Calibration and Reliability in Groundwater Modelling: From Uncertainty to Decision Making
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Simulating environmental tracer transport in unsaturated-saturated porous media

G. A. ONNIS[1], R. ALTHAUS[2], S. KLUMP[3], H. J. W. M. HENDRICKS FRANSSEN[1], F. STAUFFER[1] & W. KINZELBACH[1]

1 *Institut of Hydromechanics and Water Resources Management, ETH-Hönggerberg, CH-8093 Zürich, Switzerland*
onnis@ihw.baug.ethz.ch

2 *Climate and Environmental Physics Division,University of Bern, CH-3012 Bern, Switzerland*

3 *Department of Water Resources and Drinking Water, EAWAG, CH-8600 Dübendorf, Switzerland*

Abstract Environmental tracers have been released to the atmosphere by human activities since the early 1950s. They provide a powerful tool for investigations of subsurface dynamics, in particular for the determination of groundwater residence times. When considering deep groundwater tables (>10 m below surface), the unsaturated zone transport of these tracers becomes a key issue for the correct evaluation of the groundwater age. Delays of more than 10 years can occur in crossing the vadose zone. In the present work we investigate the subsurface dynamics of four different environmental tracers, ^{3}H, ^{3}He, ^{85}Kr and SF_6, with particular emphasis on the tracer transport in the unsaturated zone. The method developed is applied to the Baltenswil aquifer (Switzerland).

Keywords environmental tracers; groundwater; groundwater age dating; transport; unsaturated zone

INTRODUCTION

Since the early 1950s, human activities such as nuclear power production, thermonuclear bomb testing, and industrial processes have released increasing amounts of a range of environmental tracers into the atmosphere. Many of these tracers are soluble in water, thus providing an important tool with which to track water movement in the hydrological cycle. The atmospheric concentrations of these tracers have been measured at reference sites over the last decades. By measuring the concentrations in groundwater and comparing the measured values with the known atmospheric growth curve (called the atmospheric input function) it is possible to determine the groundwater age.

When considering thick unsaturated zones, the time spent by the tracer in the unsaturated zone results in a time lag—or delay—that must be considered in the calculation of the groundwater age. For water tables at a depth of less than 10 m below the surface, this effect is almost negligible, but it gains in importance as deeper water tables are considered. It was shown (Cook & Solomon, 1995) that the time lag for a 30 m water table depth can range between 8 and 15 years. Crossing the unsaturated zone also results in a modification of the temporal concentration distributions of the tracers. The shape of the input function at the groundwater table differs significantly

from that of the atmospheric input function. Consequently, the concentration history at the bottom of the unsaturated zone must be reconstructed to give a correct input function for any meaningful transport modelling in the saturated zone.

We investigate the transport of the tracers ^{3}H, ^{3}He (decay product of ^{3}H), ^{85}Kr and SF_6 in the unsaturated zone by means of a numerical solution to the vertical advection–dispersion equation. Considering different tracers has the advantage of taking into account the different transport mechanisms in the subsurface. Tracer transport in the subsurface can occur both in the liquid and gas phases. The water bound tracer ^{3}H moves by advection following the seepage water, while the transport of the gas tracers like ^{3}He, ^{85}Kr and SF_6 is advection dominated only in the few upper metres of the unsaturated zone, while afterwards they are diffusion dominated.

We apply our theoretical studies to the Baltenswil aquifer (Switzerland). The catchment of this reference site was selected as it has been relatively well studied in the last decade. The undulating topography of the site—a sandy-gravel formation formed in the Riss ice-age—gives rise to a complicated unsaturated zone geometry that makes Baltenswil a challenging test site for the validation of our unsaturated/saturated zone transport model.

TRACER TRANSPORT IN THE UNSATURATED ZONE

Theory

We consider the advection–dispersion equation in the unsaturated zone for radioactive tracers as derived by Cook and Solomon (1995):

$$\frac{\partial(\theta^* c_g)}{\partial t} = D^* \frac{\partial^2 c_g}{\partial z^2} - q^* \frac{\partial c_g}{\partial z} - \theta^* \lambda c_g$$

where $c_g(z,t)$ is the gas concentration as a function of depth z and time t and:

$$\theta^* = \varepsilon_a + \theta\rho_l K_w + (1-\theta-\varepsilon_a)\rho_s K_W K_d$$

$$D^* = D_g + D_l \rho_l K_W$$

$$q^* = q_1 \rho_l K_W + q_g$$

are the effective parameters describing the soil and tracer physical properties: water content θ, gas filled porosity ε_a, liquid and solid phase density ρ_l and ρ_s (g m^{-3}), decay constant λ (year^{-1}), water/gas and solid/liquid partitioning coefficients K_w and K_d, effective liquid and gas diffusion coefficients D_l and D_g (m^2 year^{-1}), mean liquid and gas fluxes q_l and q_g (m year^{-1}). We assume:

1. Constant values for the physical parameters describing the unsaturated zone structure: $\varepsilon_a = 0.15$, $\theta = 0.1$, $n = 0.25$.
2. Solubility equilibrium at the surface between atmospheric and soil water and gas tracer concentration. Water/air partitioning coefficient: $K_w = 0.01$ for all gas tracers.
3. ^{3}H is bound to the water molecule: $K_w = 1$ for tritium.
4. Negligible sorption. Solid/water partitioning coefficient: $K_d = 0$.

5. Advection of the gas phase is important only in the first upper metres of the unsaturated zone: $q_g = 0$. Only for tritium ^3H, is advection of the liquid phase the driving transport phenomenon: $q_l = 0.83$ m year^{-1}.

With these assumptions, the transport phenomena are controlled by the following parameters:

^3He, ^{85}Kr, SF_6	^3H
$\theta^* \approx \varepsilon_a$	$\theta^* \approx \theta\rho_l K_w$
$D^* \approx D_g = D_g^0 \tau_g \varepsilon_a$	$D^* = D_l^0 \rho_l K_w$
$q^* = 0$	$q^* = q_1 \rho_l K_W$

where D^0_g and D^0_l are the diffusion coefficients in free air and water, respectively, and τ_g is the gas phase tortuosity (constant $\tau_g = 0.25$ is assumed).

The one-dimensional (1-D) numerical solution to the advection–dispersion equation is obtained by a forward-time and central-space finite difference scheme. The conjugate gradient solver is used to get the solution of the implicit system. The unsaturated zone is modelled as an *N*-cell homogeneous 1-D column with the upper boundary at constant concentration (equal to the atmospheric one) and a lower boundary at depth *z* (*z* being spatially variable) with a constant outgoing flux (equal to the average groundwater recharge rate, $r = 633$ mm year^{-1}). For the actual recharge rate value, diffusion at the lower boundary is not considered (it may, however, become important in low recharge conditions).

Results

Simulations were performed for ^{85}Kr ($D^0_g = 440$ m^2 year^{-1}, half-life $\tau_{1/2} = 10.76$ years) and SF_6 ($D^0_g = 206$ m^2 year^{-1}), respectively, making use of the Freiburg am Breisgau (Germany) input function for ^{85}Kr and the global input function for SF_6 (data from IAEA, International Atomic Energy Agency).

The concentration profiles at different depths follow the atmospheric input function (soil surface profile), but become smoother and lose detail with increasing depth. A time delay is also apparent in the shift to the right of the concentration profiles as depth increases (Fig. 1).

Sensitivity analysis shows no apparent dependence of the concentration profiles on either water content, θ, or on gas filled porosity, ε_a, (constrained by $n = \theta + \varepsilon_a$). A remarkable sensitivity is found with respect to gas phase tortuosity, τ_g, raising tortuosity (and thus the effective diffusion coefficient) results in higher concentration values at depth, i.e. in faster tracer dynamics in the subsurface. As expected, sensitivity to the depth of the lower boundary is also important: the deeper the boundary, the slower the tracer dynamics. This consideration is of major importance when considering aquifers with overlying undulating topography, i.e. variable unsaturated zone thickness. Smaller sensitivity is found with respect to the recharge rate which enters the model as the constant outgoing flux at the lower boundary.

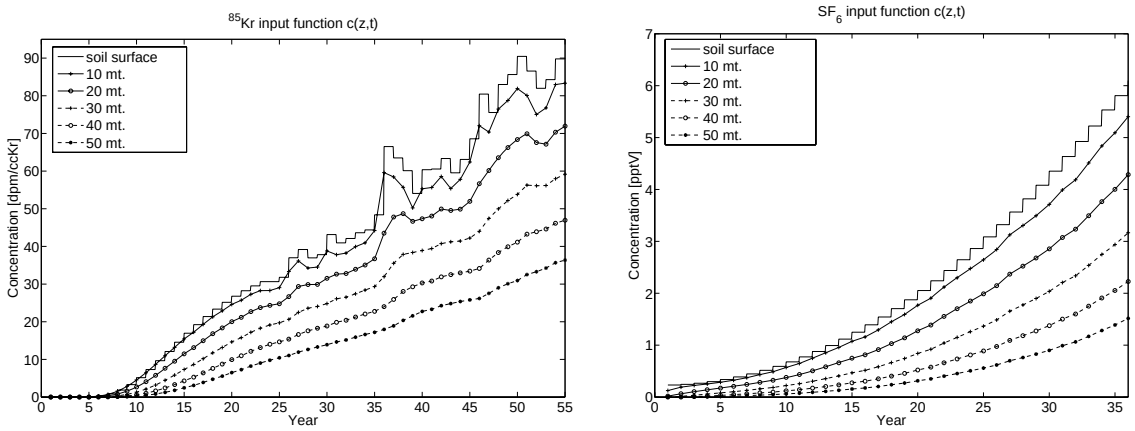

Fig. 1 ^{85}Kr (years 1950–2004) and SF_6 (years 1969–2004) time series for different depths of the unsaturated zone.

Simulations for ^{3}H (D^0_l = 0.0495 m^2 year^{-1}, half-life $\tau_{1/2}$ = 12.43 years) and ^{3}He (D^0_g = 2179 m^2 year^{-1}) as its stable daughter product were performed, making use of the Konstanz (Germany) ^{3}H input function (Fig. 2).

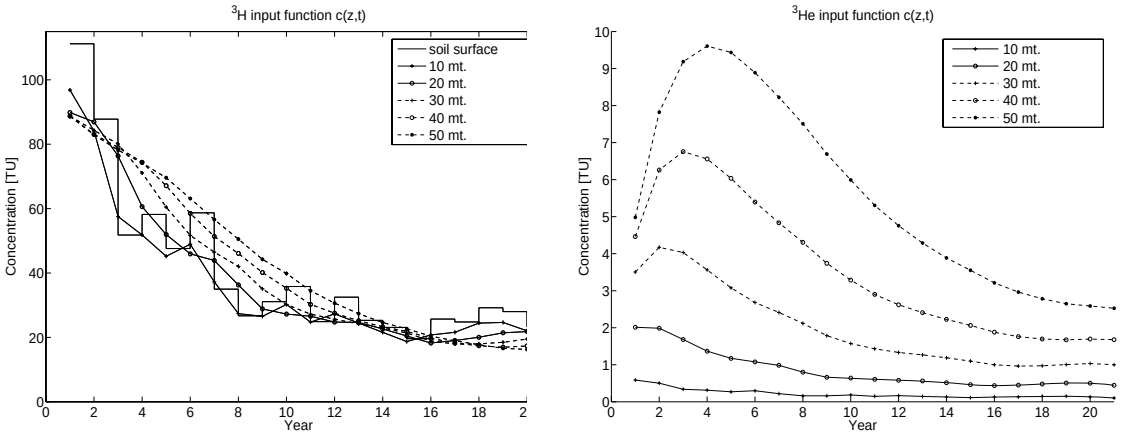

Fig. 2 ^{3}H and ^{3}He (years 1978–2004) time series for different depths of the unsaturated zone.

Longitudinal dispersivity is set equal to α_L = 5 m. The upper boundary condition for ^{3}He transport simulation is 0 TU (atmospheric concentration). The advective character of the ^{3}H transport equation results in a sharp and less smoothed shift to the right of the concentration profiles (delay effect). This explains the faster ^{3}H transport dynamics compared to other environmental tracers, which is controlled by the advection term parameter. Some sensitivity of ^{3}H and ^{3}He concentration profiles to water content θ and gas filled porosity ε_a is found, as well as to mean water flux q_l: higher water content (and thus lower gas filled porosity) results in slower dynamics of both ^{3}H and ^{3}He. Sensitivity to gas phase tortuosity τ_g is again found to be the most relevant. Some sensitivity to recharge rate as well as to the depth z of the lower boundary is also found for both tracers.

CASE STUDY: UNSATURATED AND SATURATED ZONE TRANSPORT IN THE BALTENSWIL AQUIFER

The Baltenswil (Kanton Zürich, Switzerland) aquifer is part of the Aathal aquifer, a sandy-gravel formation mainly formed in the Riss ice-age. The highly conductive layer is covered by moraine deposits and is underlain by poorly permeable silty and loamy lake deposits. Parts of the catchment are situated completely in the more or less permeable moraine layer, underlain by the impermeable rock formation. The thickness of the unsaturated zone varies between 0 and 50 m.

The domain (3×3 km^2) is divided into two aquifer zones for modelling purposes (Fig. 3): a large saturated domain, characterized by a stochastic transmissivity field ($T = 10^{-3}$ m^2 s^{-1} on average) and a second smaller inflow area (indicated with arrows, northeast of the larger area) is modelled as a very thin aquifer with low constant transmissivity ($T = 10^{-4}$ m^2 s^{-1}). The second area is defined in order to account for the inputs from the unsaturated zone lying above it, which consequently become the boundary fluxes of environmental tracer to the aquifer proper.

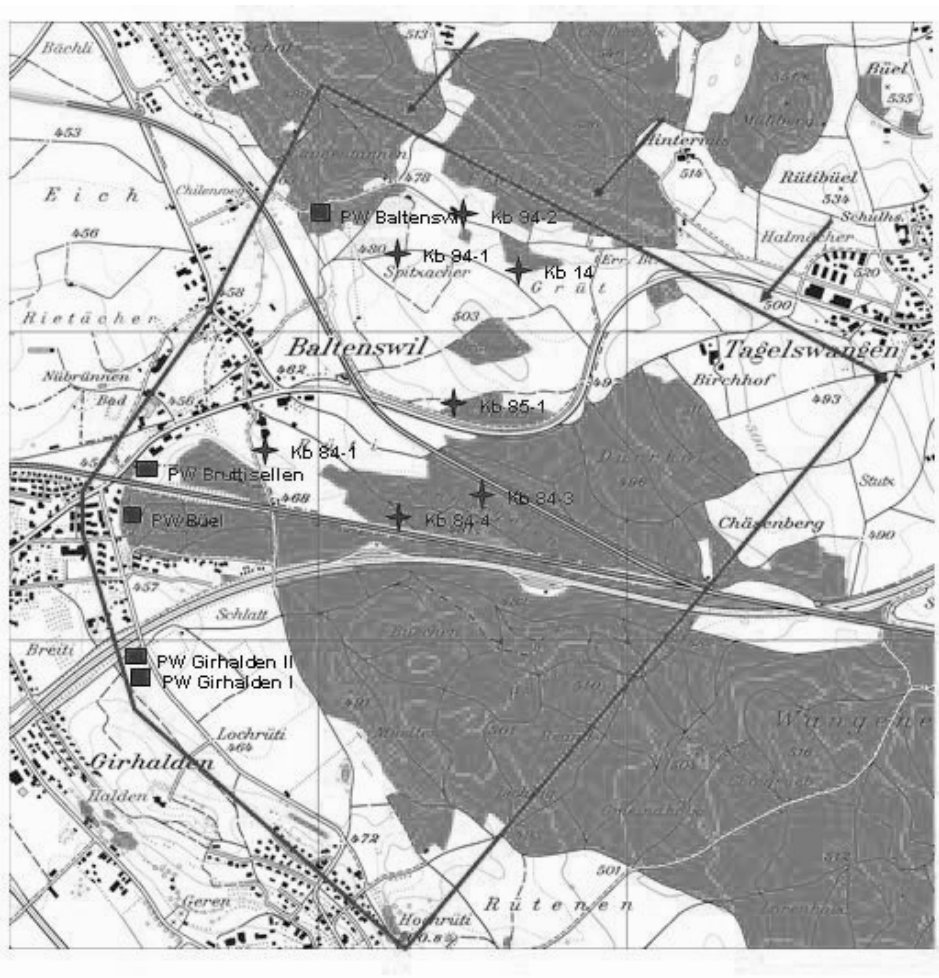

Fig. 3 Boreholes and measuring points for Baltenswil aquifer.

Saturated zone stochastic flow model

The flow model for the saturated zone is based on a set of 100 multiple equally-likely realizations of the log-transmissivity field generated with GCOSIM3D (Gómez-Hernández & Journel, 1993) conditional to transmissivity (*T*) measurements. In addition, these log*T* fields are also conditioned to steady-state hydraulic head measurements by the Sequential Self-Calibrated Method (Gomez-Hernández *et al.*, 1997a,b), implemented in the code INVERTO (Hendricks Franssen, 2001).

Transport model

For each cell of the domain (60 × 60 cells) we solve the transport equation in the unsaturated zone for a 1-D column with the lower boundary set at a depth equal to the unsaturated zone thickness, previously calculated by means of the difference between the altitude a.m.s.l. (from a Digital Elevation Model) and the calculated ensemble average hydraulic head (Fig. 4, bottom right). According to the undulating topography of the site and given the atmospheric input function, we reconstruct the concentration history at the bottom of the unsaturated zone. This was then used as concentration input flux for transport modelling in the saturated zone (performed with MT3DMS (Zheng, 1990)) for the years 1991–2004. For transport simulations in the saturated zone, we use a flow model based on one transmissivity realization drawn from the ensemble.

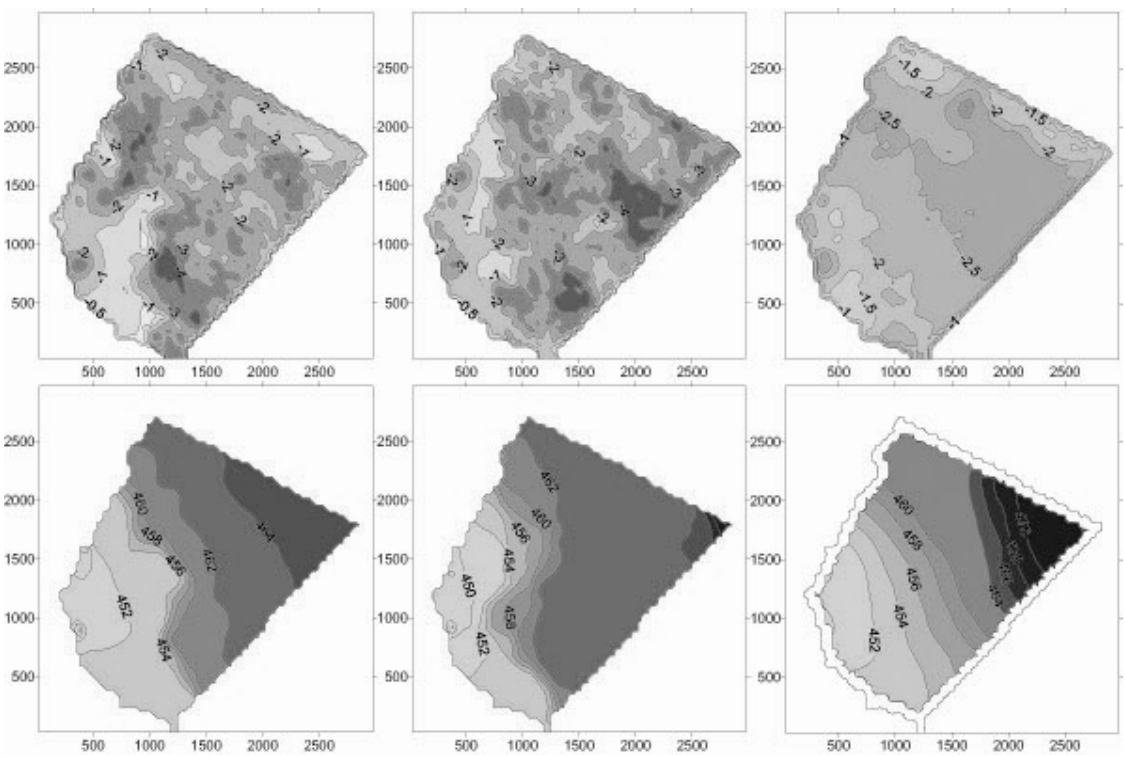

Fig. 4 Two stochastic transmissivity realizations (top-left and top-centre) and related hydraulic head fields (bottom-left and bottom-centre) from the 100 realizations set. On the right-hand side, average transmissivity (top-right) and average head (bottom-right) fields over 100 realizations.

RESULTS

Breakthrough curves at the Baltenswil pumping station show on the whole a fair simultaneous match between simulated and measured concentrations (Fig. 5).

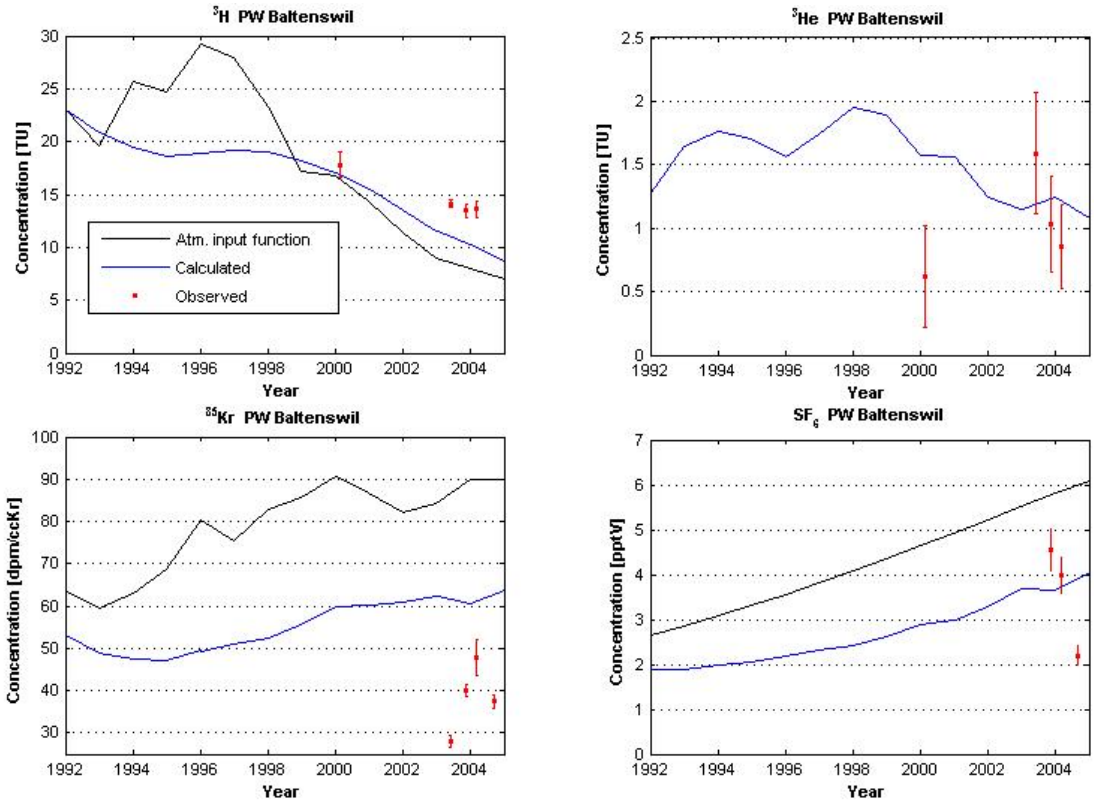

Fig. 5 Breakthrough curves at the Baltenswil pumping stations for the four different tracers.

Sensitivity analysis on the final breakthrough curves at the measuring locations shows that the mismatch between calculated and simulated values can be reduced with proper parameter calibration, the most important being the atmospheric input function. Due to its very local character (^{3}H especially is still often used in industrial activities), the input function is the most uncertain input parameter of the model. Also the unsaturated zone transport parameters, such as gas tortuosity (effective diffusion coefficient), recharge rate and water content (for the ^{3}H and ^{3}He simulations) play an important role on the final breakthrough curves.

Saturated zone transport has also been performed making use of different flow models based on different transmissivity realizations from the stochastic ensemble (Fig. 4), transmissivity being usually the most important parameter affecting the model predictions' uncertainty. No sensitivity to the transmissivity realization has been found because of the short residence times in the saturated zone. In shallow aquifers the most important role for the transport model calibration is then played by the unsaturated zone dynamics.

CONCLUSIONS

The tracer dynamics in the unsaturated and saturated zone for the radioactive gas tracers ^{85}Kr, SF_6, ^{3}He (stable), and water bound ^{3}H have been investigated. The concentration profiles in the subsurface differ significantly from the atmospheric input function, even in a 1-D and homogenous medium. In the real world, heterogeneity and neglected 2-D/3-D effects will play an important role in the modification of the input function at the groundwater table.

Different transport dynamics in the unsaturated zone are recognized as the most important factor in determining the tracer concentrations at the groundwater table. Where groundwater is recharged through a thick unsaturated zone, and especially in areas where topography is undulating, it is necessary to reconstruct the tracer input at the interface between unsaturated and saturated zone.

Sensitivity analysis showed the major importance of parameters such as the input function and the effective diffusion coefficients. Transport model results are most sensitive with respect to eventually heterogeneous parameters describing the unsaturated zone. The next step after this study would be the stochastic modelling of these parameters in order to set the modelling problem in an uncertainty assessment context.

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