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Groundwater flow and nitrate migration in a Dutch-Belgian chalk catchment; observed and future concentrations

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In the catchment of the Noor brook, by the Dutch/Belgian border, excess precipitation with high nitrate concentrations (80–120 mg·l^{-1}) recharges the Cretaceous multi-aquifer system under the plateau and foothill (agricultural area). The nitrates are transported through the aquifer (median NO_3^- > 50 mg·l^{-1} under the plateau) towards springs and wetlands of the Noorbeemden nature reserve. The major spring has a concentration of 70–80 mg·l^{-1}. The concentration in the Noor brook is somewhat lower (median 46 mg·l^{-1}) because of denitrification in the wetlands. The groundwater flow and transport model FLONET/TRANS was used to simulate the development of the nitrate distribution in the last 50 years. The general NO_3^- distribution in the northern part of the catchment could be reasonably well predicted, whereas the concentrations in the southern part were underestimated by about 20 mg·l^{-1}. The simulated future trend in the northern part of the catchment in the next 100 years shows that a reduction of nitrate leaching by 50% is required to level off the currently still increasing concentrations. A dramatic decrease of leaching by 75% is needed to achieve pre-1950's concentrations. This implies that the management plan of the Noorbeemden Nature Reserve, which is complied by the Nature Conservation Organisation, should not aim at botanical restoration in the first decades, even if severe restrictions in nitrogen application are imposed on the farmers now.

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Key words: the Netherlands and Belgium, Cretaceous chalk aquifer, groundwater flow, nitrate leaching, modelling , nitrate reduction policy.

INTRODUCTION

Since the 1950s, intensification of agriculture in the Netherlands has resulted in significant nitrate leaching to groundwater bodies. In the mid 1980s, nitrate concentrations were increasing in more than 75% of the well fields that extracted groundwater from unconfined aquifers. In some fields the drinking water standard of 50 mg NO_3^-·l^{-1} was already exceeded (RIVM, 1988). Nowadays, groundwater resources in extensive Dutch areas with fresh groundwater are at risk. Such conditions prevail, for example, in the southeastern part of the Netherlands in the area adjacent to the Belgian border, i.e. South-Limburg. In this region, about 20·10^6 m^{3}y^{-1} of groundwater with increased NO_3^- concentration is extracted from an unconfined Cretaceous chalk aquifer. In one well field the nitrate concentration has increased from 15 mg·l^{-1} in 1955 to 40 mg·l^{-1} in the late 1980s and is still rising (Juhász-Holterman *et al.*, 1989). A regional water treatment plant has been built there to remove nitrate from groundwater before it is distributed as drinking water. Increasing nitrate concentration also has adverse effects on wetlands. The botanical composition changes, often dramatically, decreasing the ecological value.

Many countries in western Europe, where chalk aquifers occur, have been facing similar problems. In some areas in France and England, for instance, nitrate concentration is over 50 mg·l^{-1} due to intensive agricultural activities. In areas of low infiltration it is likely to exceed 100 mg·l^{-1} eventually (Downing *et al.*, 1993; Hack-ten Broeke *et al.*, 1996). More than 2 million people in France and England have been supplied with groundwater containing more than 50 mg·l^{-1} over many years (Downing *et al.*, 1993). Policy makers on nature, environment and drinking water resources want to know when nitrate reduction policy measures will result in lower nitrate concentration in the groundwater and surface water systems of the chalk hydrogeological unit.

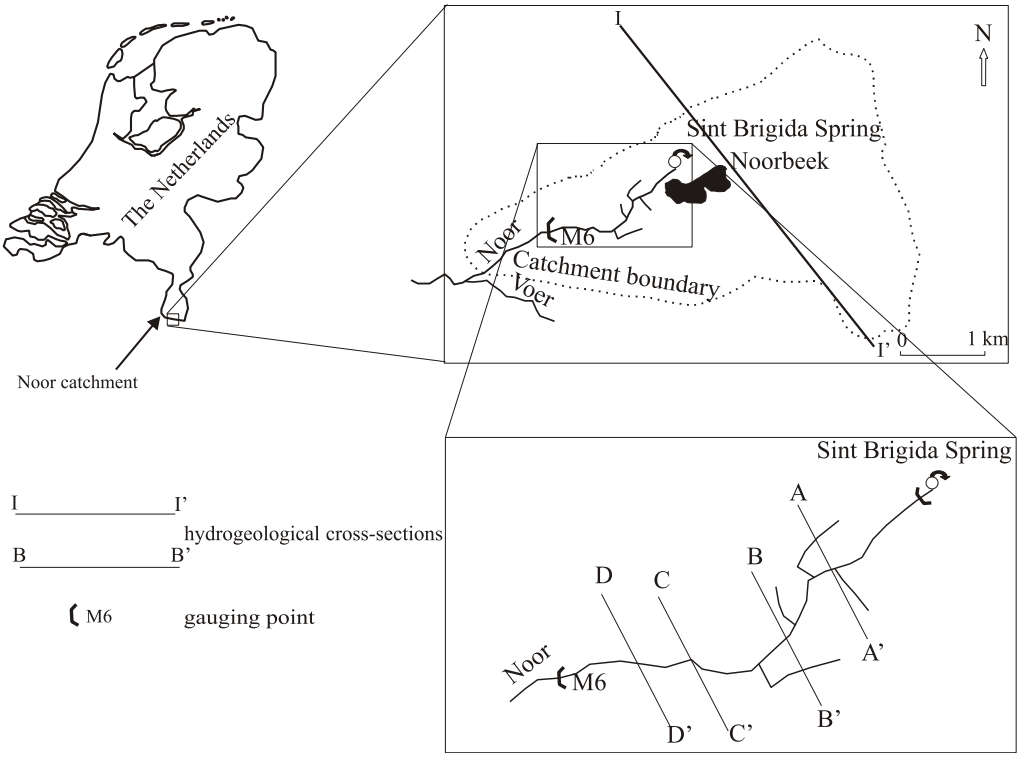

Fig. 1. Location of the Noor catchment, cross-sections and gauging station (van Lanen and Dijksma, 1999)

In recent years, several environmental laws and measures have been imposed on farmers in order to reduce nitrogen application and losses in the Netherlands. Research is being carried out to investigate the fate of nitrate and its pathways in agricultural catchments. In this framework, water flow and nitrate concentration in groundwater and surface water have been monitored in the Noor catchment since the early 1990s. This experimental catchment with deep water tables is representative of the chalk hydrogeological unit, which covers the southern part of Limburg and adjacent regions in Belgium. This hydrogeological unit is very different from the other mostly unconsolidated Quarternary hydrogeological units in the Netherlands. In the initial stage of the study in the Noor catchment, high nitrate concentrations were found in the chalk aquifer and extremely low concentrations in the riparian area. The main spring shows a clear upward trend of nitrate concentration, which has resulted in nitrate concentrations above the drinking water standard (van Lanen and Dijksma, 1999).

The objectives of this study are: (1) to describe the hydrogeological system of the Noor catchment, (2) to report the observed nitrate concentrations, (3) to develop a relatively simple groundwater flow and solute transport model to explain nitrate migration in the past and the present concentrations , and (4) to model future nitrate concentration trends in the Noor catchment as a result of the nitrate reduction policy.

DESCRIPTION OF THE NOOR CATCHMENT

HYDROGEOLOGICAL SYSTEM

The Noor catchment (1056 ha) is located in the south-east of the Netherlands and north-east of Belgium (Fig. 1). The elevation varies between 91 m a.s.l at the outlet and 240 m a.s.l. in the south-east. The Noor brook starts as the Sint Brigida spring (major spring) in Noorbeek (the Netherlands) at 138 m a.s.l. and discharges into the Voer in ‘s-Gravenvoeren (Belgium). The Voer is a small tributary of the river Meuse. The Noor brook has a length of 3 km and drains the southeastern part of a dissected Cretaceous chalk plateau. The valley has a steep northern slope (20%) and a gentler southern slope (7%). Permanent grassland and arable land (mainly maize) cover 62 and 35% of the catchment, respectively.

Consolidated Upper Carboniferous shales and sandstones, folded during the Variscan Orogeny, form the impermeable base of the major part of the basin at a depth of 50–150 m (Table 1, Fig. 2). In the downstream part in Belgium these shales and sandstones have been eroded and permeable Lower Carboniferous limestones occur, which implies that the impermeable base is at more than 800 m depth. The consolidated rocks are discordantly overlain by subhorizontal Upper Cretaceous deposits. These start with coastal environment sediments (Aken Formation) consisting mainly of well sorted sands. The sands only prevail in the southeastern part of the catchment due

Table 1

Hydrogeological characteristics of the Noor catchment (Nota *et al.*, 1988; van Lanen *et al.*, 1995; Felder, 1996; Kessels, 1997)

Time period	Formation	Description
Quaternary	Singraven (SF)	unconsolidated valley fillings (1–5 m); gravelly, fine-grained with thin peat layers; k_x=1.5 10^{-1} m·d^{-1}, k_y =7.5 10^{-2} m·d^{-1}, θ = 0.15
	Eindhoven (EF)	unconsolidated regolith or overburden (1–10 m); mixture of clay with flints and loess; unsaturated; k_x = 2.0 m·d^{-1}, k_y = 6 10^{-1} m d^{-1}
Upper Cretaceous	Gulpen (GF)	slightly consolidated, light coloured, fine-grained chalk with fissures (max. 40 m); upper part unsaturated; k_x = k_y = 2.0 m d^{-1}, θ = 0.10
	Vaals (VF)	saturated; glauconite-containing, unconsolidated clayey silts (max. 40 m) with thin fractured sandstone layers; silts: k_x = 6 10^{-1} m·d^{-1}, k_y = 3 10^{-1} m d^{-1}, θ = 0.15; sandstone layers*: k_x= k_y = 20 m d^{-1}, θ = 0.05
	Aken (AF)	saturated; light coloured fine-grained quartz sands (max. 35 m)*
Upper Carboniferous		consolidated shales and sandstones; impermeable base; k_x= k_y=1 10^{-6} m d^{-1}, θ = 0.01
Lower Carboniferous		karstified consolidated limestones*

k_x — horizontal permeability, k_y — vertical permeability, θ — effective porosity; *not included in the model

to erosion in the Cretaceous. Above the Carboniferous or Aken Formation a tidal flat unit occurs, which embodies a sedimentary succession consisting of clayey silts (matrix) with gully fillings (silt-fine sand) interbedded with thin layers (0.1–0.2 m) of consolidated and fractured sandstones (Vaals Formation). Chalk sediments (Gulpen Formation) overlie the tidal flat unit. The chalk is soft and poorly bedded and consists of nearly 90% calcite (e.g. Nota *et al.*, 1988; Felder, 1996). On top of the chalk a poorly sorted regolith (Eindhoven Formation) occurs. During the Pleistocene, the Noor brook deeply incised into these geological formations, eroding the chalk and the top part of the Vaals Formation. Therefore, in the valley centre, the chalk is missing. There, the Late Pleistocene and Holocene valley fillings (Singraven Formation) directly cover the Vaals Formation. Because of the deeply incised valley, deep groundwater levels prevail under the plateau. This implies that the regolith and the upper part of the chalk are unsaturated. A multi-aquifer system occurs in the lower part of the chalk and the Vaals Formation. The aquifer is unconfined, except for the valley centre where, due to the fine-grained nature of the valley filling, semi-confined conditions occur. At some locations in the centre of the valley, artesian conditions are even observed (van Lanen *et al.*, 1995; van Lanen and Dijksma, 1999).

Measurements show that the thickness of the unsaturated zone varies from less than 0.5 m in the valley up to 40 m under the plateau. The regolith readily allows infiltration of precipitation into the soil. Experimental research in the chalk region confirmed that surface runoff and interflow hardly occur. Analysis of the stream flow hydrograph at the gauging point M6 (Fig. 1) showed that more than 95% of the stream flow consists of groundwater discharge (Dijksma *et al.*, 1997). This is supported by the chemical composition, which indicates that almost all surface water samples belong to the $CaCO_3$ type, reflecting a strong interaction with the chalk sediments (Nota *et al.*, 1988; van Lanen *et al.*, 1995). This means that, except for the village of Noorbeek and the narrow wet valley, excess precipitation recharges the groundwater system. Deep water tables under the plateau show a delayed and smoothed response to excess precipitation. The annual precipitation varied between 616 and 987 mm in the period 1991–1996 and the calculated groundwater recharge varied between 130 and 410 mm. The daily stream flow at the gauging point M6 showed a variation between 0.25 and 0.90 mm·d^{-1} (Dijksma *et al.*, 1997).

MONITORING NETWORK

Since 1991 precipitation has been measured at three locations and groundwater levels at about 40 locations. Most piezometers are located in the cross-sections A–A' to D–D' (Fig. 1). In the wet valley, shallow observation wells (2–3 m) were installed to observe groundwater heads of the unconfined aquifer in the valley fillings and deep wells (5–8 m) to observe the heads of the semi-confined aquifer in the Vaals Formation. A few observation wells and dug wells are used to monitor the heads of the unconfined aquifer in the chalk under the plateau. Discharge of springs and surface streams have been monitored at 13 sites.

NITRATE

Since 1991, the chemical composition of the surface water system has been determined every month or two months at two springs, at seven tributaries of the Noor and at three locations in the Dutch part of the longitudinal profile of the Noor brook. Groundwater has been sampled four times a year and, since 1997, monthly in approximately 25 piezometers and wells. In the summer of 1995, numerous springs (n = 55) were sampled once at both sides of the brook (Kroon, 1995). Nitrate is one of the physical-chemical components which was measured. Two methods were used to determine nitrate concentrations, i.e. conventional ion chromatography in an analytical laboratory

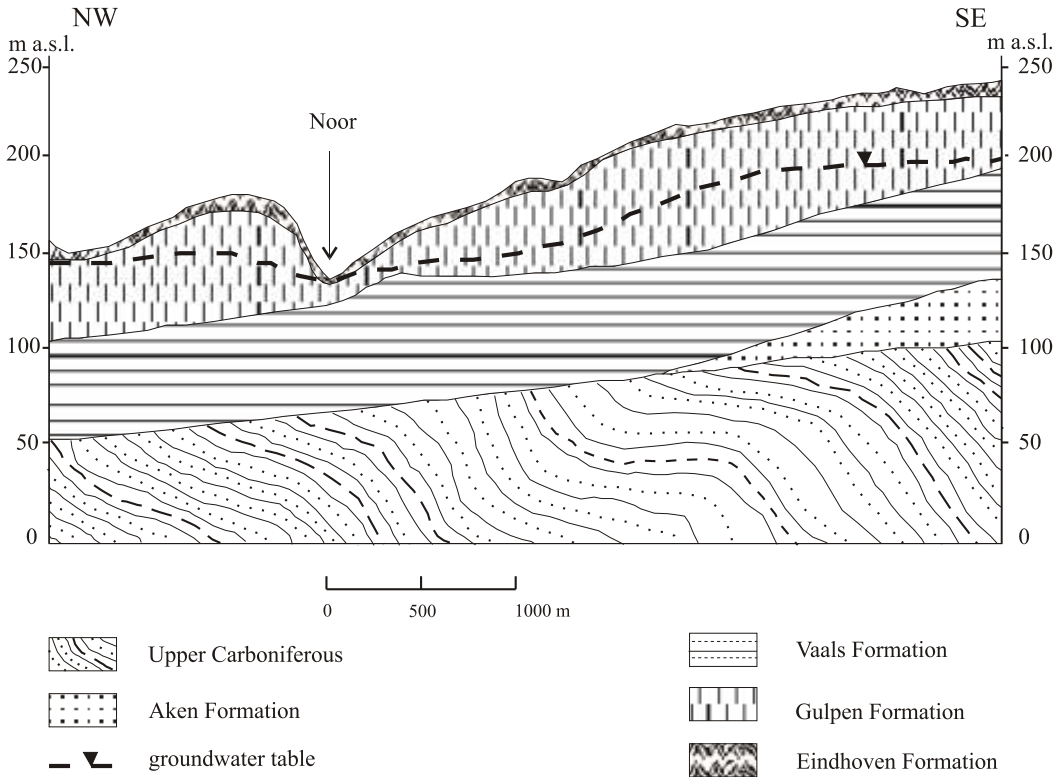

Fig. 2. Hydrogeological cross-section I–I'; cross-section line is indicated in Fig. 1 (van Lanen *et al.*, 1995)

and a set of integrated selective electrodes, which can be used either in a laboratory or in a field station. Comparison of both methods showed a good agreement between the NO_3^- concentrations of duplicates (van Lanen and Dijksma, 1999).

DESCRIPTION OF FLOW AND SOLUTE TRANSPORT MODEL FLONET/TRANS

The software code FLONET/TRANS (Guiger *et al.*, 1997) allows simulation of 2D steady-state groundwater flow and transient advective-dispersive contaminant transport in saturated porous media. The flow and transport equations used in the programme are given in Annex 1. The programme calculates hydraulic heads, streamfunctions, flow velocities, contaminant concentrations and breakthrough curves. FLONET/TRANS uses the finite element method to solve the flow and transport equations numerically. It allows heterogeneous and anisotropic domains, variable boundary conditions and source locations. The contaminant can be subject to linear retardation, diffusion and first-order decay.

The FLONET/TRANS steady-state groundwater flow model of the cross-section B–B' in the Noor catchment (Fig. 1) was developed by Kessels (1997). A groundwater contour map has been used to locate the cross-section perpendicular to the contour lines in the horizontal plane and to derive the position of the water divide. The modelled cross-section is 2700 m long and 105 m high, and it has a south-east-north-west orientation. The model mesh is subdivided into a number of cells defined by 45 columns and 43 rows. However, the calibration of the groundwater flow model has proved that the northwestern groundwater divide for the cross-section B–B' does not correspond exactly with the surface water divide. Due to this fact the simulated groundwater divide has been moved 450 m from the surface water divide towards north-west.

The boundaries of the groundwater flow model of the cross-section coincide with the natural boundaries of the multi-aquifer system, therefore zero-flux boundaries have been assigned to the water divide in the north-west and southeast (Fig. 3) and to the impermeable base of the aquifer (top of the Upper Carboniferous). A specified-flux boundary (groundwater recharge) has been used as an upper boundary condition. Transient groundwater recharge has been calculated separately for every type of land use and soil for the period 1960–1995. On the basis of the computed time — series of annual groundwater recharge, three different types of steady-state groundwater conditions have been defined: namely dry, average and wet. Groundwater recharge has been used as an upper boundary condition for most of the cross-section. In the centre of the valley, however, a constant head has been used as an upper boundary condition to simulate drainage of groundwater by springs, wetlands and the Noor brook. The constant head has been derived from the elevation of the springs, the water-table of the wetlands (near the surface) and the surface water level of the Noor brook. The drainage resistance of the springs, ditches in the wetland and the Noor brook has been defined by introducing a layer of cells with a low vertical conductivity.

The real hydrogeological conditions have been simplified (Fig. 3) and every hydrogeological unit has been assumed to be homogeneous. The hydrogeological properties of the units, such

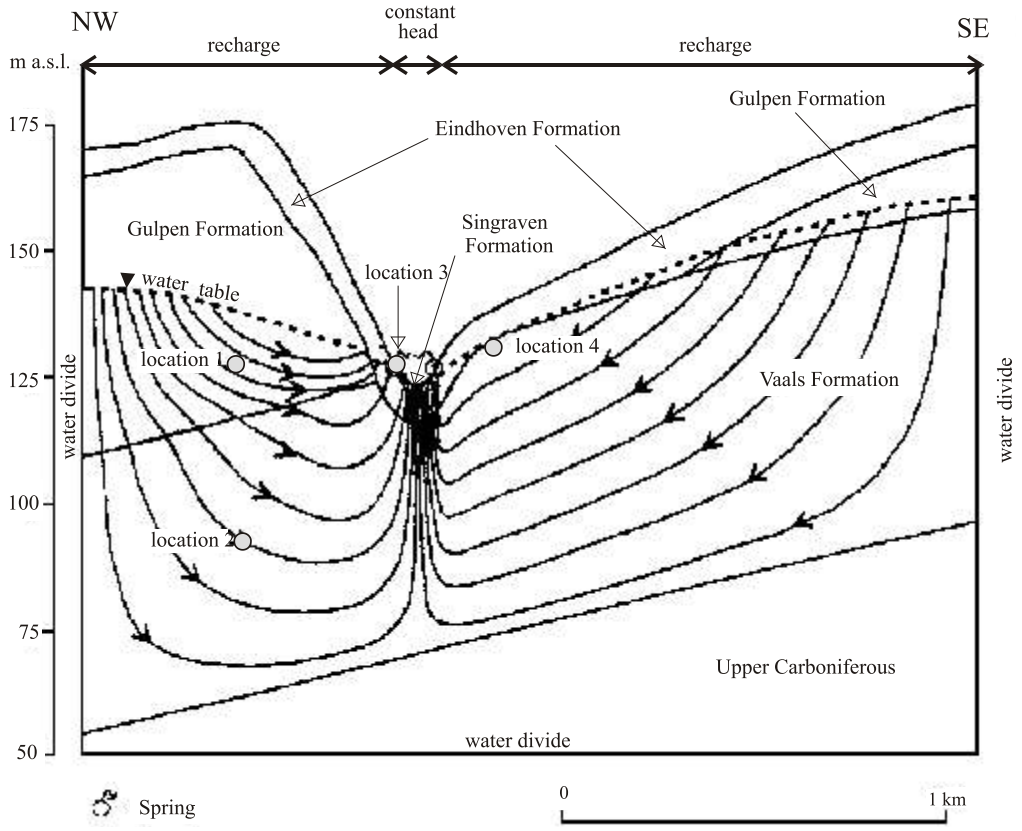

Fig. 3. Hydrogeological cross-section B–B' showing the simulated groundwater flow paths, schematized hydrogeological units (Eindhoven, Singraven, Gulpen and Vaals Formations and Upper Carboniferous), monitoring points (1 — northern plateau shallow location, 2 — northern plateau deep location, 3 — northern foothill, 4 — southern foothill) and boundary conditions

as hydraulic conductivity in the *x*-, *y*-directions and effective porosity, were initially derived from the calibrated MODFLOW model (van Lanen *et al.*, 1995). In the next stage hydrogeological properties were calibrated separately for every hydrogeological unit using the FLONET/TRANS model (Kessels, 1997). Those values are presented in Table 1. Kessels (1997) obtained a reasonable agreement between the simulated and the observed groundwater heads of the unconfined aquifer (valley fillings) and the semi-confined aquifer (Vaals Formation).

The groundwater flow model of cross-section B–B', described above, was used to develop a model to simulate transient advective-dispersive nitrate transport. First a model for the period 1950–1997 was developed to simulate historical migration of the nitrate plume and the current distribution of the nitrate concentration. Second, future nitrate migration patterns were simulated for the period 1998–2098. Three scenarios were defined, which represent different nitrate reduction policies.

The boundary conditions of the transport model fully match the boundary conditions of the flow model. Zero-concentration gradient boundaries coincide with the zero-flux boundaries of the flow model (impermeable base at the bottom and water divide in the north-west and south-east). The only place through which nitrate enters the multi-aquifer system model is the upper boundary (groundwater table), where a dispersive-flux boundary was defined. The historical nitrate input has been calculated for different land use types in South-Limburg for the period 1950–1995 (Schot *et al.*, 1996). They estimated the nitrate concentration of water leaching from the root zone in 1950 at 20 mg $NO_3^-\cdot l^{-1}$, which was used as an initial concentration in the model. This initial concentration is already somewhat higher than the lowest concentration reported in the base flow of the chalk region (about 12 mg $NO_3^-\cdot l^{-1}$; Nota *et al.*, 1988). The average NO_3^- concentrations for the different land use and soil types for the period 1950–1997 are given in Table 2. These concentrations were derived from Schot *et al.* (1996), and they were used as the upper boundary condition for the 1950–1997 model. The average NO_3^- concentration is significantly higher in the northern part of the catchment (112 mg$\cdot l^{-1}$) than in the southern part (82 mg$\cdot l^{-1}$). This is because of a higher percentage of grassland in the former part. Nitrate input is the lowest (55 mg$\cdot l^{-1}$) in the wet forested valley.

Transport parameters, i.e. contaminant decay rate, diffusion coefficient, source decay rate, longitudinal and transverse dispersivity of nitrate have been defined according to Domenico and Schwartz (1990) and Freeze and Cherry (1979). They presumed that NO_3^- is a stable form of dissolved nitrogen,

Table 2

The average NO_3^- input for different types of land use and soils assigned to the dispersive-flux boundary of the models (derived from Schot *et al.*, 1996)

Distance in the *x*-direction from N to S [m]	Land use and soil type	Computed concentration of NO_3^- leaching from the root zone $[mg \cdot l^{-1}]$				
		1950–1997 model		1998–2098 models reduction by		
		nitrate input	weighed average	25%	50%	75%
–450–500	grasskand	116	112	87	58	29
500–569	forest	55		41	27	14
569	Noor brook					
569–705	forest	55	82	41	27	14
705–780	grassland	116		87	58	29
780–2200	tuber corps on regolith	83		63	42	21

undergoing only minor transformation processes and migrating together with groundwater flow, with no or little retardation and decay. Because of these characteristics the relatively simple FLONET/TRANS model can be applied to simulate migration of the nitrate plume in the cross-section B–B'. After sensitivity analysis and the comparison of simulated and measured nitrate concentrations, modelling showed that no calibration was required of the NO_3 transport parameters, as derived from the literature. The simple model cannot be used to simulate denitrification processes. The simulated nitrate concentration distribution underneath the valley centre, where denitrification prevails, is therefore not reliable and it is not addressed in this study.

FLONET/TRANS does not account for NO_3^- transport in the thick unsaturated zone. The travel time of a water particle in this zone is assessed to be 5–10 years (Juhász-Holterman *et al.*,

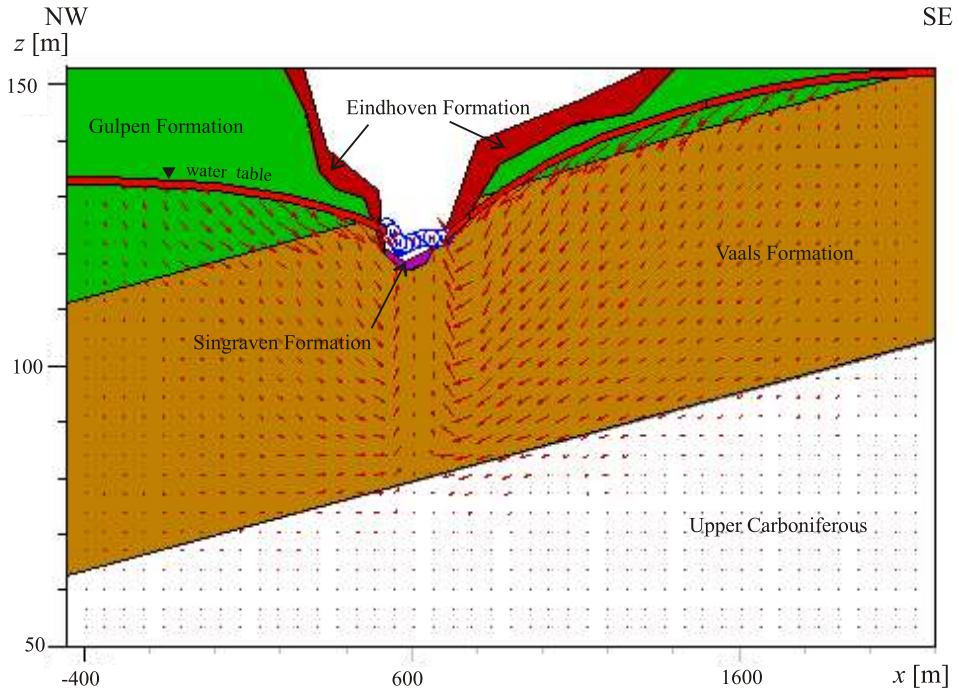

Fig. 4. Simulated groundwater flow velocities in the cross-section B–B' for average groundwater flow conditions

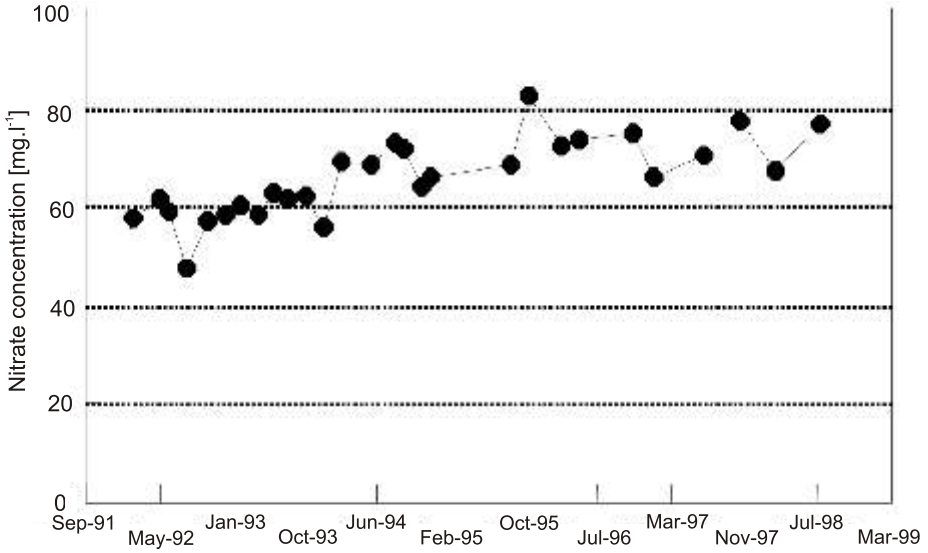

Fig. 5. Measured nitrate concentrations of the Sint Brigida spring for the period September 1991–March 1999

1989). The nitrate ions will stay at least this time in the unsaturated zone. Because FLONET/TRANS only deals with the saturated zone, the simulated travel time and response time should be corrected for the travel time in the unsaturated zone.

RESULTS

GROUNDWATER FLOW

The simulated streamlines (Fig. 3) start at the water table under the plateau and foothill (recharge area) and finally end in the wet valley (discharge area). A small part of the groundwater recharge in the catchment only flows through the chalk (foothill north of the Noor brook) and does not flow through the Vaals Formation. The majority of the recharge flows through the Gulpen and Vaals Formations. The streamlines have a strong vertical component close to the water divides and beneath the valley centre. In most of the catchment the groundwater flows downward or approximately horizontally. In a narrow zone along the brook (riparian area) upward seepage occurs. Sensitivity analysis showed that the groundwater flow pattern does not substantially change for dry and wet conditions (years with a low and high recharge, respectively).

The simulated groundwater flow velocities are highest below the valley centre and in the chalk north of the Noor brook. Low velocities occur at the bottom of the aquifer, especially near the water divides (Fig. 4).

The simulated travel times in the saturated zone are 30 and 50 years for water particles following the longest streamline in the north and south, respectively. The simulated streamlines, flow velocities and associated travel times are the key factors for the prediction of the historical and future nitrate distribution, as will be shown below.

CURRENT NITRATE CONCENTRATIONS

MEASUREMENTS

The nitrate concentration of the Sint Brigida spring (Fig. 5) characterises the groundwater contamination in the Noor catchment. Since 1991 the concentration has increased from about 60 to 70–80 mg·l^{-1}. The concentration is well above the drinking water standard (50 mg·l^{-1}). The nitrate concentration of the spring was also monitored in the early 1980s, when it increased from about 40 to 50–60 mg·l^{-1}.

Nitrate concentrations of groundwater samples show a general upward trend (Kłonowski, 1997; van Lanen and Dijksma, 1999). The highest concentrations occur in the groundwater under the plateau and foothill. The median concentration is above 50 mg·l^{-1} and the highest concentrations are well above 100 mg·l^{-1}. In the wet valley, extremely low concentrations are

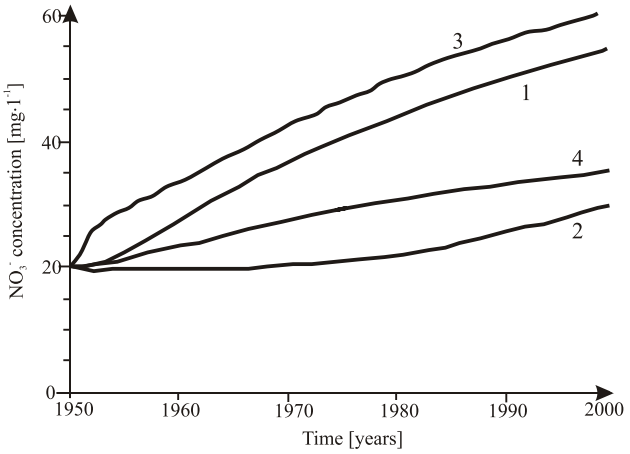

Fig. 6. Simulated nitrate concentrations at the locations 1, 2, 3 and 4 for the period 1950–1997 under average groundwater recharge conditions; locations are indicated in Fig. 3

Table 3

Statistical moments of nitrate concentrations (mg l^{-1}) in springs and surface water (after Kroon, 1995; Kordík, 1998)

Samples	Median	Mean	Standard deviation	Minimum	Maximum	Number of samples
Noor brook*	45.7	45.5	7.7	26.3	60.6	102
Tributaries north*	46.6	44.7	8.6	31.9	70.7	68
Tributaries south*	37.4	38.3	5.7	24.5	48.5	57
Springs*, **	64.0	65.7	10.3	45.2	82.5	51
Spring north 1995	64.4	59.2	15.9	26.6	87.8	33
Spring south 1995	55.4	54.2	22.0	10.9	113.6	22

* — period 1992–1997; ** — two springs north of the Noor brook

measured (median 2 mg·l^{-1}) due to denitrification. The variation is low there.

The NO_3^- concentrations of springs and surface water (Noor brook and tributaries) are shown in Table 3. The median NO_3^- concentration of the springs is higher than the tributaries and the Noor brook itself. The springs only receive nitrate-rich groundwater from the plateau and foothill, whereas the tributaries and the main brook also receive nitrate-poor groundwater from the wetlands in the valley centre where denitrification occurs. Furthermore, the median nitrate concentration in the northern springs and the tributaries is substantially higher than in the southern ones.

Except for the valley, NO_3^- concentrations of the groundwater samples show a higher spatial and temporal variability than the samples from springs and surface water (Noor brook and tributaries). The reason for this is that spring and surface water integrate the chemical composition of groundwater from a particular area (catchment area), whereas groundwater samples reflect the characteristic of particular point in a 3D flow domain. In the modelling study emphasis has been put on exploration of general nitrate patterns and not on the explanation of concentrations measured in the aquifer at a particular point.

SIMULATIONS

Nitrate migration in the past has been simulated on the cross-section B–B' (Figs. 1 and 3) for the period 1950–1997. The transient simulation of nitrate transport is based upon steady-state groundwater flow conditions. In the context of model sensitivity analysis, steady-state simulations of groundwater flow have been performed for dry, average and wet groundwater recharge conditions. Analysis of flow lines and flow velocities showed that a steady-state groundwater flow model with average recharge as an upper boundary condition is a reasonable basis for the transient simulation of nitrate distribution.

Breakthrough curves (NO_3^- concentration versus time) for the selected locations within the model domain (Fig. 3) have been calculated in order to illustrate development of the nitrate plume in the period 1950–1997 (Fig. 6). The quickest response to the contamination occurs at the foothill (location 3), where groundwater follows short and shallow flow paths with relatively high velocities in the chalk. At this location nitrate concentration rapidly increases at first, then it grows steadily to a value of about 60 mg·l^{-1} after about 50 years. Compared to location 3, the increase of the nitrate concentration under the northern plateau (location 1) is slightly delayed but is also gradually increasing. The final concentration is above 50 mg·l^{-1}. The nitrate concentration at the southern foothill (location 4) is much lower than at the northern foothill during the whole simulation time. The final concentration for this location is around 35 mg·l^{-1}. Deeper parts of the Creataceous aquifer under the northern plateau (location 2) are characterised by a very slow response to the contamination and a low but still increasing concentration of about 30 mg·l^{-1} after about 50 years.

The final NO_3^- distribution (after 48 years, present situation), under average groundwater recharge conditions, is shown in Figure 7 for the cross-section B–B'. Generally, the NO_3^- concentration is much higher in the north than in the south. The highest values, over 50 mg·l^{-1}, occur at the northern foothill, while the southern foothill is characterised by concentrations over 35 mg·l^{-1}. The nitrate concentration in the uppermost part of the multi-aquifer system (Gulpen Formation) under the plateau is also very high and varies between 40 and 60 mg·l^{-1} in the north and 30–38 mg·l^{-1} in the south. Simulation of the nitrate plume migration shows a zone of stagnant groundwater at the bottom of the aquifer, which does not take any active role in groundwater flow and is characterised by lower nitrate concentrations (Fig. 7). At the bottom of the aquifer (Vaals Formation) the concentration reaches 20–25 mg·l^{-1}.

The sensitivity of the nitrate transport model has been tested under different groundwater recharge conditions: dry, average and wet. The analysis shows that the nitrate migration is about two times faster under wet conditions than under dry conditions. For example, the drinking water standard (maximum allowable concentration) of 50 mg·l^{-1} is reached at the northern foothill after 50, 33 and 25 years for dry, average and wet conditions, respectively (Kordík, 1998). The sensitivity analysis shows that the size of the stagnant zone depends on the recharge. In reality, the recharge varies from year to year, which implies that the stagnant zone is smaller than shown in Figure 7.

As mentioned earlier it was not the objective of this modelling to predict the nitrate concentrations at particular points (location, depth and time), but emphasis was put on general, more integrated nitrate concentrations. The simulated NO_3^- concentrations at the northern foothill (55–60 mg·l^{-1}) (Figs. 6 and 7)

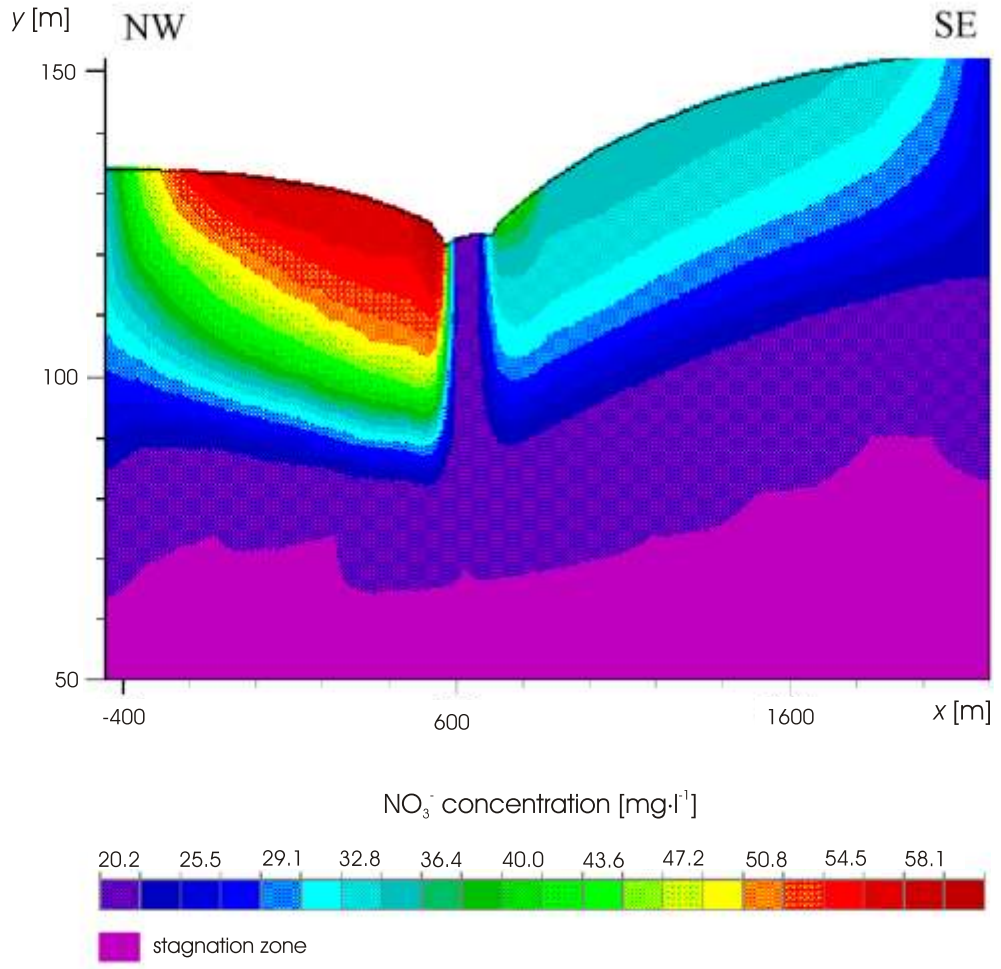

Fig. 7. Simulated distribution of nitrate concentrations in the cross-section B–B' after about 48 years (present situation) for average groundwater recharge conditions

correspond well with the recently (1995) measured concentrations in springs at the northern foothill (min 26.6, max 87.8 and median 64.4 mg·l^{-1}, Table 3). In the southern part of the catchment the measured concentration in the springs (min 10.9, max 113.6 and median 55.4 mg·l^{-1}) is underestimated by about 20 mg·l^{-1} (Figs. 6 and 7). This underestimation of the simulated concentrations might be caused by different factors, among others: lack of detailed knowledge of geological and hydrogeological settings and changes in land use in the past. Because of the underestimation of the NO_3^- concentrations, the southern part of the catchment was not further considered in the analysis of the future trends. This is allowed because the simulation shows that the southern and northern part of the catchment behave relatively independently (a vertical zero-flux plane occurs below the valley centre), which permits an evaluation of the NO_3 concentrations in the northern part only.

FUTURE TRENDS OF NITRATE CONCENTRATION

A model for the prediction of future nitrate concentrations has been developed upon the basis of the 1950–1997 flow model. The simulation of future trends starts in 1998 and terminates in 2098. The model presumes no changes in flow and transport parameters as well as in groundwater recharge conditions in the next century. The average recharge conditions have been assumed to give a good basis for the simulation of the NO_3^- distribution.

Three scenarios have been defined to study future trends of nitrate migration in the northern part of the catchment. The three scenarios reflect reduction of the NO_3^- input concentration at the dispersive-flux boundary by 25, 50 and 75%, respectively, compared to the 1950–1997 period model (Table 2). The initial concentrations for the scenarios (situation 1998) have been assigned by means of six polygons, characterised by average NO_3^- concentrations which have been derived from the final output of the 1950–1997 model (Fig. 7). Unfortunately, a more precise definition of the initial concentration (e.g. assigning a value to each element) is impossible with the current version of the FLONET/TRANS model. This implies that no smooth development of the NO_3^- concentrations from the present to the future situation can be simulated.

In the initial stage of the simulation of future trends, from 0 to about 25 years, distinct fluctuations of NO_3^- concentration can be observed (fine dashed line in Fig. 8) which are due to the

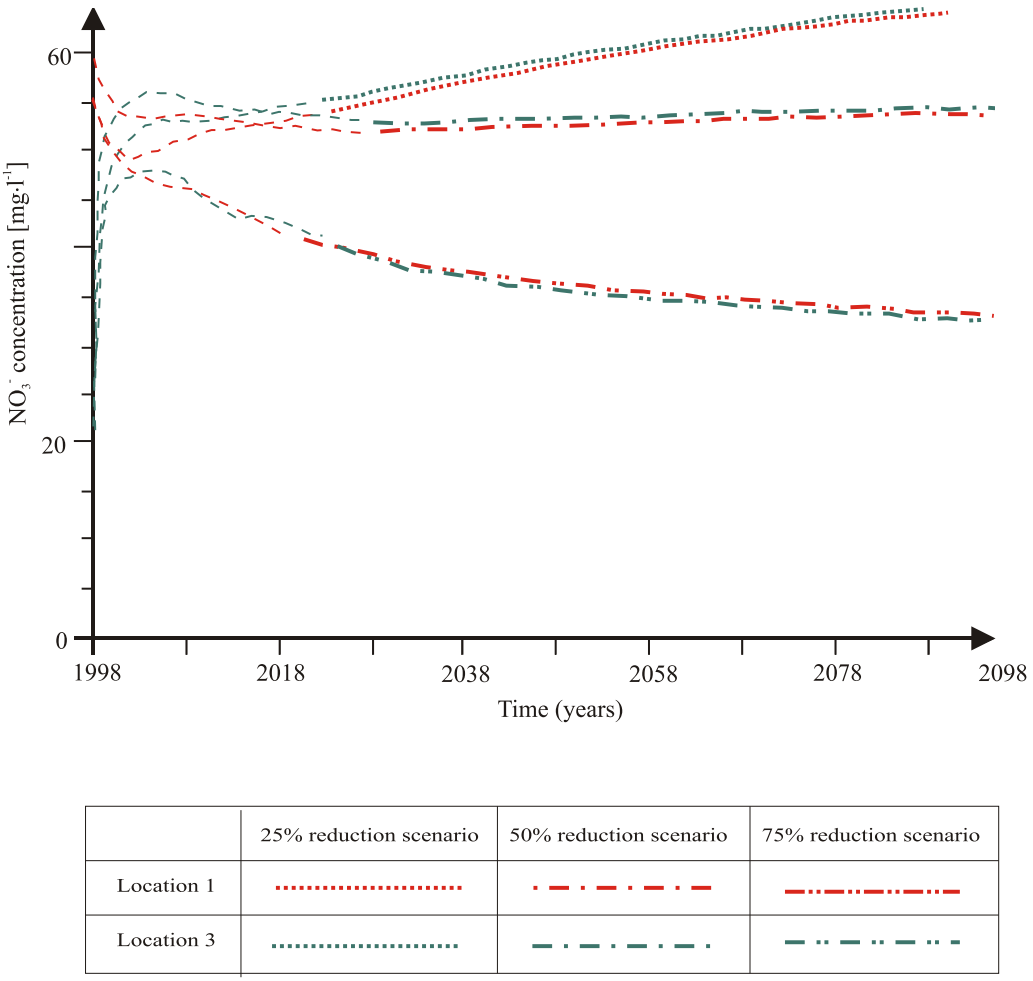

Fig. 8. Simulated nitrate concentrations for the period 1998–2098 for average groundwater recharge conditions at locations 1 and 3 in the aquifer for the 25, 50 and 75% nitrate reduction scenario

formulation of the initial NO_3^- concentration. Nevertheless, average nitrate concentrations at the starting point of the 1998–2098 models, neglecting the above mentioned fluctuation, approach the final nitrate concentration of the 1950–1997 model (Figs. 6 and 7). After a period of 25 years the model is adapted to the crude definition of the initial concentration and stabilizes. The initial fluctuation is mainly caused by the earlier mentioned design of the polygons and the limited number of the polygons allowed. Therefore, only the trend in nitrate concentration from about 25 years onwards will be discussed.

Reduction of nitrate input by 25% does not cause any decline of the calculated nitrate concentration (Fig. 8). The breakthrough curves for the northern plateau (location 1) and northern foothill (location 3) are very similar and show a gradual increase. For the whole simulation period they exceed the drinking water standard of 50 mg l^{-1} and reach concentrations of above 65 mg l^{-1}, after about 98 years.

Reduction of nitrate input by 50% leads to a stabilisation of the calculated nitrate concentration in the northern part of the catchment (Fig. 8). The nitrate concentrations for the plateau and foothill are almost constant. Nevertheless, a very small upward trend can still be recognised. The final concentration at both locations is about 50 mg l^{-1}.

Reduction of nitrate input by 75% causes a decrease of the calculated nitrate concentrations north of the brook (Fig. 8). The final NO_3^- concentration for the locations 1 and 3 is about 30 mg l^{-1} after 98 years.

CONCLUSIONS AND DISCUSSION

The hydrogeological system of the Noor catchment with deep water levels in the unconfined Cretaceous aquifer leads to hardly any generation of surface runoff or interflow which quickly feed the Noor brook (<5% of the streamflow). The groundwater recharge predominantly follows a long flow path through the chalk and underlying silts and sandstone layers with estimated travel times up to 50 years. Only in the vicinity of the wet valley, short flow paths prevail with travel times of a few years. Eventually, most of the groundwater is discharged in the wet valley as springs. The remaining groundwater flows to seepage areas and directly to the Noor brook.

In the last 20 years the NO_3^- concentration of the major spring, i.e. the Sint Brigida spring, has nearly doubled from about 40 to 70–80 mg·l^{-1}. The increase is the response to leaching of nitrates from agricultural areas. The leaching for grassland increased from about 20 to more than 100 mg NO_3^-·l^{-1} in the last fifty years. In extensive areas in the catchment (plateau and foothills) the majority of the groundwater samples has NO_3^- concentrations above the drinking water standard of 50 mg NO_3^-·l^{-1}. Low nitrate concentrations (< 10 mg NO_3^-·l^{-1}) occur only in the wet valley. This is, most probably, due to denitrification processes caused by the relatively high content of organic matter of the valley fillings. Most of the springs also have nitrate concentrations (median: 55–65 mg NO_3^-·l^{-1}) above the drinking water standard because they drain nitrate-rich groundwater. The tributaries and the Noor brook collect, other than spring water, water from seepage areas and groundwater, which might have undergone denitrification. Therefore the median NO_3^- concentration of these surface water streams is lower than that of the springs (between 37 and 47 mg·l^{-1}). The northern springs and tributaries have a higher nitrate concentration than the southern ones (median about 10 mg·l^{-1} higher). The larger grassland area in the north with higher nitrate load is the main reason for this.

The relatively simple 1950–1997 model based upon the FLONET/TRANS code simulates the general nitrate concentration patterns for the northern part of the Noor catchment reasonably well. The model reveals that the higher groundwater recharge and the lower nitrate input south of the Noor brook explain the lower nitrate concentration of the springs and tributaries there. However, the model underestimates the measured NO_3^- concentrations in the southern part of the catchment by about 20 mg·l^{-1}. The complex denitrification process could not be included in the model. This is the reason that the simulated concentrations in the wet valley are unrealistically high. The model also shows that the upper part of the Cretaceous aquifer was polluted very soon after the intensification of agriculture in the 1950s, whereas the lower part responded much later. In 1997 the nitrate concentration still increased in both parts of the aquifer. The northern springs show a quick response on the groundwater contamination because they are mainly fed by short flow paths through the chalk.

The simulation of future trends of nitrate concentrations in the northern part of the catchment, as a response to nitrogen reduction measures in agriculture, shows that a reduction of the nitrate leachate by 25% still results in an increase of the nitrate concentrations in groundwater and surface water in the next 21st. In this 25% reduction scenario the concentration in the leachate is still higher than the current concentrations in the aquifer. In the 50% reduction scenario the current nitrate concentrations level stabilises, but no significant decline occurs in the northern part of the Noor catchment. A dramatic decrease of the nitrate input, i.e. the 75% reduction scenario, results in slowly decreasing NO_3^- concentrations to a value of about 30 mg NO_3^-·l^{-1} (mean NO_3^- input). This value will be reached after decades because of the long travel times in the Cretaceous multi-aquifer system with deep water levels.

The monitoring and modelling work in the catchment of the Noor brook points out that even if agricultural practices change substantially, the groundwater resources and nature reserves in this chalk unit will suffer from too high nitrate concentrations for many years. Nature management authorities must realise that a restoration of the botanical composition, associated with groundwater rich in $CaCO_3$ and poor in nutrients, is not to be expected within the first decades. Irreversible processes might already have taken place.

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ANNEX 1

FLONET/TRANS is a scientific software package developed at the Waterloo University in Canada. It has been designed to approximate natural, three dimensional (3*D*) groundwater systems in either the two-dimensional (2*D*) area plane or vertical cross-section. It suits especially all hydrogeological domains with significant vertical variation in physical properties or with significant vertical flow gradients. The programme assumes: steady 2*D* groundwater flow, fully saturated conditions, constant density pore water, contaminant in the dissolved phase, contaminant dilution with a temperature equal to that of the pore water.

This software is a combination of two numerical finite element models: a steady-state saturated groundwater flow model and a transient advective-dispersive contaminant transport model. The model solves the flow problem using the dual formulation (Frind and Matanga, 1985) which formulates the flow equations in terms of hydraulic potential and stream function. In advective-dispersive contaminant transport FLONET-/TRANS solves the 2*D* equation with linear retardation and first-order decay. Groundwater flow velocities are derived from the stream function solution of the flow problem (Guiger *et al.*, 1996).

Governing equations for groundwater flow

The governing equation for 2*D* steady-state saturated groundwater flow can be written in the dual formulation as (Guiger *et al.*, 1996):

$$\frac{\partial}{\partial x}\left(K_{xx}\frac{\partial \varphi}{\partial x}\right)+\frac{\partial}{\partial y}\left(K_{yy}\frac{\partial \varphi}{\partial \psi}\right)=0 \qquad [1]$$

and

$$\frac{\partial}{\partial x}\left(\frac{1}{K_{yy}}\frac{\partial \psi}{\partial x}\right)+\frac{\partial}{\partial y}\left(\frac{1}{K_{xx}}\frac{\partial \psi}{\partial y}\right)=0 \qquad [2]$$

where: x, y — spatial co-ordinates [L]; K_{xx} — hydraulic conductivity in the x direction [L/T]; K_{yy} — hydraulic conductivity in the y direction [L/T]; ϕ — hydraulic head [L]; ψ — stream function [L^2/T].

Groundwater flow velocities

The relationship between the properties of the porous medium, the hydraulic gradient and the groundwater flux q(L/T), as given by Darcy's equation and written in terms of the stream function is:

$$q_x=\frac{\partial \psi}{\partial y};\ q_y=\frac{\partial \psi}{\partial x} \qquad [3]$$

The components of the average linear groundwater velocity are given by:

$$v_x=\frac{q_x}{\theta};\ v_y=\frac{q_y}{\theta} \qquad [4]$$

where: θ — effective porosity [-]; q — groundwater flux [L/T].

Numerical implementation

FLONET/TRANS uses the Galerkin finite element approach to solve equations [1] and [2]. Therefore the flow domain is covered with a grid consisting of elements and nodes. The model allows grid elements to deform, thereby providing a more accurate representation of the water table geometry and internal hydrostratigraphic layers. It also allows the principal directions of the hydraulic conductivity to vary in space. In the solution [1], an iterative approach is used where the domain is allowed to deform vertically to conform to the steady-state water table position in unconfined aquifers. In these cases, the hydraulic head solution algorithm will iterate until a user-defined convergence criterion (*crit*) is satisfied. The condition to be satisfied at the water table is (Guiger *et al.*, 1996):

$$\varphi_w(x, y) = y_w(x, y) \pm crit \qquad [5]$$

where: φ_w — head at the water table [L]; y_w — water table elevation [L].

Governing equations for advective-dispersive contaminant transport

The governing equations for 2D advective-dispersive mass transport of a dilute species within a porous medium can be written as (Bear, 1972; Spitz and Moreno, 1996):

$$\frac{\partial}{\partial x_i}\left[\frac{D_{ij}}{R}\frac{\partial c}{\partial x_j}\right] - \frac{\partial}{\partial x_i}\left[\frac{v_i}{R}c\right] - \lambda c = \frac{\partial c}{\partial t} \qquad [6]$$

where: x, y — spatial co-ordinates [L]; v_i — average linear flow velocity [L/T]; D_{ij} — hydrodynamic dispersion coefficient [L^2/T]; R — linear retardation factor [-]; λ — linear decay rate [T^{-1}]; t — time [T]; c — concentration [M/L^3].

The form of the dispersion is given by Frind and Hokkanen (1987) and is dependent on the average linear flow velocities, the longitudinal and transverse dispersivities (α_L and α_T) and on the effective molecular diffusion coefficient D [L^2/T] according to:

$$D_{xx} = \alpha_L \frac{v_x^2}{\bar{v}} + \alpha_T \frac{v_y^2}{\bar{v}} + D \qquad [7]$$

$$D_{yy} = \alpha_T \frac{v_x^2}{\bar{v}} + \alpha_L \frac{v_y^2}{\bar{v}} = D \qquad [8]$$

and

$$D_{xy} = D_{yx} = (\alpha_L - \alpha_T)\frac{v_x v_y}{\bar{v}} \qquad [9]$$

where: $\bar{v} = \sqrt{v_x^2 + v_y^2}$. [10]

The velocity in [6] is obtained from equation [4] using the steady-state stream function solution of the dual formulation.

Adsorption of a contaminant onto aquifer solids results in a retardation effect while decay processes decrease the mass. FLONET/TRANS approaches adsorption as a linear equilibrium partitioning process between dissolved and adsorbed phases and is governed by the retardation factor R given by (Guiger *et al*., 1996; Spitz and Moreno, 1996):

$$R = \left[1 + \frac{\rho_b}{\theta} K_d\right] \qquad [11]$$

where: ρ_b — bulk density of the saturated porous medium [M/L^3]; K_d — distribution coefficient [M/L^3]

and the decay constant is given by:

$$\lambda = \frac{\ln(2)}{t_{1/2}} \qquad [12]$$

where: $t_{1/2}$ — contaminant half-life [T].

Numerical implementation

The transport equation [6] is solved in FLONET/TRANS using the Galerkin finite element method (Guiger *et al.*, 1996). The transport solution uses the same finite element grid as in the flow solution. Therefore it uses identical grid co-ordinates, including any deformation made to accommodate a free water table. The transport solution is transient and employs a user-defined time step. It assumes a dilute contaminant within an isothermal hydrogeological domain.