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Environmental Geology

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Original Article

Modelling seasonal variations in nitrate and sulphate concentrations in a vulnerable alluvial aquifer

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Abstract The Eisden–Meeswijk region in Belgium has been affected by mining subsidence due to the deep coal mining activities. Groundwater levels in the alluvial plain of the Meuse River are maintained below the ground surface by drainage installations and municipal well fields. A correlation between the water level in the Meuse River and the variation in nitrate and sulphate concentrations in the aquifer has been observed. A transient groundwater model is developed for the period May 1998–May 2002 and advective transport simulations have been carried out using this model. During dry periods, the major groundwater flow is directed towards the Meuse River, thereby feeding the river. During wet periods, however, groundwater flows in the opposite direction. Due to these variations in groundwater flow direction and to the extraction of groundwater, zones of higher solute concentration exist of which the position and extension vary both spatially and temporally.

Keywords Groundwater flow · Transport · Nitrate · Sulphate · Belgium

Introduction

The city of Eisden is situated in the northeast of Belgium, along the Meuse River, which forms the border with The Netherlands (Fig. 1). In the past century, there was intensive coal mining activity in this region. From 1922 until 1987, 73,191,893 tons of coal were mined in Eisden at a depth up to 700 m in the carboniferous coal mines (Dusar 1996). As a result of the coal exploitation, irregular land subsidence was induced in the area. According to Vansteelandt (1984a), the subsidence reached up to 7 m near the city centre of Eisden, affecting an area of 44.70 km^2 (Fig. 1). These changes in topography led to (1) building damages, (2) a need for higher embankments of the main canal “Zuid-Willems” and (3) changes in the flow direction of small streams like the Vrietselbeek and the Genootsbeek. These two small streams originally drained the area towards the Meuse River, but nowadays, surface water flow is directed towards the new lowest topographical points. Since groundwater levels are still in equilibrium with the water level of the Meuse River, land subsidence and the lack of exit points for the streams could theoretically cause the water level to locally exceed the ground level. In order to prevent flooding, the agency “Dienst Mijnschade”, who is responsible for mitigating the damages caused by the coal mining activities, has installed pumping installations on both streams. Excessive water is pumped towards the Zuid-Willems Canal and the Meuse River. In Leut, a drainage system has been placed beneath the main street of the village (Fig. 1)to drain excess water towards the Meuse River. Additionally, the drinking water company VMW installed two well fields of 14 pumping wells each in Eisden and Meeswijk for drinking water production. The well screens are located in the shallow, alluvial aquifer, consisting of mixed gravel and sand deposits from the Meuse River.

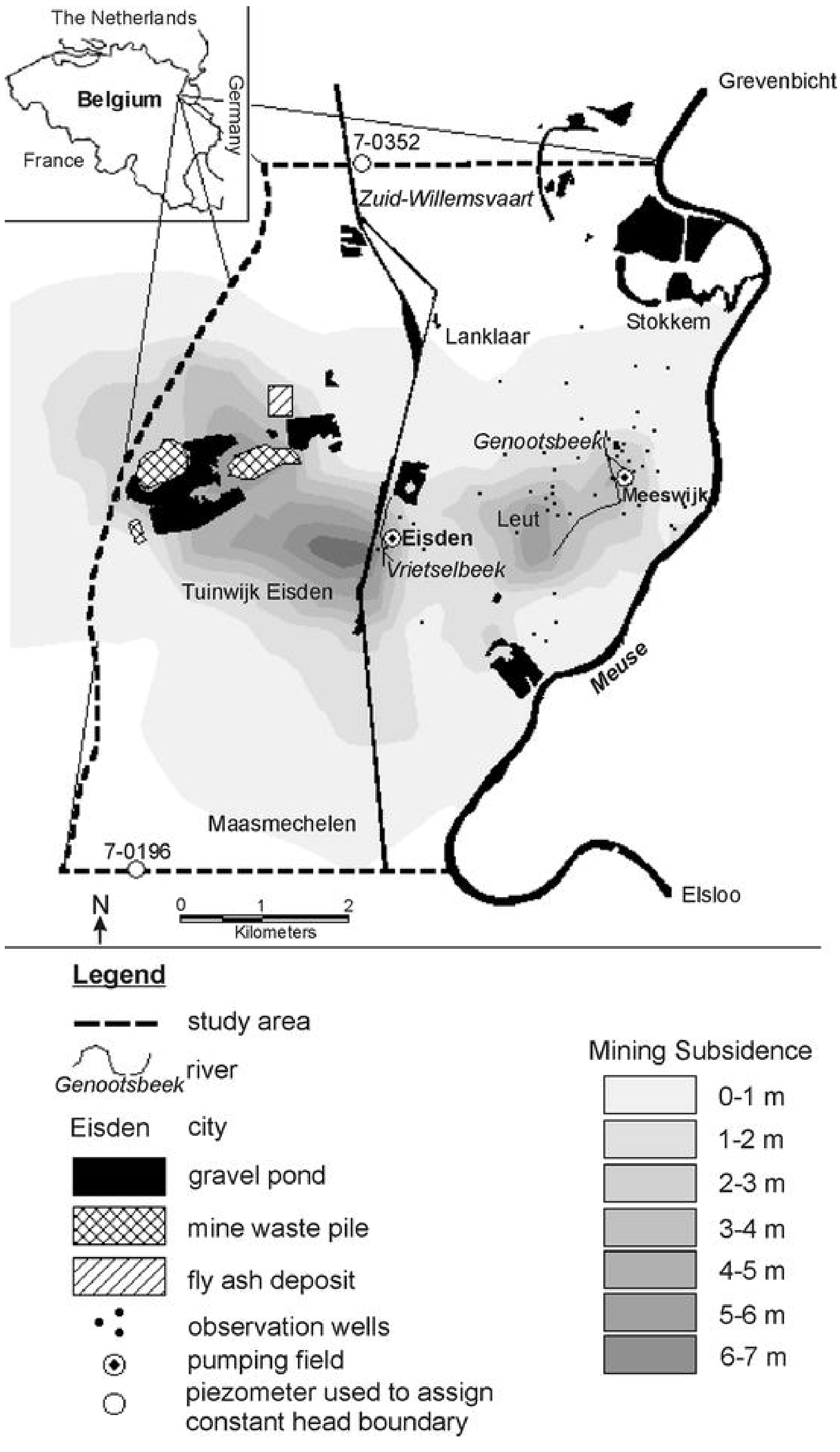

Fig. 1 Location of the region studied with indication of the calculated mining subsidence (after Vansteelandt 1984b)

The main concerns for the quality of drinking water are the highly variable sulphate and nitrate concentrations in the groundwater. The main source of sulphate in the aquifer is acid mine drainage from the mine tailings of the former coal mine in Eisden. The mine tailings cover an area of approximately 17 ha and reach a height of 80 m. The tailings mainly consist of sandstone and shale with a pyrite content of 0.31–1.39% in weight (Cammaer 1998). Oxidation of pyrite by infiltrating water results in sulphate-rich groundwater (Sams and Beer 2000; Denimal and others 2002). On the other hand, about half of the land surface in the region studied is used for agriculture, i.e. mostly pastures and cornfields. There, the use of fertilizers is the main source of nitrate in groundwater (Canter 1996; Walraevens and others 2003).

Monthly measurements of nitrate and sulphate contents in the pumping wells and biannual sampling in the monitoring wells, show an increase in nitrate concentrations when the river level of the Meuse is high, and an increase in sulphate concentrations when river levels are low.

The objective of this study is to investigate both the temporal and the spatial variations of the nitrate and sulphate concentrations in the alluvial aquifer. This is done by simulating the solute flowpaths for a 4-year period by means of a transient groundwater model.

Methods

A transient groundwater model for a simulation period from May 1998 until May 2002 is created, using modflow (McDonald and others 1988) in combination with the groundwater modelling system (GMS; Environmental Modelling Systems, Inc. 2003) as the pre–and postprocessor.

Flowpaths of solutes in the alluvial aquifer are simulated using modpath (Pollock 1994). Advective transport computations are also used for delineating the capture zones around the VMW well fields. The modelling of seasonal variations in sulphate and nitrate concentrations are modelled with mt3d (Zheng and Wang 1999) in transient conditions.

Geology

The region studied is a part of the Campine Basin (southern North Sea Basin) and consists of Tertiary clay and sand deposits dipping 0.7° NNE, discordantly overlain by Quaternary alluvial gravels and cover sands. To the south of the study area, Oligocene marine clay overlain by Oligocene clayey sands is found. Miocene sands were deposited on top of these sediments (Buffel and others 2001). There are four NW–SE oriented, east-vergent normal faults in the area, of which only the most northern fault was active until the Quaternary (Houtgast and others 2002; Fig. 2).

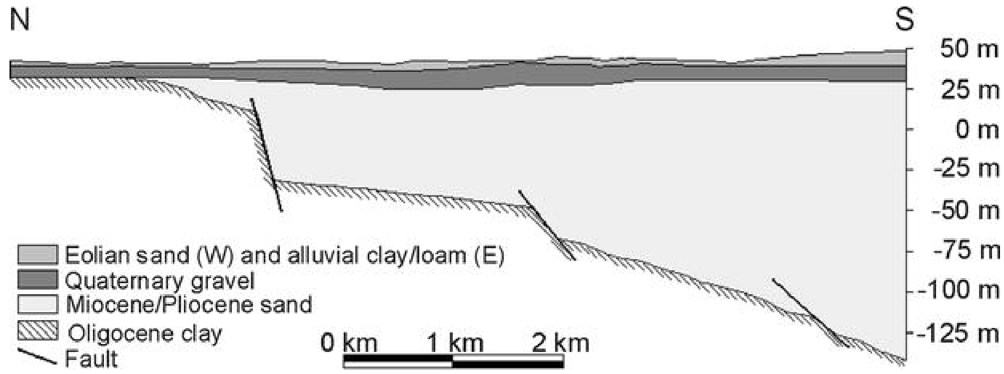

Fig. 2 N–S cross-section of the area studied

After the incision of the Meuse River during the Saale-glacial, alluvial gravels were deposited in theregion studied. Clayey or loamy lenses can be locally present in these gravels. As these gravels are mined as building material by dredging, large ponds of surface water have been left in the area (Fig. 1). The gravel deposits are characterized by a very irregular bottom due to frequent reworking of the deposits, especially in the Holocene alluvial plain, the eastern half of the area studied. The alluvial gravels are covered with eolian sands to the west, and alluvial clay and loam in the Holocene alluvial plain (Paulissen 1973).

The geometry of the different geological layers was assessed by borehole data, cone penetration tests and existing geological maps (Buffel and others 2001). However, care had to be taken since absolute elevation measurements in the log descriptions from the boreholes drilled before or during the mining subsidence were not corrected for subsidence in the archives. Similarly, elevation measurements from drillings made after the mining subsidence are often measured to a reference point that was not corrected for the subsidence. So, in the accurate description of the layer geometry, especially the geometry of the gravel layer, corrections were made based on the subsidence map calculated by Vansteelandt (1984a, 1984b). Then the corrected data were interpolated using kriging to a triangulate irregular network (TIN). Based both on the subsidence map and the geomorphologic map by Paulissen (1973), the TINs were manually adjusted to account for the irregularities in the gravel layer by abandoned stream channels.

Groundwater model

Boundary conditions

The top of the Oligocene clay-deposits irepresents the bottom of the model due to the low permeability of this deposit. Since, the dominant groundwater flow is directed to the east to the northern and southern boundaries were chosen orthogonal to the equipotential lines. In each case, the prescribed piezometric levels will be estimated from a piezometer nearby: piezometer 7–0196 in the south and piezometer 7–0352 in the north (Fig. 1). The western border was chosen at the edge of the alluvial valley, since a steep groundwater gradient is observed between the Campine Plateau and the alluvial valley (Loy 1980). The boundaries were assigned specified heads (Dirichlet boundary), variable in time, based on measurements in the aforementioned piezometers and the regional groundwater map by Loy (1980).

The model is bordered in the east by the Meuse River. Since the bottom of the river mainly consists of mixed gravels, one can consider that there is no difference in conductivity between the river bed itself and the alluvial aquifer. Therefore, a specified head condition equal to the level of the river was chosen to represent the Meuse. The specified heads are assigned along the river based on river stage measurements in Elsloo (upriver) and Grevenbicht (downriver), following a longitudinal river profile.

Groundwater abstractions in the well fields are entered in the model based on monthly abstraction data provided by the VMW. The water fluxes, pumped or drained in the area (Eisden, Leut and Meeswijk, Fig. 1) to keep the water level beneath the ground level, are introduced as a monthly areal flux applied to the drained grid-cells.

An estimation of the recharge of the aquifer is found by applying the method of Thornthwaite (1948). Rainfall and temperature data were provided by the Royal Institute for Meteorology of Belgium and the runoff is estimated to be 20% of the rainfall. The large ponds left after dredging gravel are represented in the model through cells with high conductivity (0.1 m/s; Reilly 2001).

Grid

A three-layered grid of 134 rows and 138 columns is constructed, representing, from top to bottom, the alluvial gravel deposits, the Miocene–Pliocene sands and the Oligocene clayey sands. The largest dimension of a grid cell is 100 m and the grid is progressively refined to a minimum cell with a dimension of 20 by 20 m near the pumping wells.

Hydraulic conductivity

Several pumping tests in the well fields of Eisden and Meeswijk were previously conducted by the water company. The average hydraulic conductivity values obtained for the gravel are compromised between 0.039 and 0.014 m/s. Since the pumping tests were carried out on a local scale, an initial, uniform value of 0.010 m/s was assigned to the gravel layer in the model domain. This value has been locally adapted as part of the groundwater model calibration.

No pumping test data are available in the sand deposits. Hydraulic conductivity values for these layers are taken from a previous groundwater study (IWACO 1998). A K-value of 8.0×10^{-5} m/s is assigned to the bottom layer, representing the clayey sands, and a value of 1.8×10^{-4} m/s is used for the Miocene–Pliocene sands. A storage coefficient equivalent to the specific yield is chosen at 0.1, since it is a water table aquifer (Spitz and Moreno 1996).

Calibration

Measured groundwater levels from 54 piezometers are available for calibration (Fig. 1). In a first step, the groundwater model is calibrated on two steady-state conditions: May 1998 and November 2001, representing respectively a dry and a wet period. During this calibration step, emphasis is given to changing the hydraulic conductivity values of the gravel layer by trial and error. Best results were obtained when a hydraulic conductivity of 0.010 m/s was assigned to the gravel in the Holocene alluvial plain (E), and a value of 0.0069 m/s was used in the western part of the model area. This distribution of hydraulic conductivity values can be explained by the fact that the gravels in the Holocene alluvial plain are better sorted because of the frequent reworking of the gravels during the Holocene, by meandering of the Meuse river. Figure 3 shows the calibration results, in terms of scatterplots by comparing the observed heads in the piezometers to the calculated heads in the grid cells.

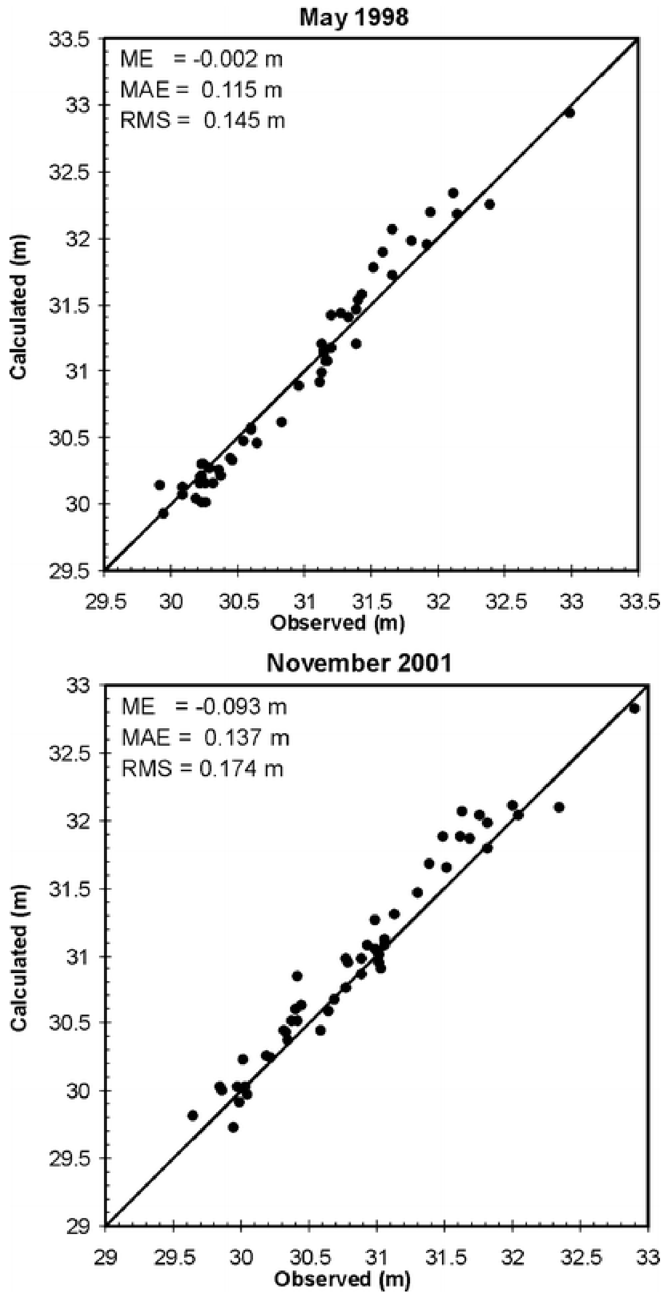

Fig. 3 Observed vs. calculated piezometric heads for the steady-state calibrations

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These steady-state calibrations are then used as starting conditions for the further calibration of the transient simulation for the period from May 1998 to May 2002. The 4-year period is subdivided in stress periods of one month and time periods of two weeks each.

The effective recharge and the abstracted fluxes were deliberately included in the parameters subject to possible changes for obtaining a calibration of the transient model. Calibration of the recharge can be justified by the rough assessment of the runoff and the uncertainty associated with the use of the empirical method of Thornthwaite. It seems that the best results of the model are obtained if calculated recharge values are assigned to the western part of the model domain, the recharge in the Holocene alluvial plain (E) being set only to 60% of this calculated value. This difference is caused by the presence of loamy and clayey soils in the eastern part compared to more sandy soils in the west (Paulissen 1973).

A slight calibration of the drained and pumped fluxes is allowed because of the large uncertainty lying in the values of the effective abstracted fluxes. The uncertainty arises because only monthly total values are available, and for the municipal well fields only values of the global production of the fourteen pumping wells together is available. Another source of uncertainty is introduced since the extent of the drained surface is not exactly known.

Calibration is performed by evaluating the performance of the model compared to measured groundwater heads for each time step. Time series comparing computed and observed piezometric heads for two example piezometers are given in Fig. 4, showing an acceptable calibration.

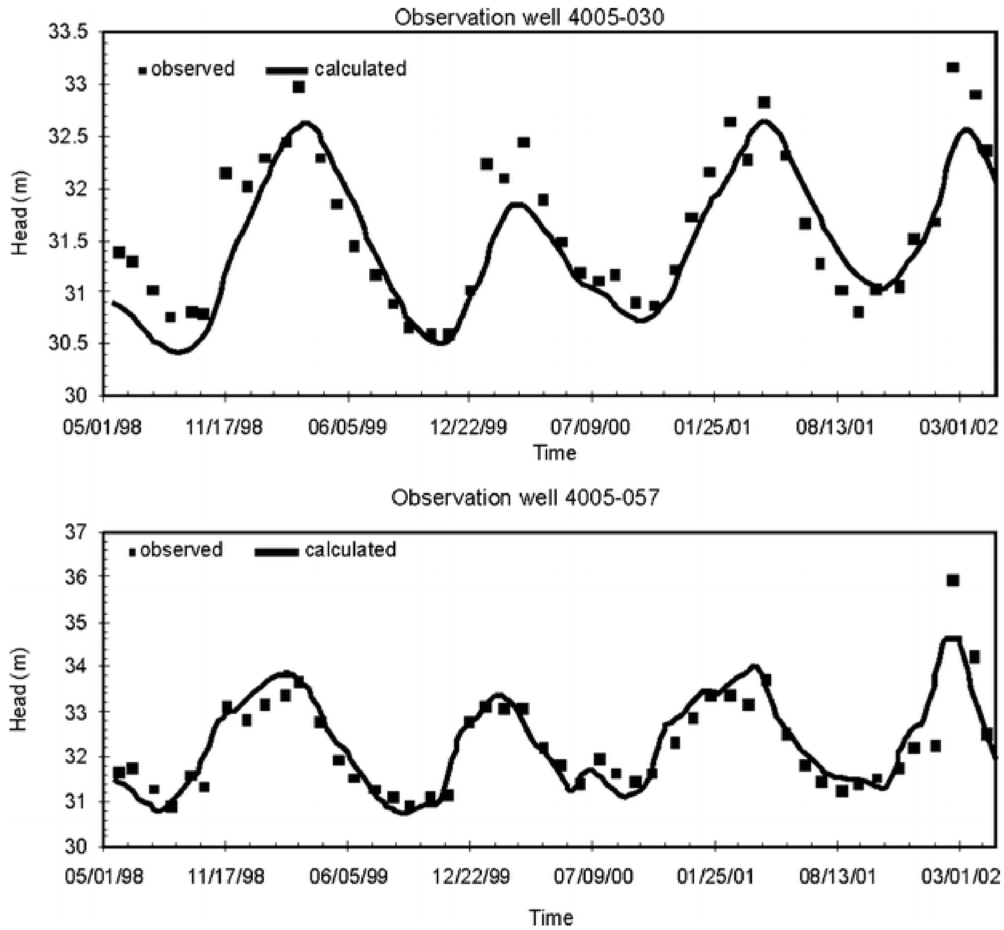

Fig. 4 Observed and calculated time series for observation wells 4005-030 and 4005-057

Results

The calculated piezometric contour maps (Fig. 5) clearly show (1) the generally eastward flow direction and (2) the local drawdowns near the main well fields and (3) to a lesser extent, a local depression near the drain. The transient groundwater flow simulation also demonstrates the great influence of the Meuse River on flow conditions in the aquifer. When river levels are low, i.e. in summer, the river is fed by the aquifer and groundwater flow is directed towards the river. When high river levels prevail, like in winter, the river feeds the aquifer which causes a direction change for the groundwater flow in the uttermost eastern part of the studied region.

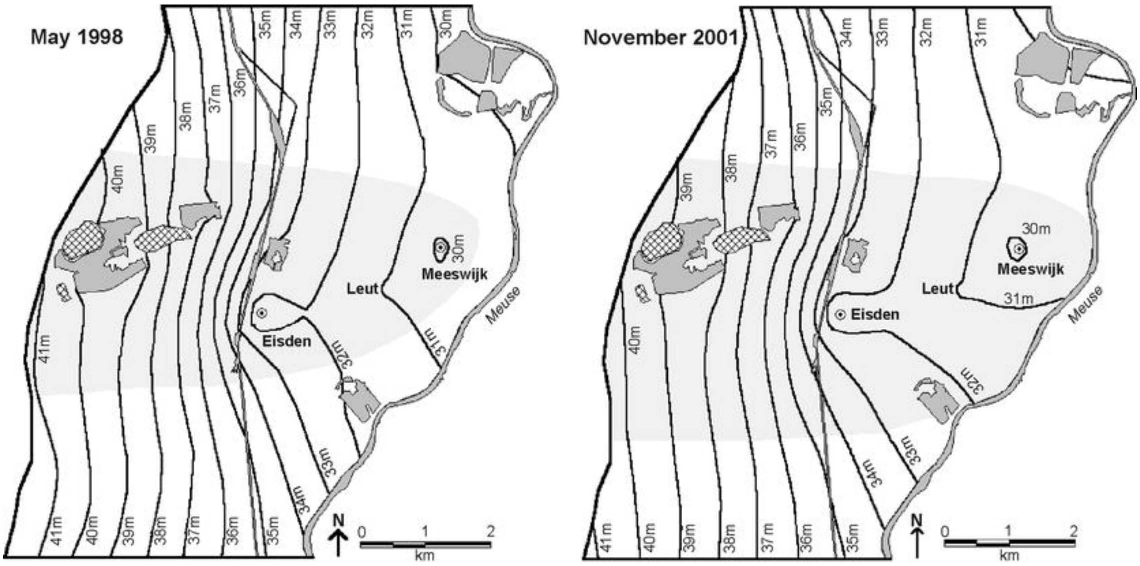

Fig. 5 Calculated piezometric maps. Captures zones for the pumping fields are indicated in *grey*

Transport simulation

Methods

Simulation of nitrate and sulphate transport in theregion studied is carried out using two different approaches. First, the capture zone delineation of the well fields is carried out by applying backward particle tracking to the two different steady state groundwater flow conditions. Secondly, simulation of solute transport from spatially and temporally variable sources is performed by a three-dimensional transport model based on the transient groundwater flow model.

The capture zones are calculated for both well fields using the backward particle tracking software modpath (Pollock 1994). As mentioned before, the effective porosity of the gravel layer is set to a value of 0.1, based on former studies (IWACO 1998) and literature values in similar conditions (Anderson and Woesner 1996).

The mt3d code (Zheng and Wang 1999) is used to simulate the transport of solutes in the saturated zone by advective transport, using the hybrid method Of characteristics (HMOC), based on the transient flow calibration. Due to numerical limitations, the time steps used for transport are determined by the computer program. The size of the time steps is approximately 18 h.

Because the dominant transport mechanism in this aquifer is advective transport, it has been decided, at this stage of the study to disregard the dispersion component of transport (Zheng and Bennet 1995). Since the aquifer is unconfined and relatively thin, dissolved oxygen is assumed to be present in the aquifer therefore denitrification and sulphate reduction processes can be neglected (Berner 1981).

Input

Assessing the actual input of sulphate and nitrate in the aquifer through vertical leaching from the mine tailings and from the agricultural land surface is very difficult. Since the goal of the study is to simulate the general dynamics and the flowpaths of both solutes, a qualitative approach is chosen. The amount of nitrate reaching the saturated zone is assessed using the land use map and the crop map. Based on a study of the Flemish government (VLM 2003), the flux of nitrate at a depth of 90 cm on a clayey or loamy soil is 344 mg/L for pasture and 188 mg/L for corn. Following this study, and correcting for the depth of the water table mostly exceeding 90 cm, the flux of nitrate reaching the water table is set to 130 mg/L for pasture and 100 mg/L for corn. The areas not intensively used for agriculture are assigned a background value of 5 mg/L for nitrate infiltrating the aquifer by percolating water due to atmospheric deposition of nitrogen.

The main source of sulphate in the region is acid mine-drainage from the waste piles of the Eisden coal mine. A distinction can be made between the waste piles and the fly-ash deposits. The water recharging the aquifer through the waste piles is assigned a concentration of 300 mg/L and water flowing through the fly-ash deposits a concentration of 150 mg/L. These values are based on sulphate measurements in nearby observation wells. For the purpose of modelling, initial concentrations, both for infiltrating water and groundwater, is set to 25 mg/L.

These concentration values chosen for the nitrate and sulphate reaching the groundwater are probably not the actual concentrations entering the saturated zone, but they are at this stage of the study, the most realistic values for the numerical simulation and tend to explain the temporal and spatial variations of sulphate and nitrate in the area studied.

Results

The capture zone delineation (Fig. 5) for the Eisden well field, shows that the capture zone encompasses the waste piles and fly-ash deposits of the coal mine at Eisden. The flow lines calculated by modpath show that most of the capture zone can be divided in two main regions: one region where groundwater streamlines reach the well field from the northwest, the other region where it is flowing from the southeast.

The biggest difference between the two periods (May 1998 and November 2001) is observed for the well field located closest to the Meuse River. Since most of the groundwater comes from the northwest of the pumping wells during the dry period, the capture zone extends towards the east and even reaches the Meuse River during the wet period. Based on flow lines, it can be observed that the major part of the groundwater abstracted is flowing from the northwest, but there is still a considerable amount coming from the southwest. Simulation of nitrate and sulphate transport during the period May 1998 through May 2002 is carried out using input concentrations as described previously.

Nitrate

The simulation shows that nitrate coming from the most northern source areas is transported towards the Meuse River and does not flow towards the pumping wells. In this simulation, the influence of the Meuse River is clearly illustrated (Fig. 6). During summer periods most of the nitrate is transported towards the river. During winter periods however, nitrate transport towards the river is not possible due to the groundwater flow inversion in the aquifer. This results in a zone parallel to the river where nitrate is temporally stored and then mainly transported towards the pumping field.

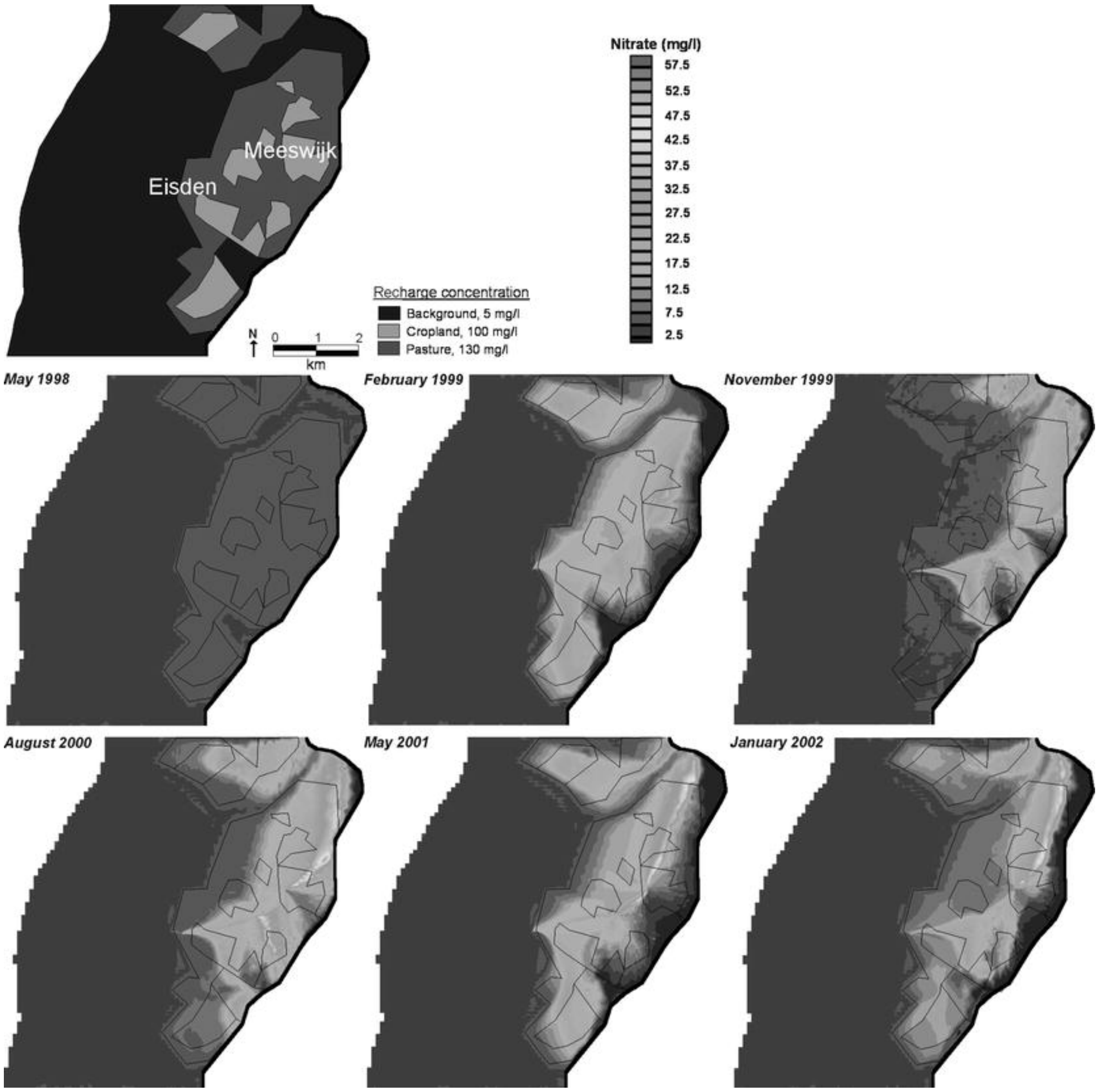

Fig. 6 Calculated nitrate concentrations in the studied region (mg/L) for different periods between May 1998 and January 2002

A similar increase in nitrate concentrations can also be observed between the pumping fields of Eisden and Meeswijk. In this zone, advection lines towards both well fields converge, as already noticed in the capture zone simulation.

Sulphate

Results of the simulation of advective transport of sulphate are provided in Fig. 7. As could be expected from the capture zone simulation, most of the sulphate leaching from the waste piles is transported in the aquifer towards the Eisden well field. It is, however, possible to differentiate between two main plumes. A southern plume reaches the pumping wells from the west, as the northern plume makes an outflanking movement to eventually reach the pumping wells from the north. Sulphate derived from the fly-ash deposit in the north is mostly transported towards the Meeswijk pumping field.

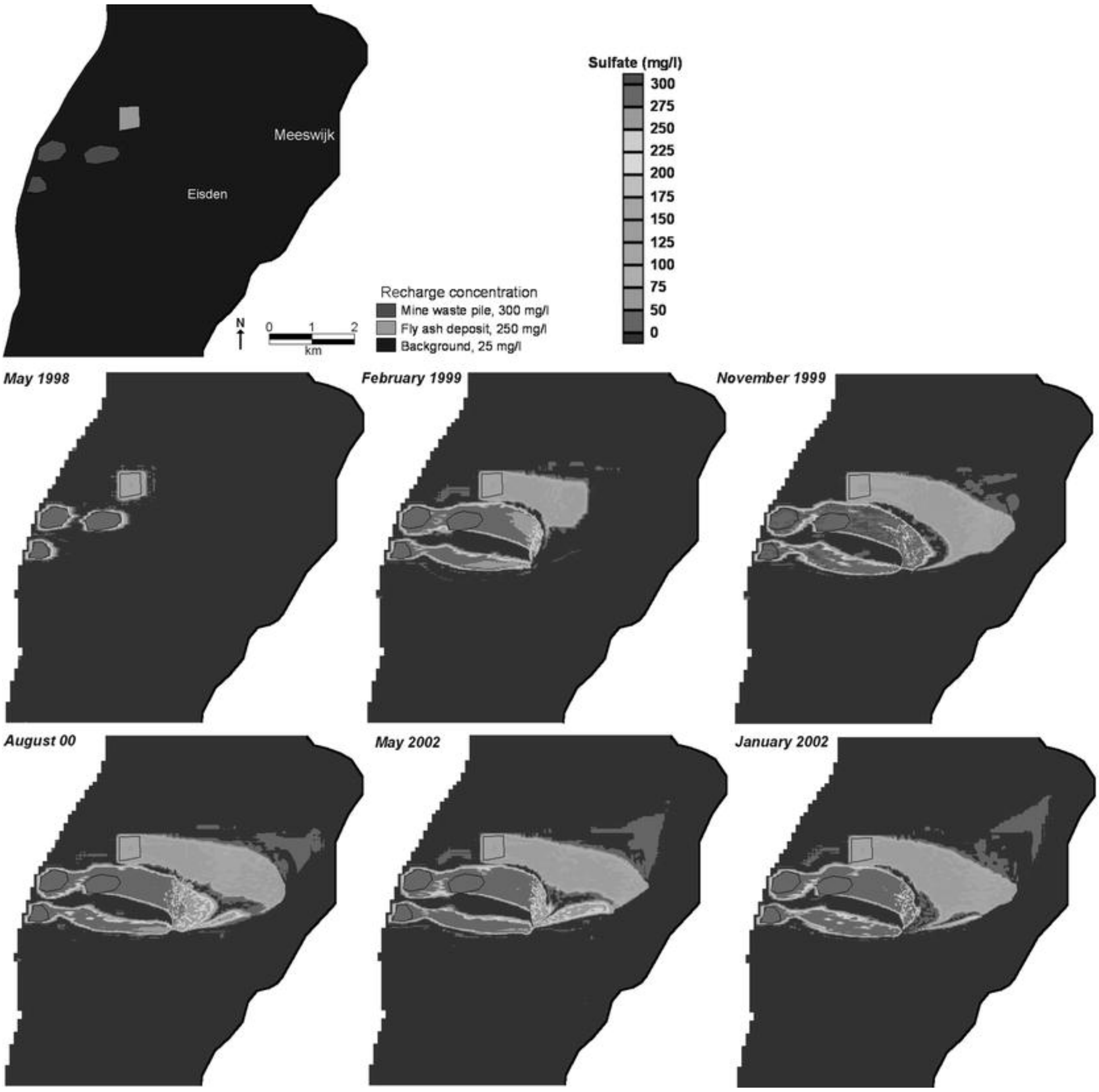

Fig. 7 Calculated sulphate concentrations in the region studied (mg/L) for different periods between May 1998 and January 2002

Again, the influence of the groundwater flow inversion during wet periods can be noticed. During dry periods, with overall flow towards the river, the extension of the contamination plumes generally increases towards the east. As soon as groundwater flow inversion occurs, the size of the plume decreases. Analogous to the nitrate simulation, a zone of increased sulphate concentrations can be observed between Eisden and Meeswijk.

Discussion

The calibrated groundwater model was able to give a good description of the groundwater flow-fields in the studied aquifer. In the future, the reliability of the model could be increased if more accurate values for groundwater abstractions and a more precise calculation of actual recharge were available.

To test the reliability of the simulations in predicting the spatial and temporal variations in solute concentrations in the region studied, a comparison is made between the simulated concentrations and the measured concentrations in the observation wells (Figs. 8 and 9).

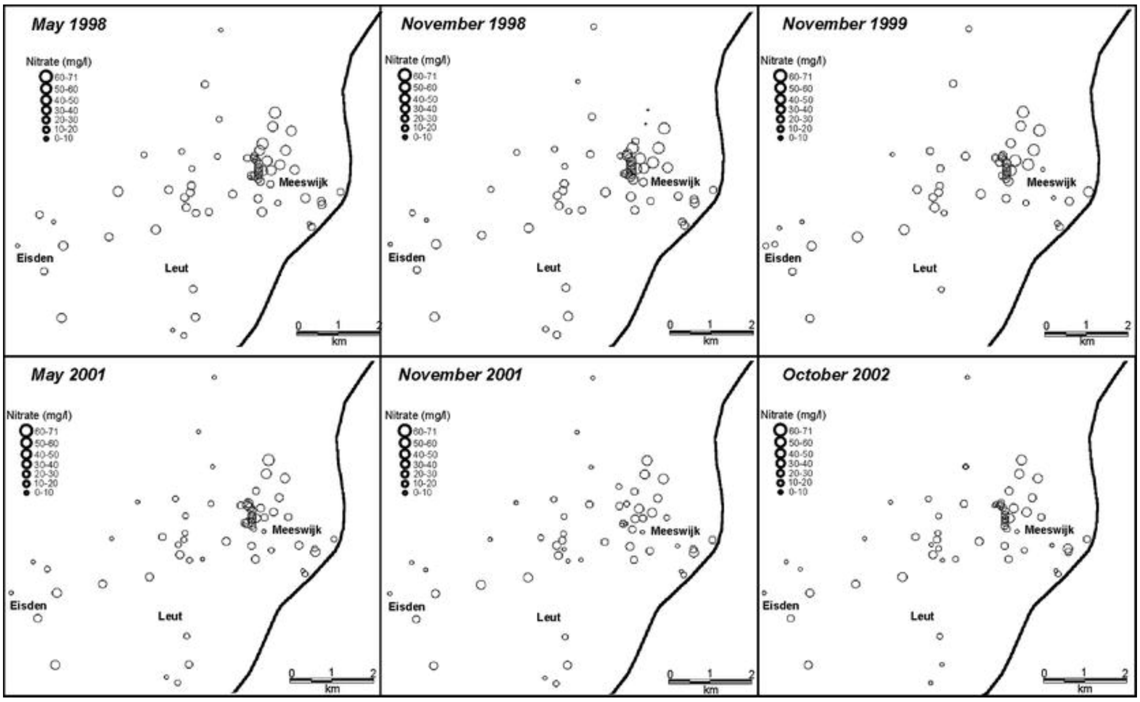

Fig. 8 Measured nitrate concentrations in the region studied (mg/L)

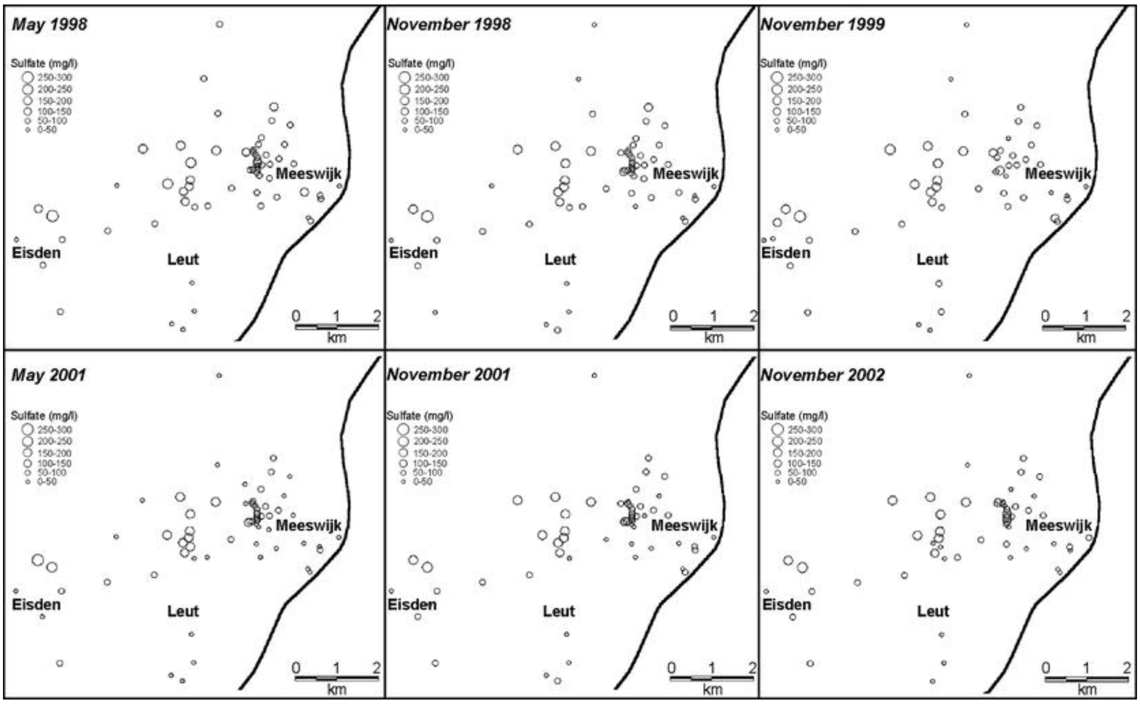

Fig. 9 Measured sulphate concentrations in the region studied (mg/L)

The measured concentration map for nitrates (Fig. 8) shows that nitrate concentrations in the northeastern zone of the Meeswijk well field are generally high. The observation wells between the Eisden and Meeswijk well fields have nitrate concentration slightly higher than the surrounding observation wells. The wells lie in the region where advection lines converge, as simulated by the transport model. However, the higher nitrate concentrations during wet periods are not only caused by the flow reversal in the aquifer. During the wet periods, recharge towards the aquifer increases and groundwater levels become higher, allowing more nitrate, stored in the partially saturated zone, to be directly mobilized in the saturated zone (Canter 1996, Oenema and others 1998; Neeteson 2000; Brouyère and others, unpublished data).

The measured concentration map for sulphate is depicted in Fig. 9. It can be seen that the highest concentrations were observed in an observation well located directly northeast of the Eisden well field. A possible explanation for these high concentrations is the outflanking movement of the northern sulphate plume, in reaching the Eisden pumping field from the northeast.

The observation well lying west of the Eisden well field shows remarkably low sulphate concentrations, despite being nearest to the waste piles. It can be seen that this well is located between the northern and the southern sulphate plume, according to the transport simulation. This well will not indicate high sulphate concentrations like the wells situated within the sulphate plumes. The higher sulphate concentrations in the observation wells located west of the Meeswijk well field can be explained by the sulphate plume induced by leaching water through the fly-ash deposits and then flowing into the aquifer towards the Meeswijk pumping field.

Conclusion

A groundwater flow model is developed for the period May 1998 to May 2002 and calibrated using monthly groundwater head measurements in 54 observation wells. Subsequently, transport simulations are carried out using modpath and mt3d. The transport simulations are limited to advection due to the lack of any data concerning dispersion coefficients. Other transport processes are considered negligible at this stage of the study.

The groundwater flow model shows an eastward groundwater flow towards the Meuse River with two cones of depression near the well fields of Eisden and Meeswijk. It is observed that during wet periods, the aquifer is fed by the river and consequently an inversion of groundwater flow occurs.

The transport simulations show the existence of zones where advection lines are converging from different directions (i.e. because of flow reversal during wet periods). The latter can be an explanation for the observation of higher nitrate concentrations in that area during wet periods.

From the simulation of advective transport of sulphate, the existence of two separate sulphate plumes can be deduced: the southern one is directly captured by the Eisden well field while the northern plume reaches the well field from the north, giving a possible explanation for the local high-sulphate concentrations in this area. Future work on nitrate and sulphate transport in the area of study will focus on incorporating transport by dispersion into the transport simulations.

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