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Combining data-based and process-based approaches to minimize the complexity of a reactive sulphate transport model

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Abstract In this study, advanced methods of nonlinear data analysis and mechanistic models are combined to investigate transport and turnover of airborne sulphate in the aquifer of a forested watershed. The objective is to understand why adjacent sites differ substantially with respect to short-term and long-term sulphate concentration time series. First, a groundwater model was parameterized as a basis for a coupled model. A detailed analysis revealed substantial uncertainties with respect to, e.g. residence times and the spatial pattern of groundwater discharge. Second, a time series of sulphate concentration in the catchment runoff was analysed using artificial neural networks. The chemical hydrograph provides spatially integrated information about the groundwater of the watershed. In addition, the pronounced short-term dynamics reflect the varying contribution of shallow and deep groundwater on stream discharge. The neural network revealed a substantial change in these dynamics during the last 14 years. Based on these results, only a few of a variety of candidate processes were selected for a mechanistic model of reactive sulphate transport in the aquifer. Sulphate transport and interaction with the matrix is described as an equilibrium sorption process. A very simplified model is used for water transport. Although none of the parameters was fitted by inverse modelling, the model matched the long-term dynamics of sulphate concentration. In addition, it succeeded in giving the envelope of observed short-term variability of sulphate concentration due to the varying contributions of different flow paths. It is concluded that using different process-based and data-based modelling approaches in an iterative way can considerably help the optimization of hydrologic models with respect to the available data, thus avoiding some of the problems associated with over-parameterized mechanistic models.

Key words artificial neural network; groundwater; process-based model; stream water; sulphate

INTRODUCTION

Bottom-up modelling by coupled groundwater and hydrochemical models suffers from the well-known problems of parameter identification and non-uniqueness of model solutions. Thus, one objective is to minimize the model complexity as much as possible, without decreasing the model performance with respect to a given set of observations.

In this study, advanced methods of nonlinear data analysis and mechanistic models were combined to investigate transport and turnover of airborne sulphate in the aquifer

of a forested watershed. In spite of substantially decreasing sulphate deposition in the last decades, stream water and groundwater still exhibit high sulphate concentrations. In addition, sulphate dynamics differ substantially at different sampling sites. The objective of the study is to identify the key processes of sulphate dynamics, taking into account the uncertainties of parameter values.

SITE DESCRIPTION

The study site is the 4.2 km^2 Lehstenbach catchment in the Fichtelgebirge mountains in southern Germany (Lischeid *et al.*, 1998). The thickness of the regolith is up to ~40 m. Due to Tertiary weathering, the aquifer is highly heterogeneous at the 10^0 m to 10^2 m scale, and comprised granite boulders, weathered granite, and sandy loam. However, there is no evidence for extended layering. Fractures in the underlying granite bedrock are supposed to have only minor effects on groundwater flow. Among the soils, dystric cambisols predominate. One third of the catchment is covered by peaty soils and bogs. The catchment is drained by a dense and irregular network of brooks and ditches and is nearly completely covered by a Norway spruce forest. Mean annual groundwater recharge in the years of the study add up to 420 mm $year^{-1}$.

Discharge at the catchment outlet has been measured continuously since 1987. Groundwater was monitored in biweekly to monthly intervals in 13 observation wells. Depth of the wells was between 10 and 24 m. None of them was fully penetrating. Samples have been taken by a submersed pump every 1–4 months since 1987. Transmissivity was determined by pumping tests at all wells (Hauck, 1999). There was no production well in the catchment area.

The region was subject to very high sulphate deposition with peak values of about 3 kmol ha^{-1} $year^{-1}$ in the early 1970s, decreasing to less than 0.5 kmol ha^{-1} $year^{-1}$ at the end of the 1990s. However, stream water and groundwater still exhibited high sulphate concentrations. The total sulphur pool in the upper 10 m layer of the aquifer was about 90 kmol ha^{-1}, exclusively due to deposition (Manderscheid *et al.*, 2000).

THE GROUNDWATER MODEL: MODFLOW

In order to better understand the turnover of nonpoint source pollutants in the subsoil, the well-known groundwater model MODFLOW (McDonald & Harbaugh, 1984) was applied. MODFLOW is a block-centred finite-difference code that simulates three-dimensional (3-D) groundwater flow under steady-state, as well as under transient conditions. The modelling environment Visual MODFLOW was used to easily set the input parameters and to visualize the results.

Hydraulic conductivity, determined by pumping tests at 13 groundwater wells, revealed a lognormal distribution with a $\log_{10}$-mean value of 2.4 m day^{-1}. Part of the pumping tests also allowed the determination of the effective porosity, with a mean value of 8%. There was no evidence for stream bed clogging, due to the dominance of sand, gruss, granite and quartz boulders. Thus, a rather high value of 50 m^2 day^{-1} was assumed. The model was not sensitive to changes in the conductance term in a range of several orders of magnitude (Hauck, 1999).

Horizontal spacing of the grid is 25 m, and four layers of about 10 m in the vertical direction. The model results given below refer to steady-state conditions. No-flow boundaries are assumed at the catchment boundary and at the bottom of the lowest layer. Recharge is spatially homogeneous. Groundwater discharge was simulated as drains with an elevation of 1 m below ground level.

Ten realizations of randomly-distributed conductivity data corresponding to the observed distribution were run. Thickness of the aquifer was 40 m in all of these realizations. In a second step, hydraulic conductivity was constant but thickness of the aquifer varied randomly, corresponding to the frequency distribution of the field data. Thus, transmissivity was varied 20 times. This had only minor effects on simulated heads. In addition, annual mean groundwater heads could be simulated with 0.1 m precision with slight adjustments of the hydraulic conductivity.

The impact on residence times was much more pronounced (see Fig. 1). Standard deviation was roughly half of the mean values. Simulated groundwater age was substantially off compared to ^{18}O data (Zahn, 1995) at two out of five wells. These two wells (GW01, GW06) are both located near the water divide. However, the range of residence times for the same groundwater exhibited substantial uncertainty, e.g. at well GW16 the standard deviations of residence time calculated by different model realizations covered the range of 2–17 years. This has severe implications for, e.g. assessing the spreading of contaminants in the groundwater, especially if kinetics come into play.

The same holds for the spatial pattern of groundwater discharge that was determined in the field by sequential discharge measurements under baseflow conditions (not shown). In addition, performance of the different model realizations was independent from that of groundwater heads. It is concluded that both groundwater residence time and spatial patterns of groundwater discharge are subject to enormous

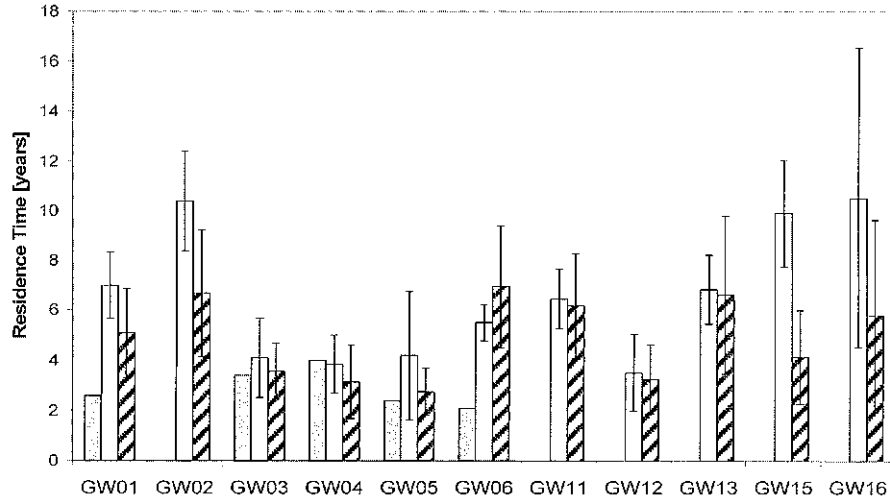

Fig. 1 Mean groundwater age measured by ^{18}O (Zahn, 1995) (solid grey column) and simulated for varying hydraulic conductivity (white column) and varying thickness of the aquifer (hatched column). For simulated data, mean values and standard deviations (10 model realizations each) are given.

model uncertainty. This uncertainty is likely to increase further when the groundwater model is coupled with a kinetic solute transport model.

THE NEURAL NETWORK MODEL

Solute concentration in the catchment runoff provides spatially integrated information about catchment groundwater concentration. The dynamics of sulphate concentration in the catchment runoff was analysed by artificial neural networks to provide information about the predominating processes (Lischeid, 2001).

The neural network model explained about 80% of the short-term variance (Lischeid, 2001). Out of a variety of candidate variables, only three were identified as relevant: daily mean discharge of the sampling day, a measure of the length of time since the end of the preceding baseflow period, and the sliding mean of annual mean throughfall concentration. In contrast, according to the model, neither air temperature, nor snowmelt dynamics or short-term dynamics of throughfall concentration had any influence on the sulphate dynamics. This provides some evidence that biological sulphate processing on the one hand, and preferential flow phenomena on the other, can be neglected. The latter was confirmed by a detailed field study (Lischeid *et al.*, 2002b).

Instead, the flashy response of stream sulphate concentration reflects the vertical gradient of sulphate concentration in the groundwater (Lischeid, 2001; Lischeid *et al.*, 2002b). Visualization of the multivariate, nonlinear regression planes indicated a clear

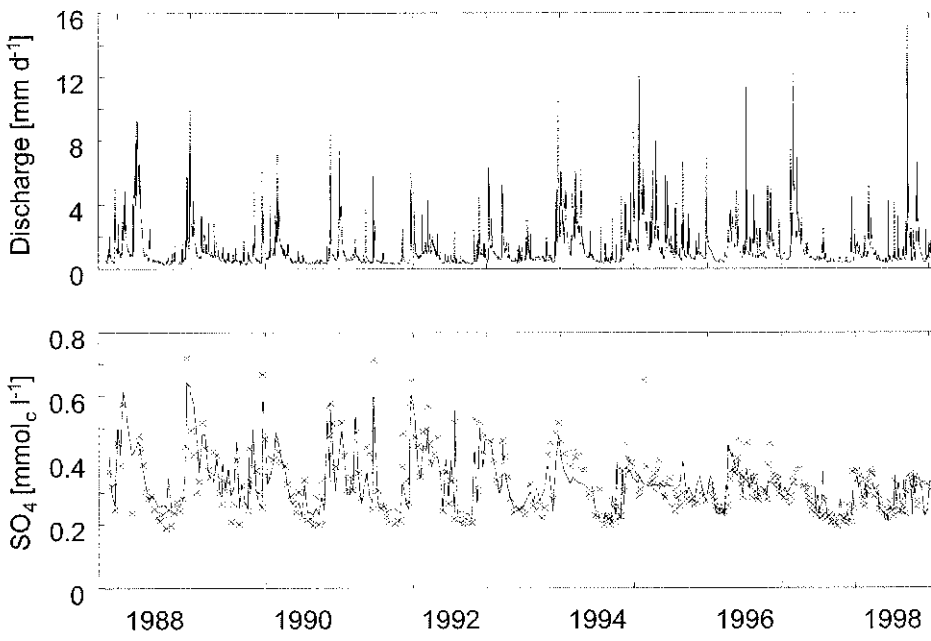

Fig. 2 Time series of discharge (upper panel), observed (x) and simulated (black line) sulphate concentration in the Lehstenbach catchment runoff.

decrease of sulphate concentration of the shallow groundwater since 1988. This is consistent with that observed in the vadoze zone. It is concluded that most of the sulphate dynamics can be described by assuming conservative mixing of shallow groundwater with high, but clearly decreasing, sulphate concentration with baseflow that contributes low, constant sulphate concentration to the stream. The process-based model that is described in the following section is intended to explain the depth-dependent long-term behaviour in the groundwater.

THE SULPHATE MODEL: SUNFLOW

In contrast to that in the catchment runoff, sulphate concentration in upslope springs did not start to decrease before 1995, and is still exceeding 0.5 $mmol_c\ l^{-1}$. Furthermore, sulphate concentration continues to increase significantly in the deeper layers of the aquifer, now clearly exceeding that of the catchment runoff during baseflow.

The SUNFLOW model was developed to investigate these spatial patterns (Büttcher, 2001; Büttcher & Lischeid, 2003). To enable a rigorous test with respect to process identification, model complexity was minimized, based on the results of the preceding studies. The model considers sulphate transport and sorption along 1-D flowpaths. The time step of the model is one year.

The aquifer is assumed to be homogeneous. The model considers vertical seepage flux in the unsaturated zone and lateral groundwater flow towards the streams. The interaction between the liquid and the solid phase is described by Langmuir isotherms, determined in samples from drilling cores of up to 10 m depth (Manderscheid *et al.*, 2000), and distinguishing between adsorption and desorption. A Monte Carlo simulation provided for spatial heterogeneity of sorption isotherms.

Groundwater recharge is assumed to be constant and to be equal to annual mean discharge measured from 1987 to 2000 at the catchment outlet. In contrast, annual sulphate deposition is reconstructed based on European data by Alewell (1995) for 1854–1987, and is set equal to annual throughfall flux, measured by bulk samplers inside the catchment thereafter.

To allow a rigorous test of the model, none of the parameters was fitted by inverse modelling. In order to account for the uncertainty of some key parameters, primarily the thickness of the regolith and the fine soil fraction of the aquifer matrix, the model was run with a "most plausible" parameterization as well as with parameter sets assumed to be extreme. The reader is referred to Büttcher (2001) and Büttcher & Lischeid (2003) for details.

In the model, the only difference between individual sampling sites within a catchment is the different shape of their subcatchments, represented by the flowpath lengths distribution. This approach was able to reproduce the different observed long-term dynamics at different sampling sites. According to the model, short flowpaths, which react very rapidly to decreasing atmospheric sulphur depositions, give rise to the decrease of sulphate concentrations at single stream water sampling sites. In contrast, the considerable delay of sulphate breakthrough of long flowpaths can explain still increasing concentrations at single groundwater wells. Moreover, the considerable short-term variability observed at single stream water sampling sites could be reproduced by considering long and short flowpaths separately (Fig. 3). Model results

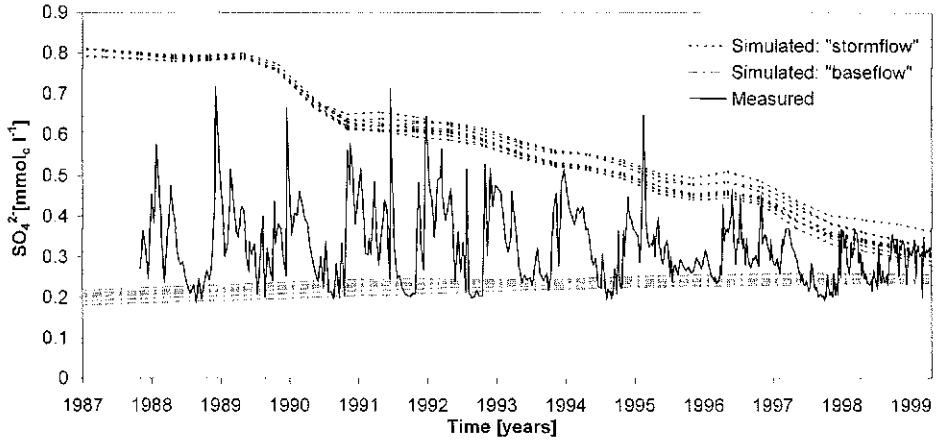

Fig. 3 Measured sulphate concentration in the catchment runoff (biweekly intervals), and simulated sulphate concentration in groundwater for flowpath lengths ≤150 m ("stormflow") and >150 m ("baseflow"), respectively, (annual mean values; 10 replicates each to account for spatial heterogeneity of sorption isotherms).

therefore give clear evidence that the observed spatial variability of sulphate dynamics can be traced back to sorption processes along flowpaths of different lengths (Büttcher, 2001, Lischeid *et al.*, 2002a).

CONCLUSIONS

Due to increasing process understanding and increasing computer power, the complexity of mechanistic hydrological models has increased tremendously in the last decades. However, the available data often do not support complex models, as could be shown in this study.

One way to handle this problem is to add additional data sets, as well as to combine different modelling approaches, comprised of process-based and data-based models. Stream water solute concentration and its short-term dynamics can be used to extract information about groundwater quality, circumventing some of the problems of the spatial heterogeneity of groundwater quality data. Advanced methods of data analysis, such as artificial neural networks, can considerably help to better understand the dynamics of observed data.

This helped considerably to reduce the number of candidate processes to be included into the process-based model, and to reduce the associated problems of model parameterization. Only then was it possible to use the model for process identification. We suggest the combination of different modelling approaches in an iterative way to optimize mechanistic models with respect to the available data.

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