The effect of field-scale solute infiltration into groundwater on surface water quality

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Abstract The effect of field-scale infiltration on surface water quality is analysed. A hypothetical example, where solute leaches from a heterogeneous field and is transported through the aquifer to surface water, is discussed. Both conservative and reactive solutes are considered, where the assumed reaction is first order non-equilibrium sorption-desorption. The impact of spatial variability in the unsaturated zone on uncertainty in predicting water quality changes is generally significant. The effect is more pronounced for a conservative than for a reactive solute.

INTRODUCTION

The impact of agricultural activities on water quality is a problem of growing interest. Different substances that are soluble in water are spread over large areas and may leach through the unsaturated zone into the groundwater. Therefrom, the solute follows groundwater movement, eventually reaching surface water and public supply wells. Decisions on appropriate administrative and technical measures are based on the assessment of potential hazard that may arise due to groundwater quality changes. In order for the potential hazard to be assessed in an objective manner, the uncertainty in predictions of regional solute transport needs to be quantified.

Regional transport of a reactive solute is generally a complex process because many different mechanisms are involved. The spatial heterogeneity in
both the unsaturated and saturated zone, however, is recognized as one of the important factors for field-scale solute movement (Dagan & Bresler, 1984; Nielsen et al., 1986). Furthermore, most of the substances used in agriculture, such as herbicides, pesticides and fertilizers, are known to react chemically and biologically in the soil. Thus, the chemical and biological reactions in the unsaturated zone may play a significant role in determining solute transport (Dagan & Bresler, 1984; Nielsen et al., 1986).

In this investigation, the effect of field-scale solute infiltration from an areally distributed source on surface water quality is to be analysed using a hypothetical example where a reactive solute leaches from a heterogeneous field and is transported through the aquifer to surface water (e.g. a lake). The uncertainty in predictions of groundwater quality changes due to the spatial variability in the unsaturated zone is to be quantified.

A simplified, steady-state flow model coupled with an analytical transport model that incorporates advection, dispersion and sorption-desorption with first-order linear kinetics is used for the unsaturated zone. A two-dimensional, areal groundwater flow and transport model for the aquifer (which in the present context refers to the saturated groundwater zone) links the source area with the surface water. In order to focus on the effects of solute infiltration, the heterogeneity and the sorption-desorption reactions are assumed in the unsaturated zone only.

The breakthrough of both a conservative and a reactive solute into the surface water is illustrated by considering Monte Carlo realizations of solute influx from vertical columns into the groundwater as a model for the field heterogeneity. The expected solute breakthrough is then analysed in order to quantify the uncertainty associated with the field-scale heterogeneity.

FLOW AND SOLUTE TRANSPORT IN THE UNSATURATED ZONE

For the unsaturated zone we assume conditions analogous to those in Bresler & Dagan (1981), namely that the field consists of a collection of vertical, independent soil columns, where each column is vertically homogeneous (i.e. soil property variations with depth are neglected and depth-averaged values are used). Furthermore, a steady gravitational water flow, vertical in each column, is assumed. Thus, the vertical specific discharge is constant and varies in the horizontal direction only.

The flow is caused by a constant rate of recharge $R$ which is defined analogous to the infiltration or "effective rainfall" by Besbes & de Marsily (1984), namely as the flux leaving the superficial soil reservoir (i.e. the root zone). The mean pore water velocity $v$ is then related to the soil hydraulic properties and $R$ in the following way (Bresler & Dagan, 1981):

\[
\begin{align*}
    v &= \frac{K(\theta)}{\theta} = \frac{R}{\theta} \quad \text{for} \quad R < K_s \\
    v &= \frac{K_s}{\theta_s} \quad \text{for} \quad R \geq K_s
\end{align*}
\]
where \( \theta \) is the soil moisture content and \( \theta_s \), the water content at saturation. \( K(\theta) \) is the unsaturated hydraulic conductivity:

\[
K(\theta) = K_s \left( \frac{\theta - \theta_s}{\theta - \theta_f} \right)^{1/\beta}
\]  

(2)

where \( \theta_f \) is the irreducible water content and \( \beta \) is a soil coefficient related to the pore size distribution; in the following we shall assume \( \theta_f = 0 \) and \( \beta = 0.14 \).

In view of the above assumptions, an advection-dispersion transport model with sorption-desorption that is governed by first-order linear kinetics can be written for each soil column as:

\[
\frac{\partial c}{\partial t} + v \frac{\partial c}{\partial z} = D \frac{\partial^2 c}{\partial z^2} - \frac{\partial c^*}{\partial t}
\]  

(3)

and

\[
\frac{\partial c^*}{\partial t} = k_1 c - k_2 c^*
\]  

(4)

where \( c \) and \( c^* \) are the concentrations of mobile and immobile solute, respectively, and \( z \) is the depth from the soil surface. Neglecting molecular diffusion, \( D \) is the coefficient of hydrodynamic dispersion, i.e. \( D = \lambda v \), with \( \lambda \) being the dispersivity of the soil. The kinetic parameters \( k_1 \) and \( k_2 \) are usually referred to as the forward (sorption) and backward (desorption) rate coefficients, respectively. Analytical solutions of equations (3) and (4) have been discussed by several investigators (Lassey, 1988; Hubert et al., 1971).

For an instantaneous injection and detection in the flux, in a semi-infinite domain, the solution for the mass flux of solute per unit area \( s \) is defined as (Lassey, 1988):

\[
s(z,t) = \exp(-k_1 t) \Omega(z,t) + k_2 \int_0^t \Omega(z,\tau) \exp(-a_1 - a_2) a_1 I_1(a_1 a_2) d\tau
\]  

(5)

where

\[
\Omega(z,t) = \rho_A \frac{z}{\sqrt{4\pi D t^3}} \exp \left[ \frac{-(z - vt)^2}{4Dt} \right]
\]  

(6)

with \( \rho_A \) being the surface density of solute injected in the profile, i.e. mass per unit area and:

\[
a_1 = k_1 t; \quad a_2 = k_2 (t - \tau); \quad I_1(W) = I_n(2W) / \sqrt{W^n}
\]  

(7)

with \( I_n \) being a modified Bessel function of the first kind. Although the
reactions of the solute with the porous matrix may in general be very complex, the equilibrium and non-equilibrium sorption-desorption models have been shown to provide sufficiently accurate approximations for a number of reactions in porous media (Nielsen et al., 1986; Lassey, 1988). In the case when \( k_1 k_2 \rightarrow \infty \), while \( k_1/k_2 \) remains finite, the equilibrium linear sorption model is obtained. The case when \( k_1 = k_2 = 0 \) yields an expression for the mass flux of a conservative solute (Kreft & Zuber, 1978):

\[
s(z, t) = \rho_A \frac{z}{\sqrt{4\pi Dt}} \exp \left[ \frac{-(z - vt)^2}{4Dt} \right]
\]

(8)

which is identical with \( \Omega(z, t) \) in equation (6).

The source area is regarded as consisting of an ensemble of profiles that are statistically equivalent. The physical parameters that are assumed random are the rate of recharge \( R \) and the hydraulic conductivity at saturation \( K_s \). The effect of variability in other parameters, such as depth from soil surface to groundwater table \( Z \) and the sorption-desorption parameters \( k_1 \) and \( k_2 \), has been discussed elsewhere (Cvetkovic et al., 1988; Destouni & Cvetkovic, 1989).

**REGIONAL SOLUTE MOVEMENT**

Consider a hypothetical aquifer (900 m x 950 m) with constant head in the upstream and downstream boundaries (11.0–8.3 m from left to right in Fig. 1); the remaining two boundaries are assumed impermeable, implying that the velocity field is unidirectional. The aquifer is assumed to be homogeneous.

Furthermore, consider a distributed source of pollution (e.g. an agricultural field) with an area of 12.25 ha. The depth from the soil surface to the groundwater table \( Z \) is assumed to be constant within the source area and

![Fig. 1 The spatial distribution of the solute concentration in the aquifer for one realization, 3.66 years after injection.](image)
equal to 3 m. The rate of recharge $R$ is assumed to vary spatially (e.g., due to varying snowmelt conditions that are a consequence of the terrain characteristics, and spatially varying root density and soil characteristics in the root zone) and the mean value is 0.3 m year$^{-1}$.

Because natural fields exhibit a large variability in their physical properties (Jensen & Butts, 1986) the water content at saturation $K_s$ is also assumed to vary spatially in the field, the geometric mean being $3.2 \times 10^{-7}$ m s$^{-1}$. Since varying soil characteristics in the root zone may be one of the reasons for varying rate of recharge, $R$ and $K_s$ should in general be correlated to some degree. For simplicity, however, we assume that $R$ and $K_s$ are independent random variables. A uniform distribution is assumed for $R$, $R(1 - \alpha_R) \leq R \leq R(1 + \alpha_R)$, while a lognormal distribution is assumed for $K_s$ (Bresler & Dagan, 1981), with $K_s^G$ and $\sigma_k^2$ being the geometric mean of $K_s$ and the variance of $\ln(K_s)$, respectively. The probability density functions (pdfs) for the random variables are assumed stationary, i.e., the moments of the pdfs do not depend on the horizontal position.

The source of solute is hypothesized as an instantaneous injection (pulse), applied uniformly over the source area (Fig. 1). The pulse nature of the source is consistent with the fact that most of the chemicals used in agriculture are applied annually during a relatively short period of time.

The movement of water in the aquifer is modelled using the code MOC (Method of Characteristics) (Konikow & Bredehoeft, 1978), which has been adopted here for phreatic conditions; thus solute transport in the aquifer is modelled as an advection-dispersion process. The influx of solute from the unsaturated zone into the aquifer is the input for the MOC model in the form of a temporal variation of the flux-averaged concentration.

DISCUSSION OF RESULTS

The spatial distribution of the solute concentration in the aquifer is illustrated in Fig. 1 for a single realization of the field and for a conservative solute. The irregular shape of the plume is a consequence of the heterogeneity in the unsaturated zone.

Figure 2 shows the expected breakthrough of a conservative solute (full line) within three standard deviations (broken lines), integrated over two cross sections of the aquifer, 75 m and 325 m from the fringe of the source area, the latter section being the surface water boundary. The concentration in Fig. 2 represents the mass of solute per unit volume of fluid penetrating the surface water (or crossing the 75-m section) in unit time. It can be seen that at the greater distance from the source area (325 m), the uncertainty associated with the field heterogeneity decreases.

The cumulative distribution of the arrival times of peak concentration is illustrated in Fig. 3. In Fig. 4, the cumulative distribution of the peak concentration values is shown. In both cases, the normal distribution function closely approximates the values obtained from Monte Carlo experiments. The effect of the sorption-desorption is to decrease the peak concentration.

Figures 5 and 6 illustrate the effect of $\alpha_R$ and $\sigma_k$, respectively, on the
Fig. 2 The expected solute breakthrough within three standard deviations, integrated over two aquifer cross sections.

Fig. 3 The cumulative distribution for the arrival times of peak concentration compared with the normal distribution function.

uncertainty of the expected solute breakthrough. This uncertainty is quantified by estimating the mean standard deviation from the expected flux concentration $\bar{c}$ averaged over a period of 20 years after injection. The mean standard deviation $\sigma_c$ is defined as:

$$\sigma_c = \frac{1}{T} \int_0^T \frac{1}{N} \sum_{i=1}^N \left[ C_i(x,t) - \bar{C}(x,t) \right]^2 \, dt$$  \hspace{1cm} (9)$$

where $C(x,t)$ is the integrated concentration over the cross section at $x$, at time $t$ for realization $i$, and $\bar{C}(x,t)$ is the ensemble mean of $C(x,t)$, i.e. $\bar{C} = \Sigma c_i / N$; $N$ is the total number of realizations and $T = 20$ years is the considered duration of the breakthrough. Figure 5 shows that $\sigma_c$ decreases
**Fig. 4** The cumulative distribution for the peak concentration values compared with the normal distribution function.

**Fig. 5** The estimated mean standard deviation of the expected solute breakthrough as a function of the variability in $R$.

**Fig. 6** The estimated mean standard deviation of the expected solute breakthrough as a function of the variability in $K_s$. 
with the distance from the source area both for the conservative and the reactive solute. Larger recharge variability yields higher uncertainty, this effect being greater in the conservative case. In comparison, Fig. 6 shows that increasing variability in the saturated hydraulic conductivity does not influence $\sigma_r$ significantly. This is a consequence of the ratio $R/K_s (= 0.03)$ being relatively small. Specifically, an increase of $R/K_s$ increases the impact of the spatial variability in $K_s$ (Destouni & Cvetkovic, 1989). For the reactive case, however, the effect of increasing $\alpha_R$ and $\sigma_k$ on the uncertainty in the solute breakthrough, at both $x = 75$ m and $x = 325$ m, is small.

**CONCLUSIONS**

The influence of variability in the parameters that control advection in the unsaturated zone decreases with the distance from the source area. This effect is more pronounced for a conservative than for a reactive solute. Larger spatial variability in the rate of recharge increases the uncertainty in expected solute breakthrough, while the spatial variability in the saturated hydraulic conductivity has less influence on the uncertainty for the considered ratio $R/K_s = 0.03$. Both the peak concentration values and the arrival times of the peak concentration are approximately normally distributed. The effect of sorption-desorption in the unsaturated zone on regional movement of solute is to decrease the expected peak concentration and reduce the influence of field heterogeneity.

The present results on regional solute transport are primarily qualitative and provide an indication of the uncertainty that may be expected in field applications. For a more realistic simulation of the regional transport process, however, the spatial structure of the heterogeneity, and the interaction between the heterogeneity and sorption-desorption in the unsaturated and saturated zones has to be incorporated.

**REFERENCES**


