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## NUMERICAL SIMULATION OF CHEMICAL TRANSPORT IN A PHYSICALLY HETEROGENEOUS SOIL PROFILE

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### ABSTRACT

Contaminated sediments stored above the soil are a typical example of a long-term source of soil pollution, for the potentially harmful chemicals present in the sediments may leach below the soil zone and ultimately contaminate shallow or deep aquifers. Risk assessment studies related to contaminated land often use the simple one-dimensional convection-dispersion model (CDE) to quantify the leaching hazard associated with disposed dredged material. Rather than using data from physical experiments, the testing of the simple model is done here by using data obtained through numerical simulation using high-performance computing techniques. Therefore, we generated two-dimensional hypothetical random fields of soil hydraulic properties which represent a heterogeneous unsaturated field soil profile whose properties are exactly known at a high spatial resolution. Following general conclusions were drawn from the study: Numerical calculations are useful in testing simple models, such as the CDE hypothesis, using procedures presented in this study. It remains difficult however to describe field-scale transport exactly using approximate models.

### KEYWORDS

convection-dispersion equation; chemical transport; heterogeneous porous media; leaching hazard; numerical simulation; random fields; risk assessment

### INTRODUCTION

The disposal of contaminated dredged material to land poses a long-term environmental threat, for toxic chemicals may leach out of dredged sediments and migrate through the soil to the groundwater and surface water. Assessing the risk of groundwater contamination from such disposal sites requires transport models in addition to site specific flow and transport parameters. Due to the natural variability of soils, modelling of the fate of contaminants should account for the heterogeneity in physical (e.g. permeability) and chemical (e.g. distribution coefficient) soil properties (Weber et al., 1992). In the present study we discuss how to incorporate spatial variability in hydraulic functions (the so-called physical heterogeneity) into a high-resolution two-dimensional solute transport model. The settings for the simulation domain are shown in Figure 1 and consist of a leaching disposal site on top of a heterogeneous soil with a relatively deep groundwater table. We further demonstrate that by means of a stochastic numerical simulation a set of high-quality solute concentration data may be generated which can serve the purpose of testing a more simplified one-dimensional transport model with macroscopic or effective parameters. Such simple models, when thoroughly tested, may be used on a daily basis in risk-assessment studies. Limitations of the simplified model are also highlighted.

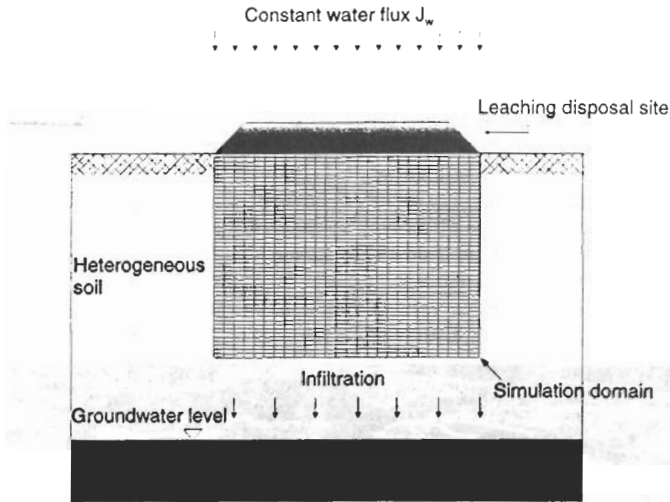


Figure 1. Schematic view of simulation domain for hypothetical disposal site.

## MATERIALS AND METHODS

### Soil Heterogeneity

Soils are known to have heterogeneous physical, chemical, and biological properties. Different properties have different degrees of variability (Jury, 1985). Heterogeneous properties relevant to contaminant transport through soils include hydraulic functions describing the water retention characteristic and the (un)saturated hydraulic conductivity (Tseng and Jury, 1994). Chemical properties such as sorption behaviour also exhibit spatial variability and cause additional spreading of the contaminant plume (Bosma et al., 1993).

In the present study the spatial variability is restricted to the physical properties, i.e., the parameters of the water retention relationship and the unsaturated hydraulic conductivity function. The first property,  $\theta(h)$ , describes the potential of the soil to retain water and is described here as (van Genuchten, 1980):

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

where  $\theta$  is soil water content ( $\text{cm}^3 \text{cm}^{-3}$ ),  $h$  is pressure head (cm), subscripts s and r denote saturated and residual water content,  $\alpha$  ( $\text{cm}^{-1}$ ),  $n$ , and  $m$  are curve shape parameters, with  $m = 1 - 1/n$ . The second property,  $K(h)$ , is used to represent the capacity of a soil to transmit water (van Genuchten, 1980):

$$K(h) = K_s \frac{([1 + |\alpha|h|^n]^m - |\alpha|h^{n-1})^2}{[1 + |\alpha|h|^n]^{5m}} \quad (2)$$

where  $K_s$  ( $\text{cm day}^{-1}$ ) is saturated hydraulic conductivity,  $l = 0.5$  and all other parameters are as defined previously.

Spatial variability in  $\theta(h)$  and  $K(h)$  in a two-dimensional space was described using the theory of microscopic geometric similitude, in which case the field heterogeneity is characterized by a single scaling factor. The local hydraulic properties in each of the nodes of the spatial domain are derived from reference properties defined here by the mean values of the hydraulic functions. The parameters for the mean  $\theta(h)$  and  $K(h)$  were  $\theta_s = 0.4$ ,  $\theta_r = 0.05$ ,  $\alpha = 0.019$ ,  $n = 1.59$ , and  $K_s = 144 \text{ cm d}^{-1}$ .

### Synthetic Heterogeneous Field

Using the turning bands method (Tompson et al., 1989), we constructed a two-dimensional random field of a lognormally distributed scaling factor  $\delta$  representing a 7.5 m wide and 4 m deep hypothetical soil profile. The spatial correlation structure of  $Y = \ln \delta$  was described with an anisotropic exponential correlation function, using a horizontal and vertical correlation length of  $L_x = 0.5$  and  $L_z = 0.1$  m, respectively. The variance of  $Y$ ,  $\sigma^2_Y$ , was chosen to represent a very heterogeneous soil, i.e.  $\sigma^2_Y = 1$ . Because the variances of  $Y$  and  $\ln K_s$  are related through  $\sigma^2_{\ln K_s} = 4\sigma^2_Y$ , we impose the variability in  $\ln K_s$  as being  $\sigma^2_{\ln K_s} = 4$ . Soil hydraulic functions (1) and (2) were described at each of the 15,762 grid points making up the uniform grid system with horizontal and vertical spacing of 10 and 2 cm, respectively. Figure 2 shows the synthetic random field of the scaling factors  $\delta$ . Dark zones correspond to sandy sediments (high permeability) whereas lighter zones represent clay layers. Note how the clay and sand layers alternate along the vertical direction, which is a direct result of the short vertical correlation length relative to the soil depth. Estimated correlograms compare well with the theoretical ones.

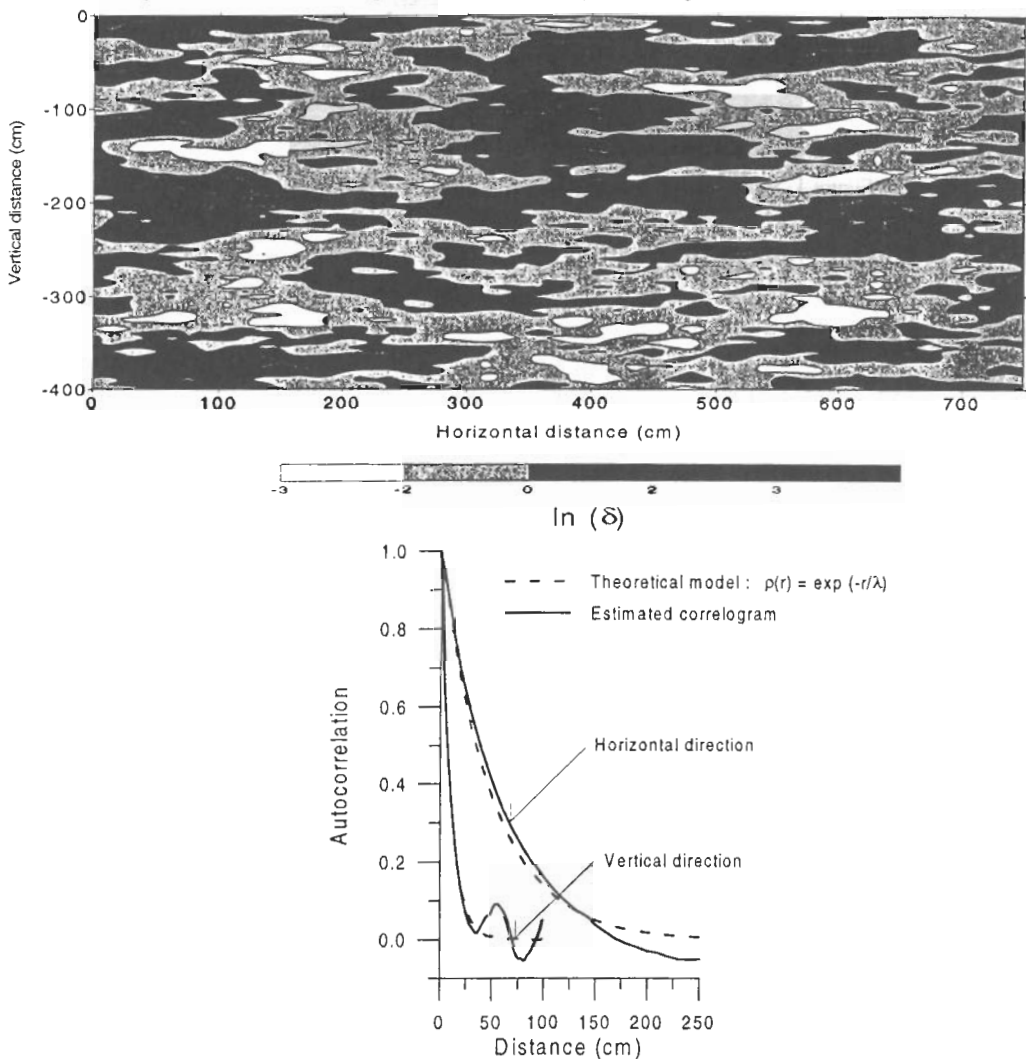


Figure 2. Synthetic heterogeneous soil profile (top), and estimated correlograms (bottom).

## Water Flow and Contaminant Transport

Two-dimensional water flow in a variably-saturated soil profile was described by Richards equation:

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial x_i} \left[ K(K_{ij} \frac{\partial h}{\partial x_j} + K_{ij}) \right] \quad i=1,2 \quad (3)$$

where  $x_i$  are spatial coordinates ( $x_2 = z$  is taken positive upward),  $K_{ij}$  are components of the dimensionless anisotropic tensor  $K$ , and  $t$  is time.

Solute transport in the heterogeneous vertical cross section is described by the CDE as:

$$\frac{\partial(\theta C)}{\partial t} = \frac{\partial}{\partial x_i} [\theta D_{ij} \frac{\partial C}{\partial x_j}] - J_w \frac{\partial C}{\partial x_i} \quad (4)$$

where  $C$  is the solute concentration in the fluid phase ( $\text{g}\cdot\text{cm}^{-3}$ ),  $D_{ij}$  is the dispersion tensor ( $\text{cm}^2\cdot\text{d}^{-1}$ ), and  $J_{wi}$  is the  $i$ th component of the water flux.

Flow and transport equations (3) and (4) were solved numerically using the finite element FORTRAN code SWMS-2D (Šimůnek et al., 1994). We assumed constant transverse and longitudinal dispersivities of 0.02 and 0.25 cm, respectively. In the numerical experiment we applied a constant water flux  $J_w = 1.44 \text{ cm}\cdot\text{d}^{-1}$  to the surface of the heterogeneous soil profile until steady state flow was established. Next, a  $0.1 \text{ g}\cdot\text{cm}^{-3}$  tracer pulse was applied during one day. Following the addition of the solute, water flux  $J_w$  continued to be applied for another 150 days until the solute plume approximately reached the bottom of the profile.

## RESULTS AND DISCUSSION

### Concentration Profiles

Spatial variability in hydraulic properties will generate tortuous, three-dimensional flow patterns of chemicals leaching from the surface to the groundwater. This can be seen from the solute concentration distributions ( $\text{g}\cdot\text{cm}^{-3}$  soil) in a heterogeneous soil at time = 15, 35, and 50 days after tracer application (Figure 3). Such concentration profiles are generally obtained through soil coring or by using the time domain reflectometry (TDR) technique (Mallants et al., 1996). Note that a sufficient number of vertical cores needs to be sampled in a contaminated soil for a proper characterization of the degree of pollution. Also shown in Figure 3 are the first, the last, and the mean breakthrough curve (BTC), where the latter has been averaged over 76 computational nodes for each depth interval. Close to the surface the BTCs are relatively smooth and symmetric. Highly irregular BTCs including the mean curve are seen deeper in the soil profile (time = 50 days) indicating an extreme heterogeneous and complex flow process. A comparison between the first and last arrivals of the peak concentrations at time = 50 days indicates that the former is approximately 100 cm ahead of the latter. These curves give an insight into the heterogeneous nature of flow and transport processes in soils. Moreover, they clearly illustrate that the local heterogeneities cannot be modelled by means of a simple one-dimensional transport model, whose application is necessarily limited to the description of the mean field-scale transport.

### Testing of simple models

Risk assessment studies dealing with the movement of pollutants through unsaturated soils have mostly been performed using one-dimensional models based on the CDE. Field-scale variability may be included by running a multitude of simulations in parallel using local values of transport parameters followed by averaging the results. Another approach would be to use effective or field-scale mean transport parameters and run the model only once. The latter approach was tested here using the detailed set of data from the numerical experiment. The macroscopic CDE with effective or macroscopic parameters is given as (Toride and Leij, 1996):

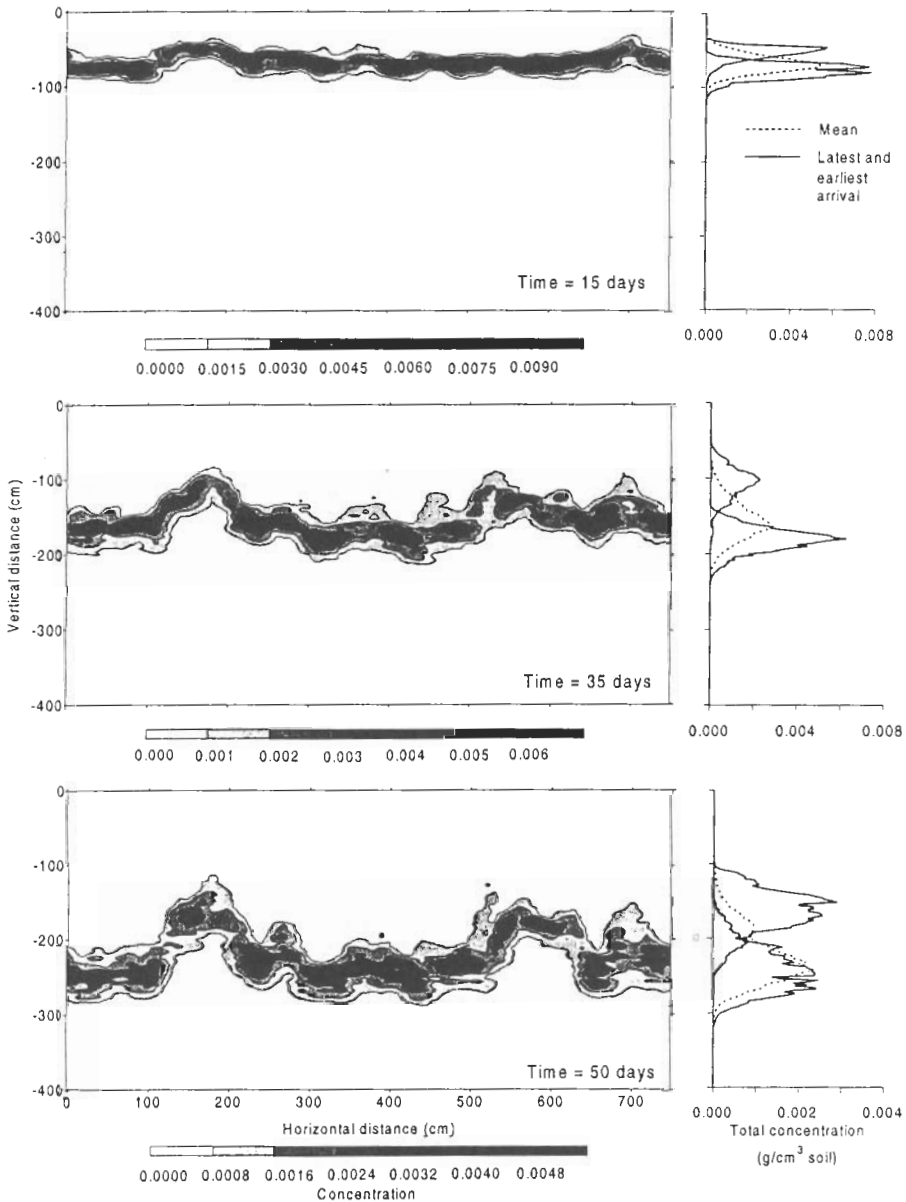


Figure 3. Resident concentration profiles at three selected times (left), and earliest, latest, and mean breakthrough curves (right).

$$\frac{\partial \langle C' \rangle}{\partial t} = D_{eff} \frac{\partial^2 \langle C' \rangle}{\partial z^2} - v_{eff} \frac{\partial \langle C' \rangle}{\partial z} \quad (5)$$

where  $C'$  is resident concentration,  $D_{eff}$  and  $v_{eff}$  are effective dispersion ( $\text{cm}^2 \text{d}^{-1}$ ) and pore-water velocity ( $\text{cm d}^{-1}$ ), respectively. The effective parameters are assumed constant with depth. We tested the predictive capacity of this simple model by calibrating the parameters at a shallow depth (or at early time) using the mean observed BTC, followed by a comparison between predicted and observed mean BTC at greater depths (or later times).

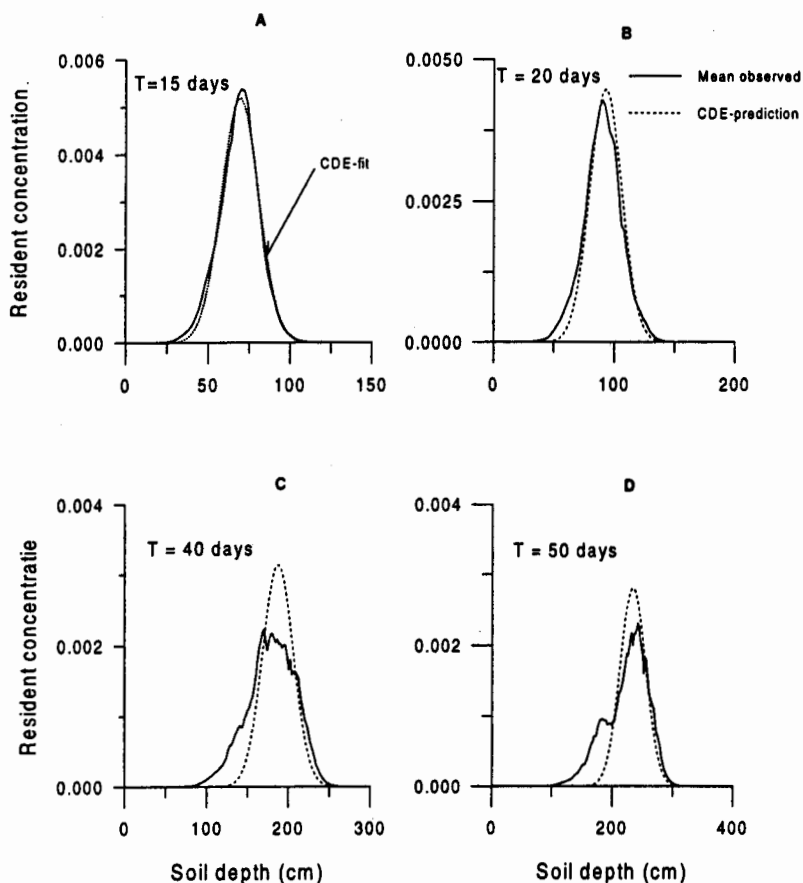


Figure 4. Predicted and observed mean resident concentration distributions at selected times. Calibration of CDE was done at time = 15 days (A).

Figure 4 shows that the fitted CDE with effective parameters accurately describes the mean observed resident concentration distribution at time = 15 days. Predicted mean BTCs at time = 20, 40, and 50 days together with observed curves are also given. At a depth close to the calibration depth the agreement is still good. Furthermore, the mean arrival time of the solute plume at all depths is well described by means of the macroscopic CDE. However, deeper in the profile the predicted curves overestimate the peak concentration and underestimate the observed tailing. This is the result of using a constant dispersion with depth, which is a requirement of the CDE hypothesis. Solute spreading or dispersion at greater depths is thus underestimated as the true dispersion or dispersivity,  $\lambda = D/\nu$  (cm), keeps on increasing with increasing depth (Figure 5). This behaviour of the dispersion coefficient is known as the dispersion-scale effect and represents the transition of an early time process without lateral solute mixing (stochastic-convective transport) to an infinite time process with complete mixing (convective-dispersive transport).

Although the current analysis did not consider reactive chemicals, sorption and desorption processes, degradation and volatilization can be easily incorporated in the numerical simulation as well as in the macroscopic CDE. In such case spatial variability in chemical properties such as the distribution coefficient will also have to be accounted for.

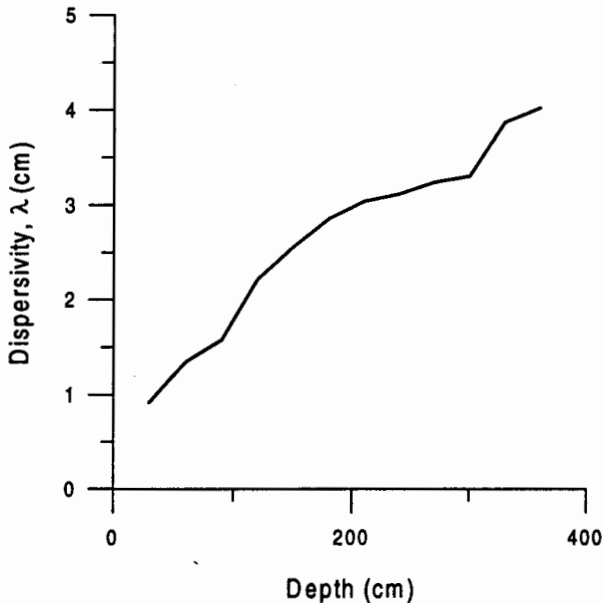


Figure 5. Growth of dispersivity with travel depth.

## CONCLUSIONS

We demonstrated how spatial variability in soil hydraulic properties can be incorporated in a numerical calculation of flow and transport processes at the field scale. Although such detailed numerical simulations using high-performance computing techniques may be used for location specific risk assessment of disposal sites, they require considerable computational resources in addition to detailed information on the spatial variability of the hydraulic functions. These requirements restrict somewhat the applicability of the sophisticated two- or three-dimensional models for management or regulatory purposes. The alternative is to use simplified one-dimensional models with average properties, such as the macroscopic CDE. The limitations of this transport model were illustrated using hypothetical though realistic data on leaching of chemicals from a disposal site. The random field represented a very heterogeneous soil profile with a distinct layering illustrative of a mixture of sand and clay sediments. Predictions with the CDE model calibrated at shallow depth overpredicted the solute peak and underpredicted the spreading at greater depths. In other words, the true field-scale dispersion at greater depths was larger than the dispersion obtained through calibration at the shallow depth. The dispersion-scale effect persisted up to a depth of at least 4 m, which corresponds to 40 vertical correlation lengths of the hydraulic conductivity. Generalization of the results obtained here is limited because of the specific flow regime established following the imposed field heterogeneity. Nevertheless, numerical experiments may help improve our understanding of field-scale transport and may be helpful in defining limitations of simple transport models.



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