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Modeling subsurface transport in extensive glaciofluvial and littoral sediments to remediate a municipal drinking water aquifer

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Abstract

Few studies have been carried out that cover the entire transport process of pesticides, from application at the soil surface, through subsurface transport, to contamination of drinking water in esker aquifers. In formerly glaciated areas, such as Scandinavia, many of the most important groundwater resources are situated in glaciofluvial eskers. The purpose of the present study was to model and identify significant processes that govern subsurface transport of pesticides in extensive glaciofluvial and littoral sediments. To simulate the transport processes, we coupled a vadose zone model at the point scale to a regional groundwater flow model. The model was applied to a municipal drinking-water aquifer, contaminated with the pesticide-metabolite BAM (2,6-dichlorobenzoamide). A sensitivity analysis revealed that hydraulic conductivity and infiltration rate accounted for almost half of the model uncertainty. For a ten-meter-deep vadose zone of coarse texture, macropore flow was found to be of minor importance for contaminant transport. The calibrated model was applied to optimize the location of extraction wells for remediation, which were used to verify the predictive modeling. Running a worst-case scenario, the model showed that the establishment of two remediation wells would clean the aquifer in four years, compared to nine years without them. Further development of the model would require additional field measurements to assess the importance of macropore flow in deep, sandy aquifers. We also suggest that future research should focus on characterization of the variability of hydraulic conductivity and its effect on contaminant transport in eskers.

1 Introduction

An increasing number of groundwater aquifers and drinking-water supplies across the world are being contaminated by chemicals from anthropogenic sources (EEA, 1999; USEPA, 1999; Erickson, 2002). Pesticides and their degradation products are some of the most common contaminants reported in European countries (Cerejeira et al., 2003;

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Lapworth et al., 2006; Papastergiou and Papadopoulou-Mourkidou, 2001; Schiavon et al., 1995) and in the US (Barbash et al., 2001; Kolpin et al., 1998). In Sweden, there are about one 900 municipal drinking-water plants that draw their supplies from natural or artificial groundwater. As part of a national monitoring program, 28% of the groundwater supplies investigated between 2005 and 2008 were contaminated with pesticides (Törnquist et al., 2007; CKB, 2009). BAM (2,6-dichlorobenzoamide) was found to be the most common contaminant, reported in 60% of the contaminated groundwater supplies. Similar levels of contamination have been found in Italy, the Netherlands and Germany (Versteegh and te Biesebeek, 2003; Wolter et al., 2001; Morvan et al., 2006). In Denmark, for example, BAM has been detected in 21% of all groundwater wells investigated (Clausen et al., 2004) and approximately one hundred groundwater supplies were recently closed due to BAM contamination (Sørensen et al., 2009).

BAM is a hydrophilic metabolite of dichlobenil (2,6-dichlorobenzonitrile) and is highly persistent in soil (Cox, 1997). Dichlobenil has largely been used as a pesticide to control various types of weeds. The use of the product Totex, which contains the active substances dichlobenil and atrazine, has been banned in Sweden since 1991, and in many other countries (KEMI, 2008). The toxicity of dichlobenil, which is classified as slightly to moderately toxic, has been investigated by several authors (Linders et al., 1994; Cox, 1997). However, relatively little is known about the toxicity and health effects of its degradation product BAM, although the available data indicate that its potential carcinogenicity does not exceed that of dichlobenil (USEPA, 1998). The potential effects of long-term exposure to low concentrations of either compound have not been thoroughly investigated.

The large number of contaminated groundwater-supplies highlights the need for tools to evaluate the subsurface processes controlling contaminant transport. The hydrogeology of groundwater aquifers is in general complex, and numerical modeling is often required. Depending on the extent to which a model can be validated using field data, it can be used to predict the behavior of contaminants over reasonably long time-scales

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and under various conditions (Hassan, 2004). Numerical models can also be used to predict the potential of remediation strategies at a specific site and aid decision making (Prommer et al., 2000; Zhou et al., 2004).

Most numerical models are optimized to solve contaminant transport problems in either the vadose or groundwater zones, however, models have been proposed which couple transport in both zones (Panday and Huyakorn, 2008; Twarakavi et al., 2008; Trefry and Muffels, 2007; Weill et al., 2009). For pesticide transport, one-dimensional vadose zone models and three-dimensional groundwater models have been coupled and applied at local (Stenemo et al., 2005; Jorgensen et al., 2004) and regional scales (Herbst et al., 2005; Christiansen et al., 2004). These models were developed for sites with fine-textured soils and shallow groundwater, where macropore flow is a significant transport process. In esker aquifers, which are generally extensive in depth and range, few regional-scale studies have considered the whole transport process, from application of pesticide at the soil surface, through transport in the vadose zone, to contamination of drinking water (Köhne et al., 2009). The lack of models for eskers at this scale is problematic since many of the most important groundwater resources in recently glaciated regions of the world are situated in such aquifers.

The objectives of this study were: (1) to model and identify significant processes that govern subsurface transport in extensive glaciofluvial and littoral sediments, focusing on a BAM-contaminated esker aquifer, which supplies a municipal drinking-water utility; and (2) to use knowledge of these processes to design and test a remediation strategy and verify the results from the model.

2 Materials and methods

2.1 Modeling approach

Simulation of pesticide movement was performed in two steps. First, transport in the vadose zone was modeled at the point scale with Hydrus-1D (Šimůnek et al., 1998).

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Hydrus-1D is a one-dimensional finite-element model which accounts for a variety of processes, including soil-water flow, solute transport, heat transport, sorption, degradation, volatilization, crop uptake and surface runoff. The model also considers mobile and immobile fractions of water flow and uses the dual permeability approach proposed by Gerke and van Genuchten (1993) to account for macropore flow. To simulate transport in groundwater at the regional scale, the calculated pesticide concentration from the vadose zone model was used as the input to the 3-D finite-difference model Modflow (Harbaugh et al., 2000) coupled with MT3DMS (Zheng and Wang, 1999). Modflow calculates flow and MT3DMS, which is designed to interface with the Modflow code, performs transport calculations (Fig. 1).

Hydrus-1D numerically solves the Richards equations for variably saturated water-flow and advection-dispersion type equations for solute transport. The water-flow equation solved by Hydrus-1D (for flow in the vertical direction) is:

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

where h is the water pressure head; θ is the volumetric water content; t is time; z is the spatial coordinate (positive upward); q_s is the sink term; and K is the unsaturated hydraulic conductivity. The van Genuchten-Mualem model was used to calculate soil hydraulic properties (van Genuchten, 1980). To consider the effect on migration of pesticides by seasonal variation in climatic parameters and extreme rainfall events, Eq. (1) was solved with daily input data. The solute transport equation with first-order reaction rate solved by Hydrus-1D is:

$$\frac{\partial(\rho s)}{\partial t} + \frac{\partial(\theta c)}{\partial t} = \frac{\partial}{\partial z} \left(\theta D \frac{\partial c}{\partial z} - qc \right) - \mu(\theta c + \rho s), \quad (2)$$

where s and c are the solute concentrations in the solid and liquid phases, respectively; q is the volumetric flux density; ρ is the soil bulk density; θ is the volumetric water content; t is time; z is the spatial coordinate (positive upward); D is the dispersion coefficient for the liquid phase; and μ is a first-order rate constant.

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The transient flow equation solved by Modflow is:

$$S_S \frac{\partial h}{\partial t} = \frac{\partial}{\partial x_i} \left(K_{ij} \frac{\partial h}{\partial x_j} \right) + q_s, \quad (3)$$

where h is the hydraulic head; K_{ij} is the tensor of hydraulic conductivity; x_i is the distance along the respective Cartesian coordinate axis; q_s is the sink/source term; S_S is the specific storage; and t is time. The advection-dispersion equation solved by MT3DMS for one species is:

$$\frac{\partial(\theta C)}{\partial t} + \rho_b \frac{\partial S}{\partial t} = \frac{\partial}{\partial x_i} \left(\theta D_{ij} \frac{\partial C}{\partial x_j} \right) - \frac{\partial}{\partial x_i} (\theta v_i C) + q_s C_s + \sum R_n, \quad (4)$$

where C is the solute concentration; θ is the porosity; t is time; x_i is the distance along the respective Cartesian coordinate axis; D_{ij} is the tensor of hydrodynamic dispersion; v_i is the seepage velocity; q_s is the sink/source term related to the aquifer; C_s is the concentration of the sink/source flux; ρ_b is the bulk density; S is the sorbed concentration; and R_n is the chemical reaction term.

To evaluate the robustness of the model's predictions, a sensitivity analysis was undertaken with respect to the physical and chemical parameters included in the model. Considered parameters were the van Genuchten-Mualem parameters, infiltration rate, thickness of the vadose zone, dispersivity, sorption and degradation of the contaminants, and hydraulic conductivity in the aquifer. The parameters in the calibrated models were perturbed with changes of ± 2 to 10%, corresponding to the observed variability (Bergvall et al., 2007). The equation used to compare the perturbed models was:

$$\text{RMS} = \sqrt{\frac{\left(c_{\text{cal}_1} - c_{\text{per}_1} \right)^2 + \left(c_{\text{cal}_2} - c_{\text{per}_2} \right)^2 + \dots + \left(c_{\text{cal}_n} - c_{\text{per}_n} \right)^2}{N}}, \quad (5)$$

where RMS is the root-mean-square deviation in the prediction; c_{cal_i} is the solute concentration at the target in the calibrated model at time step i ; c_{per_i} is the solute

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concentration at the target in the perturbed model at time step i ; and N is the number of time steps.

2.2 Field site description

The city of Umeå is located in northern Sweden (lat. 63.83, long. 20.26) and has a population of 115 000. Groundwater is an important source of municipal drinking-water for this city. Unfortunately, the concentration of BAM in the groundwater exceeds the European drinking-water standard for pesticides, $0.1 \mu\text{g l}^{-1}$ (EU, 2006). Groundwater measurements began in 1998 and, since then, BAM has been at concentrations exceeding the standard, by up to 22 times, in two extraction wells. A former forest-plant nursery situated in Piparböle, about 1.5 km up-gradient from the water-extraction wells (Fig. 2), is the source of the contaminant (Bergvall et al., 2007).

The nursery, which was active between 1956 and 2002, cultivated mainly spruce and pine seedlings. Several kinds of pesticides were used at the nursery, but only dichlobenil and its metabolite BAM were considered in this study since there is great concern over BAM contamination owing to its high mobility. Records from the nursery suggest that Totex, with the active substances dichlobenil and atrazine, was initially applied during the transition from growing bare-root seedlings on open land to their production in small containers on open land. The transition between production methods began during the 1970s and records suggest that the first application of Totex was in 1976. Totex was applied in granules, mainly along the plant beds and at the short sides where the irrigation system was located. According to historical aerial photographs, this area was estimated to cover 4% of the total $120\,000 \text{ m}^2$ open-land area. The recommended application rate of the active substance was 0.16 to 1.7 g m^{-2} .

The former nursery is situated on well-sorted, unstructured, littoral sand and lenses of silt. It is in contact with a north-south-oriented esker, which consists of glaciofluvial sand and gravel containing large quantities of groundwater. The general direction of groundwater movement is southwards, although local-scale variation in this movement does occur (Fig. 2). The groundwater table at the nursery is at a depth of approximately

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10 m, but at the extraction wells its depth is 23 to 27 m, depending on the pumping rate.

The esker is partly covered by thin layers of silt and clay, especially along the sides. Layers of silt and clay also underlie the northern and southern lakes (Fig. 2), which explains why the groundwater head at the south end of the southern lake is found 27 to 28 m below the surface water head. However, the esker is also partly located beneath the silty and clayey beds of the lakes.

2.3 Data sampling – unsaturated zone

In 2003, the soil surface at the former nursery was screened for dichlobenil and BAM. At one identified hotspot (Fig. 2), soil samples were collected at 40 cm intervals, down to the groundwater table, in November 2003. Details of the sampling have been reported previously (Bergvall et al., 2007) and the results are summarized in Table 1. In February 2009, a total of thirty soil samples were collected. They originated from five locations and were taken every 50 cm, down to a depth of 5–7 m (Fig. 2). The samples were frozen (at -18°C) until analysis. Soil samples were analyzed with liquid chromatography-tandem mass spectrometry (LC-MS/MS, Micromass Quattro Ultima coupled to an Agilent 1100 HPLC). The concentration of total organic carbon (TOC) was calculated as the percentage weight lost on ignition, in accordance with the SS-EN 12879 method (SIS, 2000).

2.4 Data sampling – saturated zone

The hydraulic groundwater head was monitored in 82 observation wells (Fig. 2) in October 2005 and August 2007. The monitoring took one week in each case. Fifty five of the observation wells were also used for sampling (Fig. 2). The groundwater collected was subsequently analyzed by LC-MS/MS. The maximum number of samples analyzed from a single observation well at the nursery was 46 (Fig. 4), whereas in peripheral wells only one sample was analyzed (Table 2). In total, 101 samples have been analyzed from the two extraction wells since 1998 (Fig. 5). One problem with

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the observations shown in Figs. 4 and 5 is that many different people from the local authority collected these samples. This means there is uncertainty as to whether the wells were purged adequately prior to sampling. In addition, the extraction wells have not operated on a regular basis, which may have affected the sampling mixture of the collected water. Since 2007, the wells have been running continuously for remediation purposes and the sampling frequency has been intensified.

The stream that drains from the southern lake crosses the aquifer above the groundwater surface. We estimated the drainage from the stream to the aquifer by measuring the stream discharge above and below the aquifer-crossing, using two water capacitance probes (TruTrack loggers WT-HR 64K, New Zealand). The water-level probes were calibrated on four occasions in 2005, using an automated salt-dilution method (Q-trace, Logotronics, Austria). On the first occasion, salt injections were also used at four other downstream locations (Fig. 2). The mean leakage was estimated to be 5% of the mean discharge rate of 160 l s^{-1} (SMHI, 2009).

Two pumping tests were conducted in the study area. One extraction well, situated 1100 m north of the model boundary (Fig. 2), was pumped for 127 days at 112 l s^{-1} . The two BAM-contaminated extraction wells, located 60 m apart, were pumped for 10 days at 83 l s^{-1} . Both tests were performed in unconfined parts of the esker aquifer. Results from the 127-day test indicated the hydraulic conductivity was between $1.0 \times 10^{-3} \text{ m s}^{-1}$ and $3.3 \times 10^{-3} \text{ m s}^{-1}$. The specific yield was between 7.4 and 9.1%. The 10-day test indicated a hydraulic conductivity of $1.0 \times 10^{-3} \text{ m s}^{-1}$ and a specific yield between 7.0 and 20.1%.

Hydraulic conductivity was also determined using slug tests in the observation wells (Fig. 2) (Hvorslev, 1951). The tests were carried out in a wide range of sediments including esker material, sand, silt and till (Table 1).

At the nursery, a local-scale, salt-injection study was conducted between August and October 2003. Salt water was injected into the groundwater in one well and transported by natural gradient flow to another well 2 m away. At a rate of 0.38 to 0.98 l min^{-1} 1200 l of salt water, with an electrical conductivity of 147 mS m^{-1} , was injected. The electrical

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conductivity was logged every 30 min at three levels in both wells (at depths of 0.1 m, 1.4 m and 1.9 m below the groundwater table). Assuming a Gaussian distribution, the longitudinal dispersion ranged between 0.2 m and 1.5 m, at the 2 m scale.

2.5 Application of modeling approach

2.5.1 Vadose zone model

The decoupling of the vadose and groundwater zones was found to be appropriate since there was little variation in the groundwater table depth (standard deviation, $SD = 0.06$, Table 1). A 1-D model was motivated since there was no evidence of tilting layers under the soil surface, which can cause lateral flow. The slope of the surface was small (0.8%) and the point-scale model, which was assumed to be representative of the total open land area, covered a depth of 10.2 m, from the soil surface to the groundwater table. It was assumed that there was no surface runoff because of the flatness and texture of the soil surface, which was classified as medium to coarse sand. The details of the vadose zone model (Larsson and Grip, 2004) were as follows. The conceptual model included variable texture and water content, at different depths relative to the groundwater table. A finite-element grid, with a total of 510 uniformly spaced cells in the vertical direction, was designed for modeling flow and transport. Ten soils with various properties were defined to describe the vadose zone, ranging from fine sandy silt to coarse sandy medium sand (Table 3).

Degradation of dichlobenil and BAM was described by first order kinetics, conditioned on the total mass balance (Table 3). The half-lives given in Table 3, agree with those of a Danish study (Clausen et al., 2007), which also demonstrated that various redox conditions do not affect degradation of dichlobenil. As in other studies, they found strong evidence that the degradation to BAM is a microbially catalyzed process. Microorganisms are most likely to be found in the upper layers of the soil and the shortest half-lives were found close to the soil surface. Clausen et al. (2007) could not correlate degradation of dichlobenil to BAM with sorption, water chemistry or composition of soils

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or sediments. As moderate concentrations of dichlobenil were found at depths greater than 0.9 m, we assumed that the linear Freundlich sorption isotherm was applicable. The Langmuir sorption isotherm was applied at the soil surface and at depths less than 0.9 m. The sorption coefficient, K_d , for dichlobenil and BAM (Table 3) varied depending on the analyzed organic carbon content and literature data (Hornsby et al., 1996). Dispersivity was initially assumed to be 10% of the soil-profile depth, but was calibrated to 5 m, except for the top meter which was calibrated to 1 m. The dimensionless Henry coefficient was set to 3.5×10^{-4} (Linders et al., 1994).

Since the pesticide was very expensive, we assumed that only 0.17 g m^{-2} was applied, twice a year between 1976 and 1983, and once a year between 1984 and 1991. This means that 19 kg of the active substance, dichlobenil, was applied to 4% of the total open land area between 1976 and 1991; this is considered to be a conservative estimate (Bergvall et al., 2007). However, since the soil has been repeatedly mixed over the years, we assumed the same load of pesticide over the entire open land area (i.e. 4% of 0.17 g m^{-2}).

Daily climate data, including precipitation, temperature, wind velocity, relative humidity and global radiation, were collected from a weather station located 10 km southeast of the nursery (SMHI, 2009). A temperature-index approach was used to model snow cover and snow melt (Anderson, 1976), and potential evaporation was calculated according to the Penman equation (Penman, 1948). To generate input data for the predictive modeling, the climate data from the preceding eight years were used; for example, climate data from 2003 were used for 2011.

The van Genuchten-Mualem parameters were calibrated against water content measured at 51 depths on 13 occasions. The mean error of the total water balance was 1.3%. The root-mean of the squared differences between the calculated and observed water contents was normalized by the observed range of water contents, giving a calibration error of 10%. The solute component of the model was calibrated to achieve the best match between the calculated and observed concentrations of dichlobenil and BAM, in the solid and solute phases. The root-mean-squares of the residuals of the

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adsorbed and liquid phases were normalized by the observed concentration range giving calibration errors of 38% and 39%, respectively.

2.5.2 Groundwater model

A 3-D, block-centered, finite-difference grid was designed to model flow and transport in the saturated zone. In the horizontal direction, a total of 73 columns and 153 rows (uniformly spaced at 20 m) were used for flow simulation, covering an area of 1.45 km by 3.05 km. The aquifer was divided into two layers, with the upper layer approximately 2 m below the groundwater table. Initially, the groundwater table was calculated in a one-layer model with the same boundary conditions.

Boundaries were identified as general heads, specified heads, no-flows and rivers (Fig. 3). The conductances of the general head boundaries were calibrated to give inflows corresponding to the net infiltration rate in the northeast and to flow estimates from the two pumping tests (conducted north of the northwest boundary and at the extraction wells); the inflow rate was calculated as 83 l s^{-1} in the northwest and 0.4 l s^{-1} in the northeast. The conductance of the southern stream, where the stream flows above the esker, was calibrated to match stream leakage. The conductance of the east river boundary was set to permit small inflows and outflows to the aquifer.

In accordance with records from the municipality, the extraction rate of the two contaminated wells varied between 0 and 61 l s^{-1} . Four different recharge rates were defined for the model: most areas had a constant net infiltration rate of $8.2 \times 10^{-4} \text{ m d}^{-1}$. The constant rate was justified by the 10 m thick vadose zone, which smoothes out variations in the net infiltration rate (Kuntz and Grathwohl, 2009) and changes in the groundwater level (Table 1). The lakes were assumed to give no recharge to groundwater (Fig. 2). Areas in the vicinity of the lakes, mainly west of the southern lake, consist of two aquifers, where the upper one (Fig. 2, consisting of silt and clay) has a leakage to the lower one (consisting mainly of sand). The upper aquifer has primarily an outflow to the lakes and is not connected to the contaminated area. A leakage from the upper aquifer was, therefore, modeled as recharge to the lower aquifer, assuming

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a net infiltration rate of $8.2 \times 10^{-5} \text{ m d}^{-1}$. The erosion-cuts along the streams (Fig. 2) consist mainly of sand and the recharge rate was assumed to be $4.1 \times 10^{-5} \text{ m d}^{-1}$.

Initially, hydraulic conductivity was quantified in accordance with the results from the pumping and slug tests. The extent of the zones of hydraulic conductivity were derived from borehole data, based on 91 borings (Wikner and Öhlén, 1994), and ground-penetrating radar measurements (RAMAC/GPR, 100 MHz antenna, Malå Geoscience, Sweden), using prior knowledge of how glaciofluvial sediments have been geologically deposited (Fig. 3).

The bedrock surface defines the bottom of the model (Fig. 3), except in small areas where silt or clay is deposited beneath more coarse-grained textures. The calibration of hydraulic conductivity zones in the two layers was conducted in two steps to match the calculated and observed heads at the observation wells. First, assuming no pumping at the extraction wells, the model was calibrated to the observations from 2005. Secondly, with the total pumping rate set to 44 l s^{-1} in the extraction wells, the model was calibrated to the heads observed in 2007. The calibrated flow model had a mean head error of 0.11 m. The calibration error, defined as the root-mean of the squared head differences normalized by the observed head range, was 1.6%, which is less than the calibration target of 5%.

A transient verification of the model was conducted using data from the 10-day pumping test performed in the extraction wells. The verification was also used to confirm that the mean of the specific yield evaluated from the pumping test was accurate. Since the aquifer is unconfined at the location where the pumping test was conducted, the mean specific yield (0.13) could be used as the effective porosity in the transport model.

According to Clausen et al. (2007), only insignificant degradation of dichlobenil and BAM has been observed in groundwater. Since dichlobenil has not been detected in our aquifer, only transport of BAM was modeled. The solute concentration data from the Hydrus-1D model was evenly distributed in the grid cells corresponding to the open land area of the former nursery. Time was divided into half-year intervals, which provided sufficient resolution to respond to changes in the input of the contaminant

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and mass conservation. The simulation started on 1 January 1976 and the calibration model was run for 33.5 yr until 1 July 2009. The prediction model was run for 45 yr until 31 December 2020.

Calibration of dispersivity in the three dimensions was conducted manually, to correlate the calibrated with the observed concentrations of BAM in the observation and extraction wells (Table 2). By considering the integral of the observed concentrations, the aim was to fit the concentrations in observation well #7705 (Fig. 2) and the extraction wells with the least-squares method. The root-mean-squares of the residuals of the observed and modeled BAM concentrations at the observation well and at the extraction wells, were normalized by the observed range of concentrations giving calibration errors of 15% and 14%, respectively.

Calibration of the transport model gave a longitudinal dispersivity of 5 m, a transverse horizontal dispersivity of 0.1 m and a vertical dispersivity of 0.04 m. This is consistent with the results from the local-scale, salt-injection study (α_L between 0.2 and 1.5 m), according to the empirical relationship between longitudinal dispersivity (α_L) and field scale (L), suggested by Schulze-Makuch (2005):

$$\alpha_L = 0.085 L^{0.81}, \quad (6)$$

Equation (6) gives a longitudinal dispersivity of 0.96 m at the grid-cell scale and 0.15 m at the scale of the salt-injection study. The relatively low transverse horizontal dispersivity and vertical dispersivity values are realistic since advection dominates transport and sorption is limited.

The ratio between the grid-cell size and the longitudinal dispersivity gives a grid Peclet number of 4, which is relatively high. In the horizontal transverse direction the grid Peclet number is 200. To avoid numeric model dispersion with the finite difference method, the third-order total-variation-diminishing (TVD) method was chosen. The TVD method is suitable for advection-dominated transport problems (Zheng and Wang, 1999).

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2.6 Remediation strategy and model validation

Since May 2007, water from the two contaminated extraction wells has been pumped into a nearby river. Because these wells are situated 1.5 km from the contamination source, this has not been a cost-effective way to remediate the area. The contaminated vadose zone amounts to approximately 1.2 million m³ of sand. Therefore, application of a treatment system to the vadose zone is unfeasible.

The calibrated model was used to design a more cost-effective remediation strategy. Locations, dimension and the number of new extraction wells for remediation purposes were optimized. Two new extraction wells were established (Fig. 2) and a 7-day pumping test was conducted in order to verify the calibrated hydraulic conductivity values and predicted BAM concentrations.

3 Results

The saturated hydraulic conductivity was found to be the most sensitive parameter for determining the predicted concentration of BAM in the extraction wells (Table 4). Together with the infiltration rate these parameters account for almost half the uncertainty. The degradation rate of dichlobenil, which was expected to be one of the most uncertain parameters, was found to be the sixth most sensitive parameter.

The simulated and observed concentrations, at one observation well at the nursery and at the extraction wells, are shown in Figs. 4 and 5. The correlation coefficients between the observed and the simulated concentrations are 0.63 for the observation well and 0.40 for the west extraction well. The simulation indicates that the guideline limit was exceeded in 1985 and that the concentration reached a maximum at the end of 1995.

A worst-case scenario was run, based on the uncertainty of the observed and simulated concentrations at the extraction wells. The standard deviations of the residuals of observed and simulated concentrations were used to calculate a 98% confidence

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limit ($\pm 0.095 \mu\text{g l}^{-1}$). In the worst-case scenario, it was assumed that the model underestimates the true concentration, corresponding to the 98% confidence limit. With this condition it was predicted that the BAM concentration will fall below the guideline limit in 2019.

5 The simulated BAM plumes in groundwater from 1985, 1995 and 2012 are shown in Fig. 6. This figure shows how the plume moves in the direction of the extraction wells and that the plume gets progressively smaller towards the front. The main leakage takes place at the southern end of the tree-nursery area, but there is also a small amount of leakage in the northwest. The two plumes enter the esker and become a
10 single plume before reaching the extraction wells. This scenario is supported by the BAM samples in the area (Table 2).

In the modeled scenario, 19 kg of the active substance, dichlobenil, was applied between 1976 and 1991. At the end of the predictive simulation, 0.8 kg remained in the vadose zone as either dichlobenil or BAM, 2.6 kg had left the vadose zone as dissolved
15 dichlobenil or through volatilization of dichlobenil or degradation of BAM, and 15.6 kg had degraded from dichlobenil to BAM and entered the groundwater. Of the 15.6 kg that had entered the groundwater, 8.2 kg was predicted to have been extracted in the wells and 2.5 kg was predicted to have passed the extraction wells because they have been periodically closed.

20 The simulation of the vadose zone gave a median transit time to groundwater of 4.3 yr for water and 7.2 yr for BAM. This corresponds to a median retardation factor of 1.7, although this value varies depending on moisture content, depth, and sorption coefficients at different depths. The simulation of groundwater gave a median transit time between the nursery and the extraction wells of 2.7 yr for water and 4.7 yr for BAM,
25 which also corresponds to a retardation factor of 1.7.

The results of the 7-day pumping test in the two newly established remediation wells (Fig. 2) are compared to the model predictions in Table 5. During the pumping test, groundwater was analyzed twice and the mean concentrations were -1% and $+33\%$, compared to the model results. According to the calibrated model, the recharge rate of

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the nursery's catchment area is 2.6 l s^{-1} . During the test, the pumping rates in the two wells were 0.82 l s^{-1} and 0.27 l s^{-1} and the simulation was run with pumping rates of 1.5 l s^{-1} and 0.75 l s^{-1} . With the remediation wells running from July 2010, and imposing the conditions of the worst-case scenario, it was predicted that the concentration of BAM in the contaminated extraction wells would decrease to less than half the EU guideline limit in 2014. This corresponds to a reduction of five year compared to the situation without the remediation wells.

4 Discussion

The numerical approach applied to a contaminated aquifer in this study, can be compared to the stationary mass-balance model results, based on Monte-Carlo simulations, described by Bergvall et al. (2007) for the same site. The time resolution in the numerical model was days, in contrast to years in the mass-balance model. In groundwater, the transit times from the nursery to the extraction wells were found to be twice as long in the numerical model, compared to the mass-balance model. Mainly, this difference can be explained by differences in aquifer thickness and hydraulic conductivity. In particular, the thin layers of silt that partly cover the esker were not included in the mass-balance model. However, in the vadose zone the models gave similar median transit times for both water and BAM. This is probably a consequence of the thickness of the vadose zone, which results in a transit time of four years for water. Thus, the effect of the different resolution of the infiltration rate in the two models is small. However, extreme infiltration events can lead to higher contaminant concentrations, which may result from macropore flow, reduced biodegradation and sorption. In our case, the effect of the extreme rainfall events in the summer and autumn of 2000 can be seen in the vadose model: they resulted in increased migration and higher concentrations of dichlobenil, but lower concentrations of BAM. In situations where macropore flow is dominant, transient flow conditions may have a large effect on the prediction.

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We considered whether macropore flow should be included in the model. During our simulation time of 33.5 yr, the maximum infiltration rate was 40 mm day^{-1} . Compared to the saturated hydraulic conductivity at the soil surface, this is 10^3 times smaller and indicates that matrix flow is the dominant process. Macropore flow can occur when the water-entry pressure head is higher than -0.15 m (Jarvis, 2007). The pressure heads were studied with our model, but they were not higher than -0.75 m at any time-step or depth. The assumption of no macropore flow was also appropriate, given the analyzed soil textures, which showed that 70 to 94% of the soil above a depth of 2.6 m was classified as medium sand or coarser. At greater depths the fractions of fine sand and silt increase, which may increase the probability of macropore flow. However, at such depths, changes in the water flow are smaller than changes in the infiltration rate at the soil surface. Hence, changes in pressure heads are small. However, macropore flow could be incorporated in the model through the dispersion term, using an apparently higher dispersion than the true dispersion. This approach is valid because of the thickness of the vadose zone and the large area of the source of contamination. For example, the transit time of groundwater from the north to the south end of the nursery is more than one year. The assumption of no macropore flow is supported by Haria et al. (2003), who demonstrated in two field experiments in shallow and deep groundwater, that macropore flow is highly dependent on the depth to groundwater and only occurred at shallow groundwater. The authors explained this finding as being due to the capillary fringe sustaining a higher moisture content in the vadose zone at the shallow groundwater site.

The municipality's sampling procedure has been improved since 2007. Figure 4 shows that the observations do not fluctuate as widely after 2007. This was the motivation for using the integral of the observed concentrations during calibration. A plot of the moving average shows that the integrated result is close to the simulated breakthrough curve, giving a correlation coefficient of 0.99.

The concentration gradient at the front of the plume is steep (Fig. 6). The steepness can be explained by large differences in hydraulic conductivity between the esker and

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the surrounding material, which has a finer texture (Fig. 3). The distinct change from fine to coarse material alters the flow direction of the contaminated water to follow the border of the zone of coarse material. This affects the plume by narrowing the front as it moves towards the extraction wells.

5 In a non-pumping scenario, the BAM-contaminated water plume still moves directly towards the extraction wells, in particular to the west well. In this well, the concentration increases when the pumps are stopped because the contaminated water becomes less diluted (Fig. 5). However, we can see that the opposite happens to the east well, where the concentration decreases when the pumps are stopped. Although we know that all
10 samples since 2007 were taken in the west well, it is not certain which extraction well the older samples come from. If the results from the model are valid, it appears that samples older than 2003 were taken in the east well, but the remaining samples were taken in the west well. This assumption gives a correlation coefficient of 0.61 between observed and simulated concentrations, in comparison to 0.40 if all observations are assumed to come from the west well.
15

In order to speed up the cleaning process, the model was applied to optimize the location of remediation wells (Fig. 2). A 7-day pumping test was conducted in the newly established wells and the results verified the calibrated transmissivities and the predicted BAM concentrations. Transmissivities from the test were found to be 20–
20 30% higher than the calibrated model values (Table 5). The values from the pumping test may, however, have been overestimated because of intense rainfall during the pumping test (16% of the yearly net infiltration rate in one week). The predicted BAM concentration and the result from the pumping test agreed within the analysis error of 35% for both wells. With the remediation wells, it was predicted, as a worst-case scenario, that the extraction wells supplying the municipal drinking-water utility would be clean in four years. Without the remediation wells a similar scenario predicted the
25 extraction wells to be clean in nine years.

One limitation of our model is that breakthrough measurements of the contaminant at the extraction wells are not available, which may be crucial for predicting future

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transport behavior (Eggleston and Rojstaczer, 2000). However, the transport model is not only based on measured head and hydraulic conductivity values, but also on data from twelve years of contaminant measurements at several locations, including the source and the extraction wells. We believe that the results of this model show that it is possible to obtain satisfactory predictions of contaminants, despite the lack of breakthrough measurements, if supported by abundant field data.

5 Conclusions

This paper describes a model for pesticide transport and its application to a pesticide-contaminated esker aquifer consisting of extensive glaciofluvial and littoral sediments. A sensitivity analysis revealed that hydraulic conductivity and infiltration rate account for almost half of the model uncertainty. With the combination of a deep vadose zone and coarse texture, macropore flow was found to be of minor importance for contaminant transport. This is likely to be the case for similar aquifers since glaciofluvial and littoral sediments usually consist of well-sorted, unstructured sand and gravel. Nevertheless, the model can allow for extreme rainfall events since it is run with daily time steps.

The model was used to optimize the location of extraction wells for remediation. Running a worst-case scenario, the prediction showed that the establishment of two extraction wells would clean the aquifer in four years, compared to nine years without them. A pumping test of the two new extraction wells locally verified the calibrated transmissivities and the predicted BAM concentrations.

Further development of the model would require additional field measurements to assess the importance of macropore flow in deep, sandy aquifers. It is also important to characterize the variability of hydraulic conductivity and its effect on contaminant transport in eskers.

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Table 1. Analysis of soil samples taken from the nursery site, including data from Bergvall et al. (2007), and slug tests carried out in the aquifer (Fig. 2). Except for the hydraulic conductivity (K) measurements from slug tests, all data refer to the vadose zone. For each parameter the mean, standard deviation (SD), observed range and number of observations are reported. When the concentration was less than the detection limit ($0.1 \mu\text{g kg}^{-1}$) $0.05 \mu\text{g kg}^{-1}$ was used in the calculation of the mean and standard deviation.

Parameter	Unit	Mean	SD	Min	Max	No.
TOC soil	g kg^{-1}	2.6	1.2	1.0	6.3	55
TOC pore water	mg l^{-1}	4.1	0.8	3.3	5.1	7
pore water content	%	13.8	5.5	2.8	35.2	673
porosity	%	37	2.9	33	43	26
bulk density	kg m^{-3}	1660	78	1500	1780	26
dichlobenil soil	$\mu\text{g kg}^{-1}$	36	137	< 0.1	1100	83
BAM soil	$\mu\text{g kg}^{-1}$	1.2	3.9	< 0.1	27	68
depth to gw	m	10.20	0.06	10.08	10.34	482
K slug test	m s^{-1}	-4.6^{a}	2.4^{b}	-8.1^{b}	-2.6^{b}	44

^a value refers to geometric mean of the common logarithm

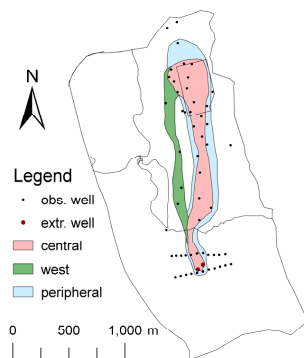
^b value refers to the common logarithm

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Table 2. Concentrations of BAM for the four zones (see attached figure) and three time intervals. For each zone and time interval, the mean, standard deviation (SD), range and number of samples are reported. When the concentration was less than the detection limit ($0.01 \mu\text{g l}^{-1}$) $0.005 \mu\text{g l}^{-1}$ was used in the calculation of the mean and standard deviation.

Years	Central		West		Peripheral		Outside plume	
	Mean (SD)	Range (No.)	Mean (SD)	Range (No.)	Mean (SD)	Range (No.)	Mean (SD)	Range (No.)
1998–2003	6.1 (5.7)	< 0.01–25(30)	0.058 (0.048)	< 0.01–0.13(8)	0.022 (0.028)	< 0.01–0.07(5)	< 0.01 (–)	< 0.01(6)
2004–2007	1.4 (1.3)	0.01–4.3 (50)	0.11 (0.12)	< 0.01–0.33(11)	0.046 (0.035)	< 0.01–0.14(14)	< 0.01 (–)	< 0.01(23)
2007–2009	0.85 (0.78)	< 0.01–2.9(55)	0.013 (0.011)	< 0.01–0.02(2)	0.026 (0.009)	< 0.01–0.06(38)	< 0.01 (–)	< 0.01(23)



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Table 4. Results of the sensitivity analysis for the concentration of BAM in the extraction wells.

Parameter	Symbol	Relative sensitivity (%)
Hydraulic conductivity in aquifer	K_{gw}	27.0
Infiltration rate	I	19.7
Tortuosity factor	n	8.4
Saturated water content	θ	8.1
Sorption of BAM in vadose zone	$K_{d_BAM_vz}$	7.3
Degradation of dichlobenil	μ_{diklo_vz}	7.1
Thickness of vadose zone	d	6.2
Sorption of BAM in groundwater	$K_{d_BAM_gw}$	5.8
Dispersivity in vadose zone	α_{vz}	3.2
Degradation of BAM	μ_{BAM_vz}	3.0
Sorption of dichlobenil	$K_{d_diklo_vz}$	2.0
Characteristic pore size	α	0.9
Hydraulic conductivity in vadose zone	K_{vz}	0.9
Dispersivity in groundwater	α_{gw}	0.4



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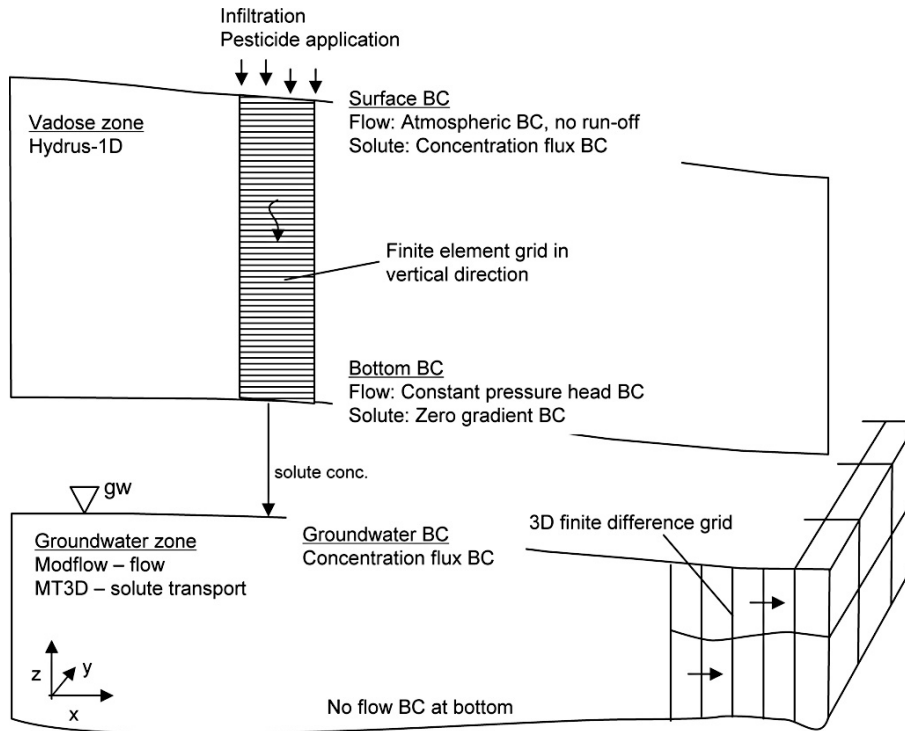


Fig. 1. Modeling procedure and boundary conditions (BC) of water flow and solute transport.

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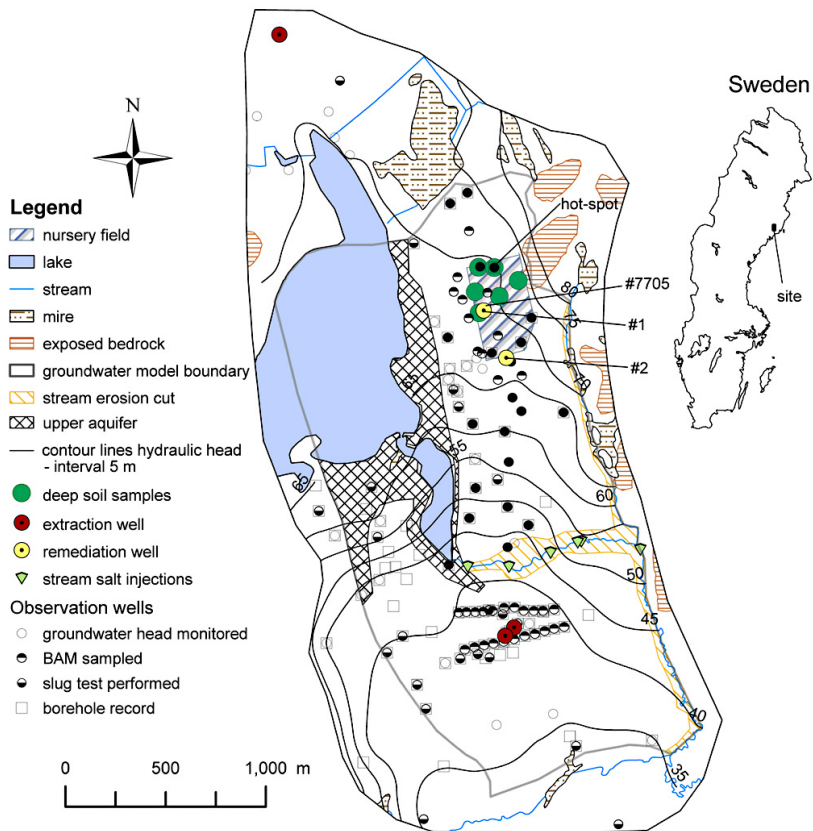


Fig. 2. Location of the site within Sweden and map showing location of the nursery site in the modeled area, observation wells, extraction wells, deep soil samples, and salt injections. The contour lines of the hydraulic head are drawn every 5 m, and lakes, streams, erosion cuts, mires and exposed bedrock are represented. #7705 refers to an observation well; #1 and #2 to remediation wells.

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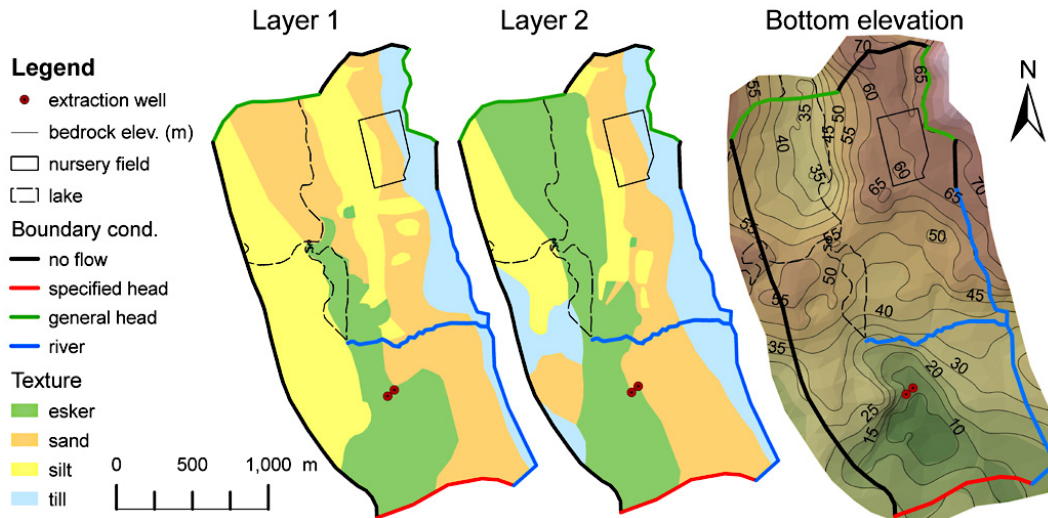


Fig. 3. Boundary conditions of the groundwater flow model. The model consisted of two layers, which were divided into four texture groups with various hydraulic conductivities. The calibrated conductivities were as follows: 25 to 200 m d^{-1} for esker material, 2.0 to 20 m d^{-1} for sand, 4.05×10^{-4} to 1.0 m d^{-1} for silt and 0.20 to 4.0 m d^{-1} for till. The modeled bottom elevation consisted mainly of the bedrock, but surfaces of silt or clay were found in some locations underlying more coarse-grained textures.

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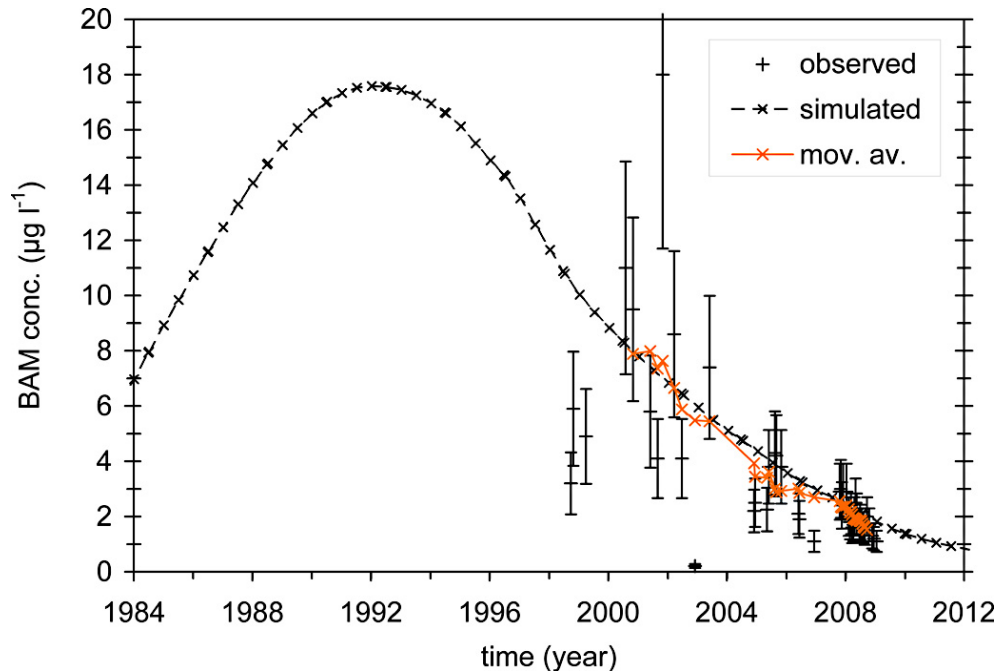


Fig. 4. Simulated and observed concentrations of BAM at observation well #7705 located at the nursery field (see Fig. 2). The analysis error was $\pm 35\%$ and is indicated by error bars. The moving average is the average of the closest four observations, before and after each observation. The observed peak value in 2001 was characterized as an outlier according to Dixon's Q-test.

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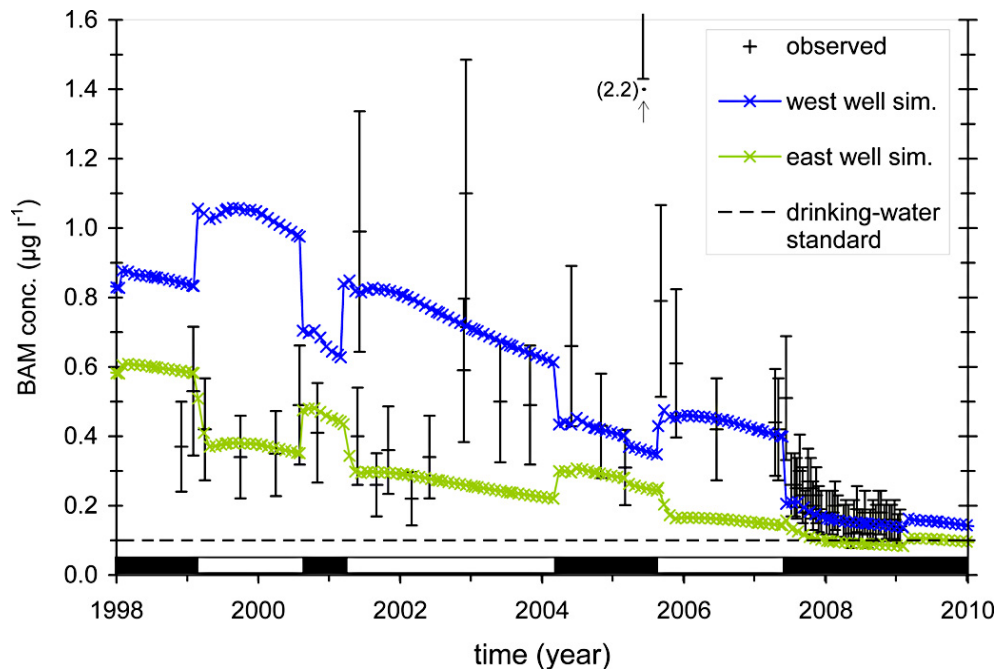


Fig. 5. Simulated and observed concentrations of BAM at the extraction wells. The analysis error was $\pm 35\%$ and is indicated by error bars. Black bar at the bottom of the graph indicates that the wells were running; white bar indicates that they were closed. Note that before 2007, it is unclear whether the observed values were sampled in the west or east well. As from 2007, all observed values refer to the west well. The observed peak value in 2005 was characterized as an outlier, according to Dixon's Q-test. The European drinking water standard is $0.1 \mu\text{g l}^{-1}$ for pesticides.

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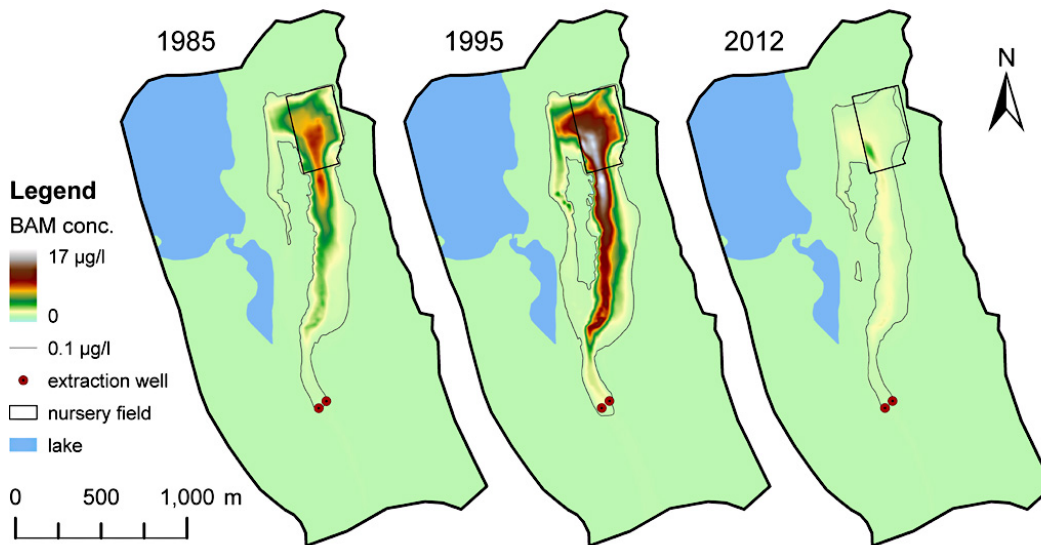


Fig. 6. Simulated BAM plumes in groundwater from 1985, 1995 and 2012. The $0.1 \mu\text{g l}^{-1}$ front of the plumes (drinking-water standard) is indicated by black contours.

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