2000; Birgisson and Ruth, 2003). An accurate description of this unsaturated flow would be helpful for understanding contaminant release in field conditions. In unsaturated conditions, carbonation and oxidation reactions may affect the contaminant release by modifying the matrix pH and chemistry (Townsend *et al.*, 1999; Sanchez *et al.*, 2002). Details of pavement hydrology are also needed to determine the dominant physical release and transport processes of contaminants in the field. Either diffusion or solubility may limit contaminant release in the field, and the relative importance of these processes may depend on water flow conditions, which will vary spatially (e.g., below cracks/joints, unpaved shoulders versus below intact pavement sections) and temporally (e.g., dry and wet periods).

Some of the recent modeling work in highway environments includes advective and diffusive transport of contaminants and spatial variability of the hydraulic regimes in two dimensions. For example, de Haan *et al.* (2003) investigated variably saturated water flow in highway pavements using laboratory and field experiments as well as numerical simulations using HYDRUS2D. Experimental data and two-dimensional modeling results suggested that there might be lateral water movement from the shoulder to the area below a relatively impermeable pavement. Using HYDRUS2D, Bin-Shafique *et al.* (2002) reproduced concentration measurements from coal fly ash in full-scale field studies and laboratory column experiments. Huber *et al.* (2001) developed the IMPACT model specifically for predicting the impact of beneficial use of secondary materials in roads. While unsaturated flow was not considered for simplicity, multiple transport, removal, and retardation processes (e.g., advection, dispersion, sorption, biodegradation, photolysis, and volatilization) were included in the IMPACT model. Pagotto *et al.* (2003) used CESAR and PHREEQC codes to model leaching from municipal solid waste incinerator ash and 3FLO code for mass transfer in the underlying soil. The model was one-dimensional and considered variably saturated flow, advection, and diffusion, as well as precipitation/dissolution reactions.

This paper describes the probabilistic application of a finite element model to simulate variably saturated flow and contaminant leaching in one dimension in a highway embankment. The goal of the research was to develop a probabilistic model for water movement in an existing highway embankment and to evaluate the hydrological and leaching response of the embankment if part of the embankment was replaced by a secondary material, coal fly ash. A probabilistic approach was used because the confidence in release estimates can be expressed by explicitly considering the variability and uncertainty in the complex physical and chemical factors affecting leaching. Many authors have treated uncertainty (degree of ignorance about the precise value of a parameter) and variability (inherent variation in the value of a particular parameter within the population of interest) separately (Frey and Rhodes, 1996; Rai *et al.*, 1996; Maxwell and Kastenberg, 1999); in this paper, we do not make that distinction, and assume that the probability distributions we use represent the combined true uncertainty and variability. A probabilistic approach provides information on the confidence of the output, which is not obtainable from deterministic modeling. Water movement was simulated in an embankment at the Minnesota Department of Transportation's (MnDOT) MnROAD instrumented, outdoor test facility. Unsaturated hydraulic parameters of the model are often not known for many virgin and secondary pavement materials. Thus, a Bayesian approach was taken where the uncertainty in unsaturated parameters was propagated through the model and then updated using Bayes' theorem and embankment water content data for 14,641 simulations. This updated information was combined with literature data to investigate a hypothetical leaching scenario for Cadmium when coal fly ash is used in the embankment.

METHODS

Field site

The MnROAD test facility consists of 40, 152-m-long hot mix asphalt and Portland cement concrete test sections with varying structural designs. Each test section is instrumented to monitor strength and hydraulic properties. The hydraulic properties of the embankment were predicted from water content measurements made in the embankment of test section 12, a Portland cement concrete pavement with an asphalt shoulder (Fig. 1a). Water content was measured every 3 h using an automated time domain reflectometry (TDR) waveguide. The Ledieu *et al.* (1986) calibration equation was used to convert dielectric values to volumetric water content. A 16-day period (23 July–8 August 1997) was used for Bayesian uncertainty analysis of the parameters. This period was selected because (1) no major rain events had occurred prior, (2) the water contents had been constant for days, and (3) it included several rain events of varying intensities.

Finite element model

HYDRUS2D, a Windows-based finite element code, was used for all simulations (Simunek *et al.*, 1999; Rassam *et al.*, 2003). HYDRUS2D numerically solves the

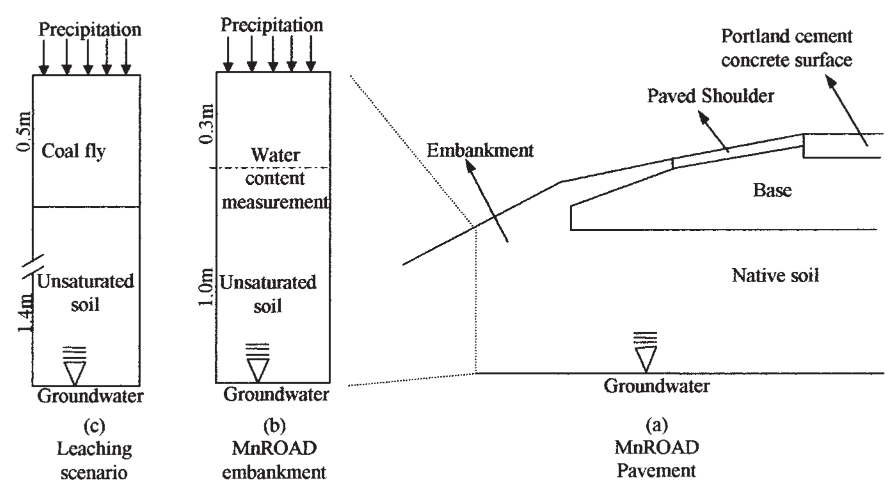

Figure 1. Cross section of MnROAD test section 12 (**a**), conceptual model of the MnROAD embankment (**b**), and coal fly ash scenario (**c**).

following equations for variably saturated water movement and solute movement:

1. Advection–dispersion equation with retardation for unsaturated medium:

$$\frac{\partial(\theta C)}{\partial t} + \rho_b \frac{\partial(K_d C)}{\partial t} = \frac{\partial}{\partial z}\left(\theta \frac{\partial C}{\partial z}(\tau D_m + \theta v D_i)\right) - \frac{\partial(vC)}{\partial z} \quad (1)$$

where ρ_b is bulk density, [M/L^3), K_d [L^3/M] is the partition coefficient, τ is the tortuosity factor [−], θ is volumetric water content [−], C is aqueous concentration [M/V], D_m is the molecular diffusion of the metal in free water [L^2/T], D_i is the Dispersivity [L], and ν is the advective velocity [L/T]. The assumptions of this equation are local equilibrium and linear sorption.

2. Richards' equation for water movement in unsaturated media:

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial z}\left[K(h)\left(\frac{\partial h}{\partial z} - 1\right)\right] \quad (2)$$

where h is the pressure head [L].

3. Closed-form expression of the van Genuchten (1980) formulation for the soil moisture retention curve:

$$\theta = \theta_r + \frac{\theta_s - \theta_r}{(1 + |\alpha h|^n)^{1-1/n}} \quad (3)$$

where θ_r [−] is (volumetric) residual water content, θ_s [−] is (volumetric) saturated water content, and α [1/L] and n [−] are fitting parameters.

4. Variation of hydraulic conductivity with water content:

$$S_e = \frac{\theta - \theta_r}{\theta_s - \theta_r} \quad (4)$$

$$K(\theta) = K_{sat} S_e^{0.5}[1 - (1 - S_e^{n/(n-1)})^{1-1/n}]^2 \quad (5)$$

where S_e is effective water content [−], and K_{sat} [L/T] is saturated hydraulic conductivity. Equations (3) and (4) assume that the soil moisture retention curve and the hydraulic conductivity curve for a soil can be estimated using five parameters (α, n, θ_s, θ_r, and K_{sat}).

Embankment infiltration model

Precipitation was input into the model in 15-min intervals as a time-varying flux boundary condition at the surface of the embankment (Fig. 1b). Maximum pressure head at the embankment surface was set to 2 mm to allow for runoff. The effect of groundwater was modeled by setting a constant zero-pressure head at 1.3 m below ground surface, which is the actual depth to groundwater. Time varying water content values at 0.32 m from the surface were stored for each simulation for posterior probability calculations. Ground freezing and evaporation were not considered.

The initial water content distribution was specified to be as close to steady-state conditions as possible. In HYDRUS2D, equilibrium conditions can be specified based on the pressure at the bottom of the mesh. When equilibrium pressures are assigned, the initial water content at the measurement depth may be considerably higher or lower than the first measured water content value. To overcome this problem, initial conditions were set at equilibrium pressure and these pressure values were converted to water content values based on the most likely parameter set of preliminary modeling exercises.

Probabilistic calibration of hydraulic parameters

There were two major criteria for selecting a model for calibrating the hydraulic parameters: to be able to incorporate all available information on the system, and to be able to present the results in a probabilistic way such that the confidence in results would be explicit. A Bayesian approach satisfies both criteria because Bayes' theorem [see Equation (9)] can statistically weight, and thus update, the prior information about the model parameters (e.g., from literature values, preliminary modeling exercises), with the degree of agreement between model predictions and observed field water content data. Bayesian approaches differ from classical statistics by allowing use of a subjective probability distribution, which represents the information on the system prior to new data collection. In this research, uniform prior distributions for the parameters were assumed, which resulted in each simulation having equal probability prior to comparison of simulations results with the field data. Through a Bayesian updating procedure, the probability of each simulation and the probability distributions of the parameters were recalculated.

The Bayesian approach used in this research for probabilistic calibration of hydraulic parameters (θ_r, θ_s, K_{sat}, α, and n) was adopted from Sohn *et al.* (2000). Uniform prior probability distributions were assigned to the unsaturated parameters based on literature data (Bigl and Berg, 1996) and previous embankment modeling experience. Uniform prior distributions were selected because the prior available information suggested that the parameters could take any value within the expected ranges. The parameter θ_r was kept constant at 0.25 based on values reported by Bigl and Berg (1996), and water content observations of the embankment in the field. The ranges of the remaining parameters are shown in Table 1. Parameters were grid samples (11 samples each) from uniform distributions and HYDRUS2D was run for all com-

Table 1. Prior and posterior mean and standard deviations of the updated parameters

Parameters/ranges	*Prior* Mean	*Prior* Standard deviation	*Posterior* Mean	*Posterior* Standard deviation	% Change in standard deviation
θ_s (0.33–0.43) (m^3/m^3)	0.380	0.029	0.356	0.011	−60.7
α (2.7–4.7) (1/m)	3.700	0.577	3.788	0.673	16.5
n (2.6–4.1) (−)	3.350	0.433	3.854	0.215	−50.4
K_{sat} (0.3–4.3) (m/day)	2.300	1.155	0.353	0.246	−78.7

binations of the parameters ($11^4 = 14{,}641$ simulations). Visual basic code was used to sequentially run HYDRUS2D for each parameter combination and store the necessary output in a designated folder. The simulated output from HYDRUS2D was compared to field measurements of water content at a depth of 0.32 m into the embankment by calculating the likelihood of each simulation. Posterior probabilities of each simulation were obtained from Bayes' theorem.

Assuming the error is distributed normally, the likelihood for any observation in time for the *u*'th simulation was calculated using the following equation:

$$L(\mathrm{O}(t)|Y(t)) = \frac{1}{\sqrt{2\pi\sigma_u}} \exp\left(-\frac{1}{2}\left[\frac{O(t) - Y(t)_u}{\sigma_u}\right]^2\right) \quad (6)$$

where σ_u is the variance of the difference between measured and modeled water content values in time; $O(t)$ is observed and $Y(t)$ is modeled water content value at 0.32 m below ground surface.

The likelihood of observing all data points in time (129 observations for 16 days) and thus the likelihood for the *u*'th simulation is given by the product of likelihood of each observation, which quantifies the difference between the observations and the model output:

$$L(O)|Y_u) = \prod_{t=1}^{T} L(O(t)|Y(t)_u) \quad (7)$$

The variance of each simulation was calculated as:

$${\sigma_u}^2 = \frac{1}{T}\sum_{t=1}^{T}(O(t) - Y(t))^2 \quad (8)$$

where t is 3-h intervals up to 16 days.

The posterior probability of each simulation, p_u, was calculated from the likelihood using Bayes theorem (see Gelman *et al.*, 2000 for a general reference).

$$p'_u = p'(Y_u|\mathrm{O}) = \frac{L(O|Y_u)p(Y_u)}{\sum_{u=1}^{U} L(O|Y_u)p(Y_u)]} \quad (9)$$

The prior distribution for the stimulations was uniform (each simulation had equal probability initially) resulting in a constant $p(Y_u)$ value, which was canceled out from the denominator and the numerator. Thus, the posterior probability of each simulation was the normalized likelihood value.

Once the posterior probabilities were calculated for each simulation, the profile likelihood ratio concept (Kalbfleish and Sprott, 1970; Vrugt and Bouten, 2002) was used to determine the confidence intervals of the posterior parameters. Chi-square significance of $p < 0.05$ were chosen as a cutoff for deciding which parameter combinations (simulations) were significant.

Probability weighted mean and variance of the unsaturated parameters were calculated from the posterior probabilities that passed the likelihood ratio test and the corresponding parameter value using:

$$\mu'_\theta = \sum_{u=1}^{U} \theta_u \cdot p_u' \quad (10)$$

$$\sigma'_\theta{}^2 = \sum_{\mathrm{i}=1}^{\mathrm{U}} (\theta_\mathrm{u} - \mu'_\theta)^2 \cdot \mathrm{p}'_\mathrm{u} \quad (11)$$

where θ_u is one of the four parameters investigated, and U is the total number of simulations (14,641).

In calculating the likelihoods, each of the data points in time in the measured data set were treated as independent from one another. The temporal correlation between consecutive measured water content values was not included in the statistical model because the posterior probability distributions were used as inputs for other simulations designed for more general hydrology and contaminant transport predictions in embankments. An autoregressive error model for the residual time series (field measured minus HYDRUS2D predicted) was included in the likelihood function during this research, and the posterior probability distributions for the hydrological parameters fit the data better with less variable posteriors. However, considering that the posteriors were estimated based on a limited data set for 16 days, more variable probability distributions obtained without incorporation of temporal correlation were considered more appropriate as inputs for a general coal fly ash embankment model.

Coal fly ash scenario model

Contaminant leaching was simulated for 10 years for a hypothetical coal fly ash embankment scenario where 0.50 m of coal fly ash was placed on top of the MnROAD embankment material modeled in previous simulations (Fig. 1c). The groundwater table was set at 1.9 m, which is within the range (1.3 to 4.6 m) observed at the MnROAD test site. An entire year's precipitation data repeated 10 times was input as the variable flux boundary condition. The molecular diffusion coefficient of Cadmium in free water was input in the model as a constant (6.2×10^{-5} m^2/day) (Li and Gregory, 1974) and tortuosity factor was calculated within HYDRUS2D as a function of the water content using Millington and Quirk's (1961) equation ($\tau = \theta^{7/3}/\theta_s^2$).

The probability distributions of unsaturated hydraulic properties of the embankment, given in Table 2, were determined from parameter posterior probabilities obtained from embankment infiltration simulations. Probability distributions were fit to the four parameters (θ_s, α, n, K_{sat}) based on the posterior probabilities generated from HYDRUS2D simulations that passed the profile likelihood ratio test. Weighted moment equations were applied to calculate the means and standard deviations for the normal distributions (Table 2). Saturated hydraulic conductivity and saturated water content were assigned joint lognormal distributions with correlation ($\log(\theta_s)$,$\log(K_{sat})$) = 0.87.

Probability distributions for all other parameters were based on literature data. Probability distributions for α, n, θ_s, θ_r, K_{sat}, and bulk density of the coal fly ash were based on six sources of coal fly ash with measurements on both drying and wetting of the samples (Young, 1993). The saturated hydraulic conductivities reported in Young (1993; 10^{-2}–10^{-1} m/day) are similar to those reported by Bowders *et al.* (1987; 10^{-2} m/day) but higher than those reported in Vesperman *et al.* (1985; 10^{-7} m/day) and Creek and Shackelford (1992; 10^{-5}–10^{-3} m/day). The bulk density distribution for the embankment was estimated by measurement of the subgrade material in this research (1.88 kg/L) and by Bigl and Berg (1996) (1.74, 1.69, 1.84 kg/L).

Partition coefficients reported in soil and coal fly ash vary three to four orders of magnitude for different conditions of pH and liquid to solid ratios. In unsaturated conditions, the pH of secondary materials during leaching may vary more than in saturated conditions (Townsend *et al.*, 1999). To account for the variability, uniform distributions (Table 2) were assigned to partition coefficients based on the values reported by U.S. EPA (1999) and van der Sloot et al. (1992) for soil and coal fly ash, respectively. The temporal and spatial variability of K_d that would be expected in the field was incorporated in the modeling approach by probabilistically varying K_d values of the subgrade and the coal fly ash for each simulation. The value of K_d is often considered as a measure of the strength of the adsorption of a contaminant on the soil. In this research, K_d is the lumped parameter for multiple processes such as dissolution/precipitation, surface complexation, surface precipitation, and diffusive transfer from particle core to the bulk solution. Use of K_d in the finite element model can be viewed as a method for interpreting pH and liquid to solid specific leachate data.

Our decision to use K_d values for predicting contaminant release is supported by the work of Bin-Shafique *et al.* (2002) on another metal; selenium. Bin-Shafique *et al.* (2002) estimated material specific K_d values from laboratory column studies and then used these estimates in HYDRUS2D to predict leaching from coal fly ash-stabilized pavements. Field data was available for only 2 years; yet a deterministic HYDRUS2D simulation accurately predicted field release for Selenium for this short period.

Uncertainty in parameters was propagated through the model by running HYDRUS2D with parameters ran-

Table 2. Probability distributions for unsaturated parameters.

Coal fly ash	*MnROAD embankment*
Log (θ_r) ~ Normal (−2.881, 0.559)	θ_r = 0.25
θ_s ~ Normal (0.455, 0.035)	Log (θ_s) ~ normal (1.033, 0.031)
α ~ Uniform (0.08, 0.45)	α ~ Normal (3.788, 0.673)
n ~ Normal (2.567, 0.378)	Log (5 − n) ~ normal (0.113, 0.186)
Log (K_{sat}) ~ Normal (−3.18, 0.96)	Log (K_{sat}) ~ normal (−1.125, 0.315)
K_d (L/kg) ~ Uniform (0.3, 2000)	K_d (L/kg) ~ Uniform (1, 4000)
ρ_b ~ Normal (1.303, 0.109)	ρ_b ~ Normal (1.756, 0.074)
τ ~ Uniform (0.1, 0.5)	
D_i ~ Uniform (0.05, 0.36)	

domly sampled from the parameter probability distributions. To be able to run consecutive automated simulations randomly sampled from probability distributions, the initial aqueous concentration assigned to the coal fly ash was set to unity. HYDRUS2D allows input of initial aqueous concentration and calculates the equilibrium solid concentration of the contaminant based on the K_d value. As long as the simulation results can be normalized, the absolute value of the concentration is not important since the concentration term appears throughout the advection dispersion equation. Contaminant leaching was evaluated by comparing the total mass of Cadmium leached across a point 0.01 m below the coal fly ash to the initial Cadmium mass input in the HYDRUS2D.

RESULTS AND DISCUSSION

Updating of uncertainty

A majority of the posterior probabilities were much lower than 10^{-6}, suggesting that many of the parameter combinations were unlikely to be representative of the system modeled (Fig. 2). Applying the likelihood ratio criteria, simulations with posterior probabilities less than 10^{-6} were eliminated leaving 510 simulations (with total posterior probability of 0.999345) that were further analyzed statistically. Only three simulations had posterior probabilities greater than 0.1.

The summary statistics of the prior and posterior distributions are shown in Table 1. The mean of the distribution of n increased, while its standard deviation decreased in the posterior estimates. The standard deviation of α increased, suggesting that the prior distribution assigned to this parameter was too tight. The mean and the standard deviations of K_{sat} and θ_r distributions decreased after Bayesian updating. The posterior mean of K_{sat} is on the verge of the range of the prior distribution suggesting it is possible that K_{sat} may have a lower value. Laboratory measured values by Bigl and Berg (1996) (0.0002–0.003m/day) also included much lower values for K_{sat}. However, K_{sat} is probably not much lower than 0.03 m/day since preliminary Bayesian analysis of this problem had eliminated the possibility of these low values representing field saturated hydraulic conductivity. The results of the Bayesian analysis suggest that embankment hydraulic conductivity may be higher than laboratory measured values, possibly due to preferential flow paths.

Many authors noted that calibrated parameters may not be unique (Poeter and Hill, 1997; Lambot *et al.*, 2002; Valota *et al.*, 2002). The Bayesian updating technique inherently addresses this concern by probabilistic conceptualization of the calibration problem. First, all parameters were varied (except θ_r simultaneously since the parameter space was sampled in all four dimensions. The updated parameters are representative of all combinations within the parameter ranges sampled from. This approach is more representative of the parameter space than varying one parameter at a time. Second, the updated parameter values are not viewed as unique solutions where the possibility of other parameters representing the system is mainly ignored. In the Bayesian updating approach, the possibility of other values of the parameters is viewed as the uncertainty related to the model, model parameters, and the modeler. Bayesian posteriors, which define the most relevant regions of the multidimensional

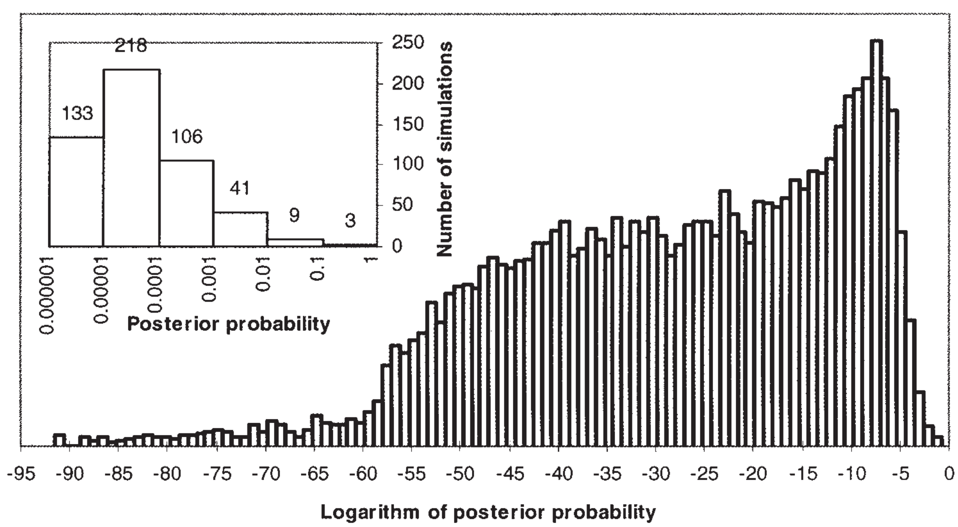

Figure 2. Histogram of posterior probabilities of all simulations and distribution of accepted posterior probabilities (inset).

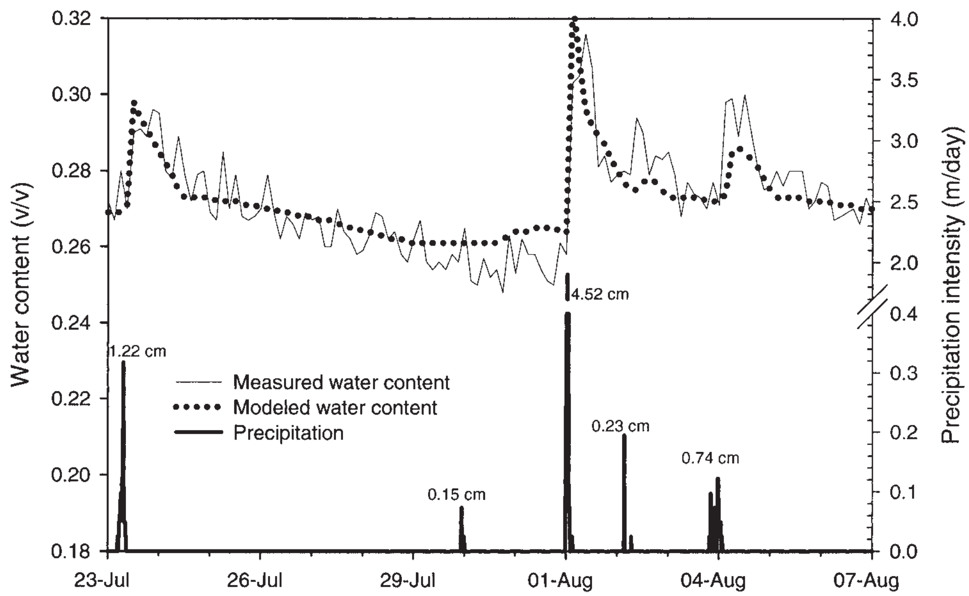

Figure 3. Modeled and measured water content data at 0.32 m from the surface and the corresponding precipitation quantity and intensity for a 16-day period in 1997.

parameter space for this particular problem, contain all information about the system such as prior expectations of the parameters and the measured field water content data.

The posterior means of the parameters provide a reasonable match between the measured and simulated water content data (Fig. 3), and similar results were observed when the posterior means were tested on another set of time series available for the same embankment (Fig. 4). Presence of only a single data point for calibration is one weakness of the current research compared to other studies where close matches between measured and modeled water contents were also observed in controlled field and

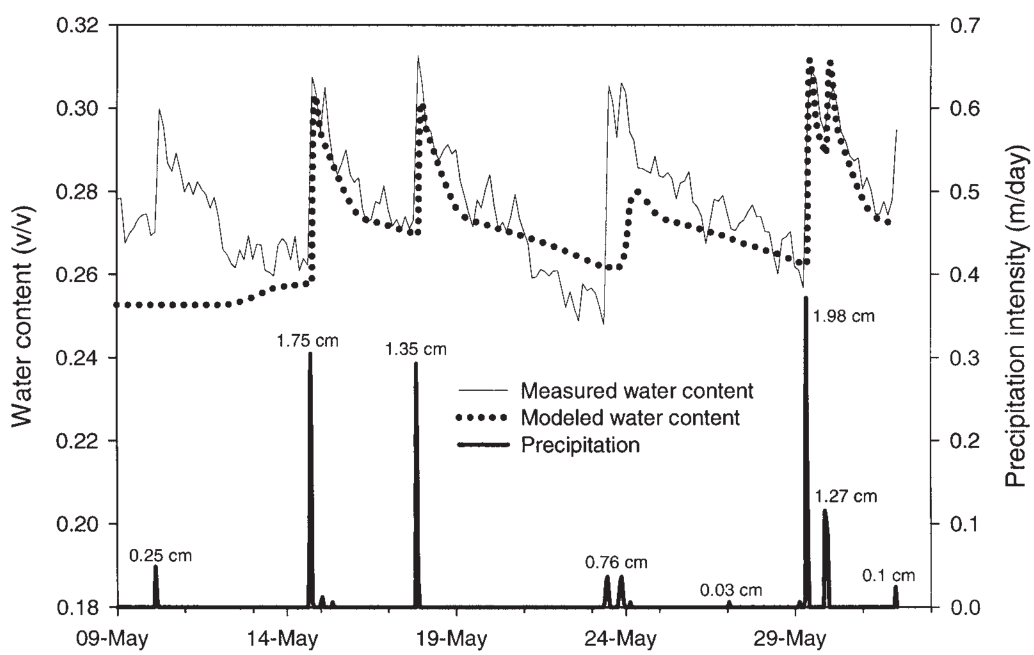

Figure 4. Modeled and measured water content data at 0.32 m from the surface and the corresponding precipitation quantity and intensity for a 23-day period in 1998.

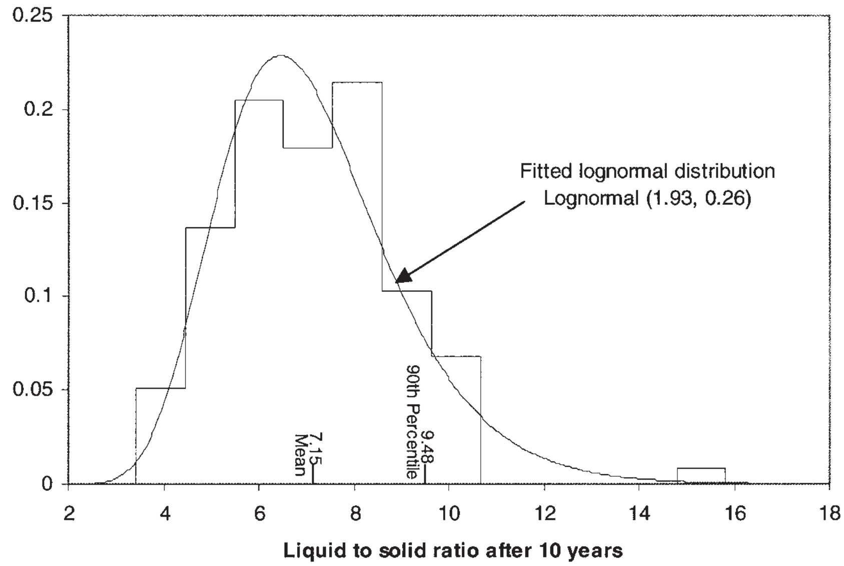

Figure 5. Histogram and fitted probability distribution for liquid-to-solid ratio after 10 years.

laboratory experiments (Jacques *et al.*, 2002; Lambot *et al.*, 2002). In this research, overall, the model adequately reproduced water content for precipitation events exceeding 0.01 m, but underpredicted the response of smaller rain events. Multiple factors may be the cause of the minor deviations between the model output and field data. Presence of lateral flow, limitations of the van Genuchten model, evaporation, and hysterisis may have contributed to the deviations between measured and modeled output. Acting as an impermeable boundary, the TDR rod might also affect the local distribution of water content (Ferre *et al.*, 2002). The impermeable volume of the TDR rod was not considered in the HYDRUS2D model.

Coal fly ash simulations

Infiltration and liquid-to-solid ratio. Water and Cadmium mass balance errors in the 113 randomly sampled simulations were less than 2.7 and 1.0%, respectively.

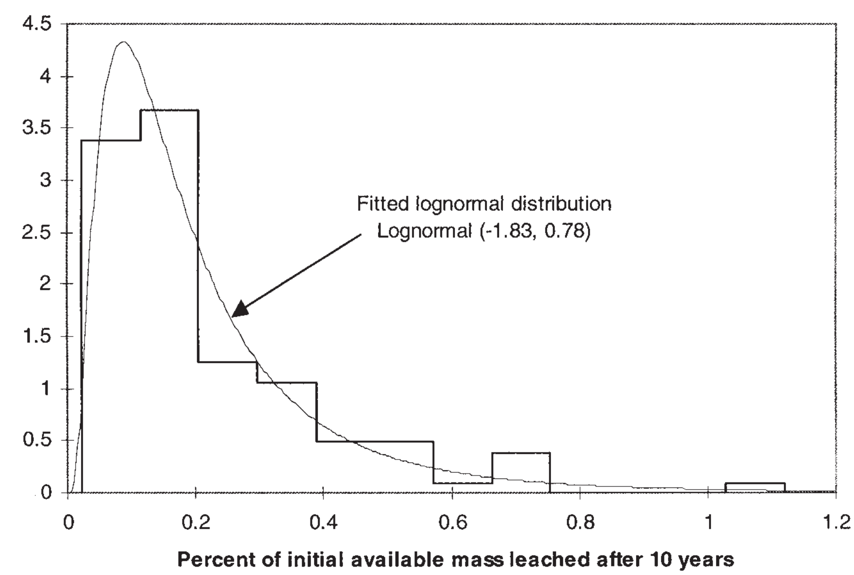

Figure 6. Histogram and fitted probability distribution for percent of initial available mass leached after 10 years (as observed 0.01 m below coal fly ash).

The total potential infiltration (annual precipitation) was 0.65 m/year, and 92% (0.60 m/year) of this flux actually infiltrated into the embankment at the 90th percentile of uncertainty. Simulation results showed that the actual infiltration could be as low as 34% (0.22 m/year) of the potential infiltration, while the mean infiltration was 71% (0.46 m/year). Hjelmar (1990) measured percentage of precipitation passing through large-scale lysimeters filled with slightly compacted coal fly ash. The values reported by Hjelmar (58, 53, 55, 42, 46, 59, and 61%) for multiple lysimeters and two different types of fly ashes are slightly lower than the mean value estimated in this research possibly due to differences in site conditions.

The liquid-to-solid ratio is a measure of the amount of water that has passed through the secondary material application, and is calculated by dividing the product of time and flux of water through the material by the mass of the material. The mass of the secondary material varied among simulations due to randomly sampled bulk density values. The liquid-to-solid ratio in 10 years was 7.15 and 9.48 L/kg on average, and at the 90th percentile of uncertainty, respectively (Fig. 5). The liquid-to-solid ratios measured by Hjelmar (1990) normalized to 10 years and 0.50 m of coal fly ash are 4.6, 4.85, and 3.92 L/kg. Higher liquid-to-solid ratios estimated in this research may be due to differences in climate, groundwater table depth, hydraulic conductivity, and material below the coal fly ash (low density polyethylene liner versus soil).

Leaching from coal fly ash. The average percentage of initial available mass leached after 10 years, as observed 0.01 m below ash, was 0.21 ± 0.20%. As also suggested by the high value of the standard deviation, the probability distribution is skewed to the left (Fig. 6). At the 90th percentile of uncertainty, the percentages of initial available mass leached are 0.02, 0.20, and 0.48% for 1, 5, and 10 years, respectively (Fig. 7). No significant Cadmium fluxes were observed 0.25 m below the coal fly ash or at the groundwater table depth. After 10 years, the fraction of initial available mass leached was 5×10^{-6} percent at 0.25 m below the coal fly ash, and 0% at the groundwater table depth (at the 90th percentile of uncertainty).

Results of HYDRUS2D simulations were so far expressed in percentage of initial available mass leached. These results can also be interpreted in mg/kg units by use of the following equation:

$$\text{Release (mg/kg)} = PL \times T_{\text{Cd}} \times \text{MA} \qquad (12)$$

where PL is the percentage of initial available mass leached [−], T_{Cd} is the total concentration of Cadmium in the sample [mg/kg], and MA is the maximum availability of Cadmium expressed as percentage of total contents [−].

Probability distributions were assigned to each term in equation (12). A lognormal distribution was fit to the percent leached data (Fig. 6). The total Cadmium content in

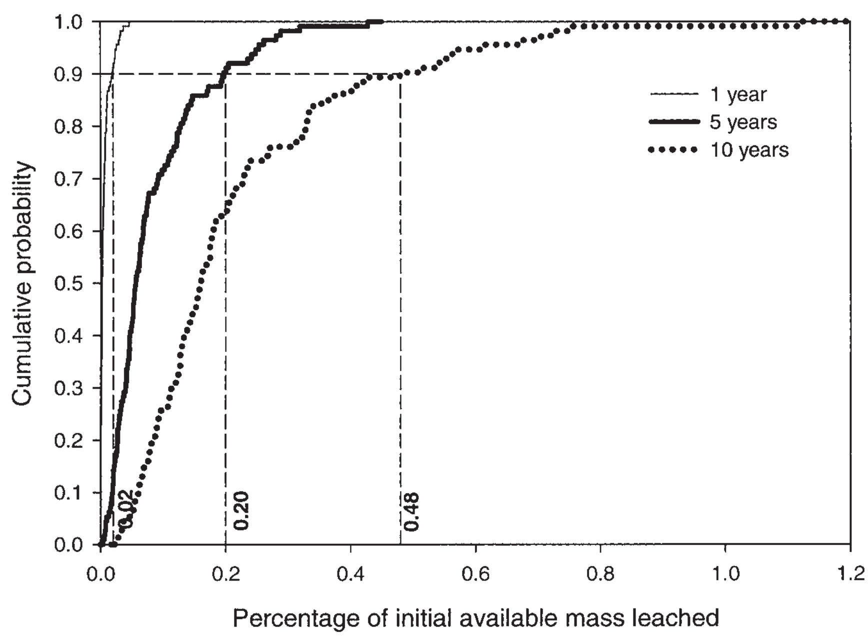

Figure 7. Cumulative probabilities of percentages of initial available mass leached (as observed 0.01 m below coal fly ash) after 1, 5, and 10 years.

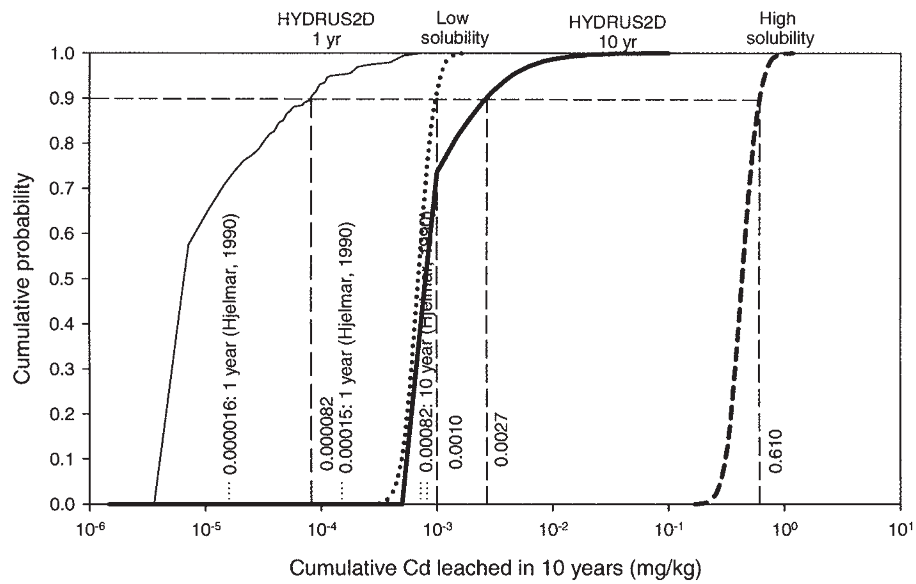

Figure 8. Cumulative probabilities of mass of Cadmium leached.

coal fly ash was modeled as a lognormal distribution ($\log(T_{Cd}) \sim \text{normal}(0.3, 1.14)$) based on values reported for 95 coal fly ash sources reported in the literature (Wu and Chen, 1987; van der Sloot et al., 1991, 1992; Schwab, 1993; Garavaglia and Caramuscio, 1994; Alva *et al.*, 1999a, 1999b; Khandekar et al., 1999; Mukherjee and Kikuchi, 1999; Twardowska, 1999a, 1999b; Brunori *et al.*, 2001; Hassett *et al.*, 2001). The percentage of maximum observed availability of total content was modeled with a uniform distribution with minimum and maximum values of 5% and 36%, respectively (van der Sloot *et al.*, 1992; Kim and Kazonich, 2001; Chaudhuri *et al.*, 2003).

The cumulative release at the 90th percentile of uncertainty, calculated using equation (12) and the appropriate probability distributions, was 2.65×10^{-3} mg Cd/kg ash after 10 years (Fig. 8). The mean of the release estimate was 1.15×10^{-3} mg Cd/kg ash. Hjelmar (1990) measured Cadmium concentrations in coal fly ash lysimeters and reported below detection limit concentrations (<0.0001 mg/L) after 0.5, 2.6, and 2.6 years for three different lysimeters. To estimate total release of Cadmium in 10 years, we assumed (1) detection limit concentrations for those times when Hjelmar's (1990) measurements were below detection limit, and (2) a value of 7 L/kg for the liquid to solid ratio. This approach normalizes the comparison to the liquid to solid ratio as opposed to the number of years. With this conservative approach, the total release estimates extended to 10 years vary from 7.2×10^{-4} to 8.2×10^{-4} mg Cd/kg ash. Thus, the mean of the HYDRUS2D simulation results were within one order of magnitude of the estimate from Hjelmar's (1990) field experiments when his results were extrapolated conservatively to 10 years.

A more direct comparison of HYDRUS2D results with field values is possible for the first year where field measurements from Hjelmar (1990) were not below detection limit. Hjelmar (1990) presented results for the 10th and 14th months for three different lysimeters which released 1.6×10^{-5} to 1.5×10^{-4} mg Cd/kg ash. HYDRUS2D results for the 90th (8.2×10^{-5} mg Cd/kg ash) percentile and the mean

Table 3. Comparison of deterministic applications of the two approaches for calculating release.

	Percolation equation	*HYDRUS2D simulations*
Considers diffusion?	No	Yes
Considers dispersion?	No	Yes
Considers multiple layers?	No	Yes
Considers tortuosity?	No	Yes
Solubility assumption	Constant	Varies linearly with the partition coefficient
Infiltration	Obtained from other resources	Calculated
Source of release	Point source	Release throughout the material

(3.2×10^{-5} mg Cd/kg ash) for the first year of leaching fall within the range observed by Hjelmar (1990) (Fig. 8).

Comparison with the percolation equation method. In the percolation equation suggested by Kosson *et al.* (1996) the cumulative mass released in is calculated from the product of solubility of Cadmium and the cumulative liquid to solid ratio.

$$\text{Release (mg/kg)} = S_{\text{Cd}} \times \text{(Liquid-to-Solid Ratio)} \quad (13)$$

where S_{Cd} is the solubility of Cadmium [mg/L].

The underlying assumption of this approach is that Cadmium will be transported at its solubility value and the advection rate is equal to the annual infiltration rate. Spatial and temporal scales are not explicitly considered in this equation; the release is assumed to be a point source with constant concentration transported at the infiltration rate. This approach is a simplified application of the HYDRUS2D simulations (Table 3).

The distribution assigned to the liquid-to-solid ratio for use in the percolation equation method was the lognormal distribution fitted to the HYDRUS2D outputs (Fig. 5). Leachate Cadmium concentrations from coal fly ashes can vary from 0.0001 to 0.063 mg/L as a function of pH and liquid-to-solid ratios (van der Sloot *et al.*, 1992). When the low value of leachate Cadmium concentration was used, the release estimate from the percolation equation was within one order of magnitude of the release estimate from the HYDRUS2D simulation at the 90th percentile of uncertainty (Fig. 8). The release estimate from the high value of leachate Cadmium concentration was two to three orders of magnitude higher than the other estimates. Figure 8 shows that the release estimates predicted from Hjelmar's (1990) field lysimeters data are close to the HYDRUS2D results, and the percolation equation results estimated from the low Cadmium solubility value. Use of high solubility values results in a significant over predictions of Cadmium release.

CONCLUSIONS

In this research, the dynamics of the water content in a highway embankment was simulated using a probabilistic Bayesian updating approach for parameter calibration. Updated parameters and literature values were used to predict release of Cadmium from a hypothetical coal fly ash embankment using two different approaches. If a low solubility value is used for Cadmium, the release estimates are close to those measured by Hjelmar (1990) and also predicted from HYDRUS2D simulations. If high solubility is used, the percolation equation significantly overpredicts the actual release. The use of the advection dispersion equation for transport and retardation in unsaturated medium is a powerful method for predicting contaminant release, especially when it is coupled with probability. One implication of the use of the advection dispersion equation is the ability to extend the release estimates to two-dimensional systems where the percolation equation may no longer be applicable due to lateral flows and more complicated hydraulic regimes. The authors recommend application of the proposed approach to two dimensions to more realistically predict both the water flow and the contaminant release and transport processes in highway environments.

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