

Nitrate concentrations in the Gulp catchment: some spatial and temporal considerations

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ABSTRACT

In a chalk catchment in the Belgium-Dutch boundary region the median NO_3^- concentration was 30 mg/l in 1991. Groundwater in wells, which are mostly located close to the villages, is more polluted than spring water of surface water from the Gulp brook. Median NO_3^- concentrations were 39, 22 and 17 mg/l, respectively. Since 1980 time-series of NO_3^- from two gauging stations in the Gulp brook have showed a distinct seasonal pattern. In wet periods (winter), when the discharge is higher, the nitrate concentration also is higher (30-35 mg NO_3^- /l), whereas in dry periods the opposite occurs; i.e. lower discharge and nitrate concentrations (10-15 mg NO_3^- /l). The positive correlation between the discharge and the NO_3^- concentrations cannot be explained by the contribution of overland flow and interflow (quick flow components) probably having higher NO_3^- contents. Saturated groundwater modelling shows that water following different flow paths might be an explanation. In periods with high groundwater recharge, young groundwater in the upper chalk layer can bypass medium-aged and old groundwater in the lower chalk and greensands. Young groundwater is likely to have higher NO_3^- concentrations than old groundwater. So, in periods with high groundwater recharge the stream flow of the Gulp brook might consist of more young groundwater with higher NO_3^- concentrations, whereas in periods with low recharge mainly old groundwater with lower NO_3^- concentrations feeds the Gulp.

INTRODUCTION

In the southeast of the Netherlands the groundwater quality has been deteriorated by an increase of nitrate. In this region 20.106 m³ groundwater per annum (about 20% of the groundwater supplied by the Drinking Water Company of the Province of Limburg) is extracted from

unconfined chalk aquifers. In one well field, i.e. IJzeren Kuilen, the NO_3^- concentration has increased from 15 mg/l in 1955 to about 40 mg/l in the late 1980's. The NO_3^- concentration is expected to exceed the maximum allowable concentration of 50 mg/l in the early 2000's in spite of possible nitrogen reduction measures (Juhász-Holterman et al., 1989). The NO_3^- concentration of extracted groundwater does not continuously increase, but is related to groundwater recharge. The NO_3^- concentration significantly increases when wet years with a high groundwater recharge occur. In dry years the concentration drops again. Leaching of NO_3^- from the thick unsaturated zone, inclusion of a nitrate-rich unsaturated zone in the saturated zone due to a rise of the water table, and preferential saturated flow in the upper part of the chalk causes the positive correlation between NO_3^- concentrations of extracted groundwater and groundwater recharge in wet years (Juhász-Holterman, 1991).

The increased NO_3^- concentrations in the groundwater system also affect the surface water quality. In some chalk streams, e.g. the Mechelderbeek, the NO_3^- concentration has doubled (rise from 16 to 33 mg/l) in the period 1973-1984 (Schouten et al., 1986). Nota et al. (1988) show that a typical chalk brook like the Gulp has a distinct seasonal pattern. NO_3^- concentrations are positively correlated with discharge. In the winter period both discharge and NO_3^- concentrations are higher than during the summer. They explain these seasonal NO_3^-

differences by assuming that during the winter hillslope subsurface water flushes NO_3^- from the top layers towards the valley. Although they suggest this flow path, no specific investigations are carried out yet to obtain firm evidence. The objective of this paper is: (1) to investigate spatial differences of NO_3^- concentration of groundwater and spring water in distinct parts of the Gulp catchment, (2) to investigate if the observed seasonal NO_3^- patterns by Nota et al. (1988) still exist in the early 1990's, and (3) to explore possible groundwater flow paths using groundwater modelling, which might explain the seasonal variability of NO_3^- concentrations of surface water in the Gulp brook. We assume that groundwater that follows a shallow flow route through the upper part of the chalk has a higher probability on increased NO_3^- concentrations than the groundwater following a deeper route. Therefore the proportional distribution of groundwater following these different flow paths for various groundwater recharge regimes was investigated.

METHODS AND MATERIALS

Gulp catchment

The Gulp brook is a stream that drains the dissected chalk plateau in the southeast of the Netherlands and adjacent Belgium areas (Fig. 1). The total area of the basin is about 4600 ha, the length of the valley is 18 km, while the maximum width is 4 km. The Gulp brook rises at 285 m a.m.s.l. in Henri-Chapelle (Belgium), and joins the Geul, which is a tributary of the Meuse, in the village of Gulpen at 88 m a.m.s.l.. Near the village of Slenaken the Gulp brook enters Dutch territory. The cross profile of the Gulp catchment is asymmetrical (Fig. 1); the steepness of the eastern slope amounts to 17%, whereas the western slope averages 5%. Permanent grassland covers more than 90% of the catchment. Forest predominantly occurs on the steep, eastern slopes. The annual precipitation averages some 800 mm in Hombourg, and the average stream flow at the outlet is about 500 l/sec.

The geology of the Gulp catchment and some hydrogeological features are given in Fig. 1 and Table 1. Tilting and faulting because of the uneven Cenozoic uplifting has resulted in a fault zone in the western part of the basin.

The Gulp brook is deeply incised into the chalk plateau, which implies that surface water levels are far be-

low the plateau surface. Outside the valleys the unsaturated zone is thick (up to 60 m). The earlier-mentioned Upper-Cretaceous sediments form a multiple-aquifer system. The chalk and the sands of the Aken Formation are aquifers, which permit horizontal groundwater flow. The permeability of the chalk varies with depth; the upper layer has a significantly higher permeability than the lower layers because of weathering processes (Juhász-Holterman et al., 1989; Rooijen & Amkreutz, 1982). From the lower chalk layers the bottom layer (bottom chalk layer) has a permeability of about 1 m/day, whereas the overlying layer (intermediate chalk layer) has a somewhat higher permeability, e.g. between 3 and 5 m/day. The silty layers of the Vaals Formation do not allow substantial horizontal groundwater flow. The interbedded fractured sandstone layers (thickness 0.1-0.2 m), however, act as horizontal groundwater drains. On the plateaus and the slopes the overburden allows readily infiltration of precipitation into the soil. Experimental research confirmed that surface runoff and interflow hardly occur (e.g. Jansen & Verhagen, 1991). Interflow was defined as temporal saturated flow in the unsaturated zone of the hillslopes. Hydrograph analysis showed that at least 70% of the stream flow consists of groundwater discharge (baseflow). This means that outside the villages (including the roads) and the narrow wet valley, the excess precipitation recharges groundwater. The deep water tables show a delayed and smoothed response on excess precipitation. Water reaching the unconfined Upper-Cretaceous aquifer flows through the chalk and fractured sandstone layers in the Vaals Formation to springs in the valley. Probably minor leakage prevails through the clayey silts of the Vaals Formation towards the deep aquifer in the Aken Formation.

Groundwater and surface water monitoring

In the period 1975-1985 groundwater and surface water was intensively monitored in the Gulp catchment. Groundwater heads were observed in several dug wells and observation wells. On some of these locations every second month groundwater was sampled for the analysis of the chemical composition. Discharge of some selected springs and tributaries of the Gulp brook was measured and the chemical composition was analyzed. Stream flow of the Gulp brook was continuously measured at five locations along the longitudinal profile of the Gulp brook, e.g. F6 and F9 (Fig. 1). Once a month surface water was sampled for chemical analysis. Nota & Van de Weerd (1978; 1980), Nota & Bakker (1983),

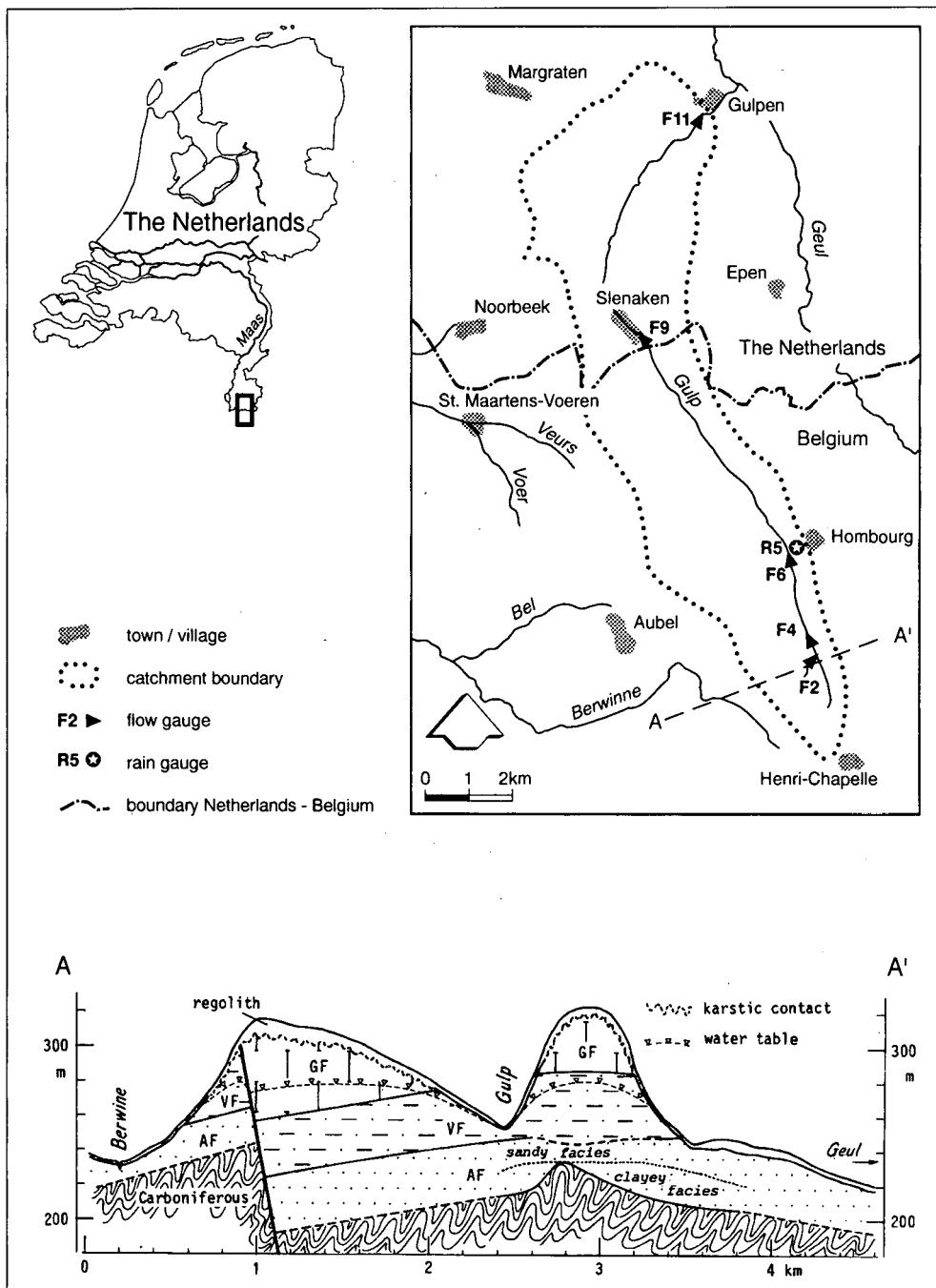


Figure 1. Location of the Gulp catchment and a cross-section showing the general hydrogeological conditions in the south of the catchment (derived from Nota et al., 1988).

and Nota et al. (1988) provide all relevant information. Since 1991 monitoring has started again on some strategic locations to investigate possible trends. In this recent period groundwater heads and surface water discharge including the chemical composition were occasionally measured for specific purposes on nearly all locations of the 1975-1985' network (e.g. Van Duinen, 1992).

The NO_3^- concentration of groundwater and surface

water was measured in the period 1980-1984 and since 1991 again. In the laboratory ion chromatography was used as an analytical procedure to determine NO_3^- concentration of the samples. For the incidental measurements NO_3^- was analyzed in a field laboratory using an ion-selective electrode. Comparison of both methods showed a good agreement (Van Duinen, 1992).

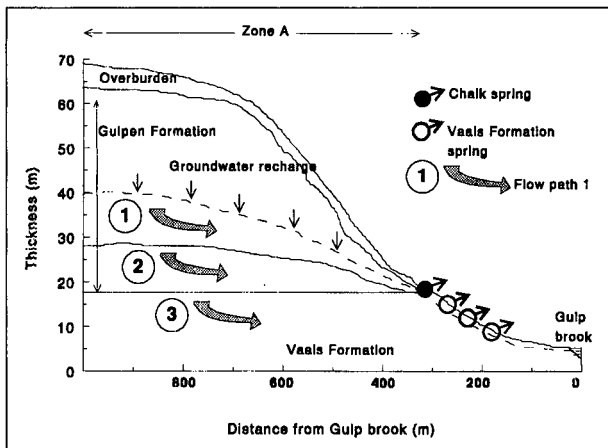


Figure 2. Schematic flow paths as modelled with MODFLOW for periods with a high groundwater recharge

Groundwater flow simulation

In the major part of the Gulp catchment excess NO_3^- can only reach the Gulp brook through various flow paths in the saturated groundwater system. Except from the villages and roads, no overland flow occurs. Groundwater flow modelling was applied to explore that flow route groundwater follows in the multi-layered aquifer system of the Gulp catchment. The amount of groundwater (specific discharge) following each of these flow paths was simulated. Although NO_3^- is a reactive ion no transformations, e.g. denitrification, were simulated in this phase of the study; NO_3^- was supposed to behave like a conservative ion. Saturated groundwater flow was simulated with a finite-difference groundwater flow model using the computer code MODFLOW (McDonald & Harbaugh, 1988). MODFLOW enables simulation of the groundwater head distribution and the water balance (e.g. groundwater flow towards a stream). In this reconnaissance stage modelling was restricted to the simulation of steady-state water flow in a vertical cross-section of the valley perpendicular to the Gulp brook. Such cross-sectional or profile models have been used frequently in an interpretive sense to study patterns in regional flow systems before designing a full three-dimensional model (e.g. Anderson & Woessner, 1992). The cross-section was subdivided in cells. In the horizontal direction from the watershed to the Gulp brook 100 cells were distinguished with a length of 10 m. In the vertical direction three cells, i.e. representing different hydro-stratigraphic layers, were selected. In the area where chalk is overlying the Vaals Formation (Fig.1) the upper layer represents the permeable intermediate chalk layer; the less-

permeable bottom chalk layer and the Vaals Formation are represented by layer 2 and 3. In the area next to the Gulp brook, where the chalk is eroded, the three vertical cells represent different layers of the Vaals Formation. No leakage to, or seepage from the underlying Aken Formation was assumed to occur. Flow in the thick unsaturated zone was not considered in the model. Groundwater recharge can follow three different flow routes in the model (Fig. 2): (1) horizontal flow through the permeable intermediate chalk layer towards a chalk spring at the margin of the chalk area (represented as a drain with MODFLOW), (2) vertical flow through the permeable intermediate chalk layer to the less-permeable bottom chalk layer and from there in a horizontal direction towards the earlier-mentioned chalk spring (see 1), and (3) vertical flow through both chalk layers to the Vaals Formation and from there in a horizontal direction towards springs or seepage areas at locations where the Vaals Formation outcrops (represented as drains at different elevations with MODFLOW). Part of the groundwater in the Vaals Formation seeps directly away into the Gulp brook (diffuse groundwater drainage). In reality the chalk springs are not found exactly at the permeability break between the chalk and the Vaals Formation, but at slightly lower elevations.

The results of MODFLOW permit computation of the water balance of different parts of the defined cross-section. For the area where the chalk is found (western part of the catchment, zone A in Fig. 2) we calculated the specific discharge of groundwater following the earlier-defined flow routes. This implies that the proportional distribution of the groundwater recharge over the three distinguished flow routes was computed.

The simulations were carried out for four different groundwater recharge regimes ranging from dry to wet under Dutch conditions, i.e. 0.0005, 0.0007, 0.001 and 0.002 m/day (Van Lanen, 1979). In the dry situation part or the entire permeable chalk layer (intermediate layer) might become unsaturated. Under these conditions no groundwater follows flow path 1 (Fig. 2). The schematic conditions in Fig. 2 apply to wet conditions, where part of the groundwater flows through the permeable intermediate layer (flow path 1). MODFLOW can handle a variable saturated thickness of the upper layer and allocates groundwater recharge to the first saturated layer. Heijnen & De Jong (1994) provide more details about the modelling in the Gulp catchment.

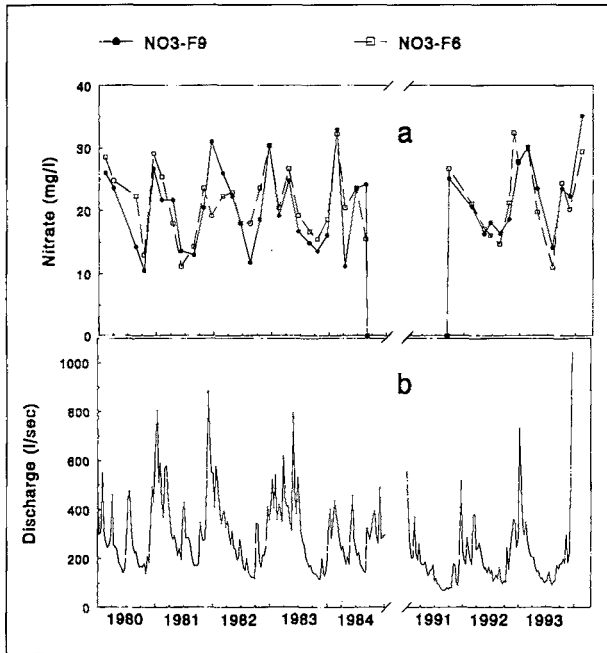


Figure 3. NO_3^- and stream flow rate of the Gulp brook: (a): variation of NO_3^- of surface water at two gauging stations along the Gulp brook (F6 and F9), and (b): 10-day' average discharge at Slenaken (F9).

RESULTS

Spatial distribution of nitrate observed in the Gulp catchment

The water analyses from the period 1981-1984 showed that NO_3^- pollution of groundwater was concentrated around the hamlets, where most of the groundwater observation wells are found. In some areas the NO_3^- concentration exceeded the 100 mg/l. Surface waters had significantly lower NO_3^- contents (Nota et al., 1988).

In June 1991 the NO_3^- concentration was determined of 53 groundwater wells and 50 springs. Surface water of the Gulp brook also was sampled on a few locations (Van Duinen, 1992). The results are summarized in Table 2.

Groundwater sampled from the wells contains higher NO_3^- concentrations than groundwater from the springs (median: 39 and 22 mg/l, respectively). This supports the conclusion of Nota et al. (1988) that groundwater taken from the wells reflects more local features (point observations) than spring water or surface water that shows more areal features. On the western slope groundwater taken from half the wells has NO_3^- contents exceeding the maximum allowable drinking water limit of 50 mg NO_3^- per litre. The occurrence of forests and the lower

agricultural nitrate load on the steeper eastern slopes is clearly reflected in the lower NO_3^- concentrations of the eastern wells and springs. The groundwater discharge from the eastern part of the basin is substantially lower than from the western part. Stream flow of the Gulp mainly consists of groundwater from the western part of the catchment. Therefore nitrate concentrations in this part have a higher impact on the nitrate concentrations of the Gulp than the eastern part. Groundwater is more polluted than the surface water of the Gulp brook; the median NO_3^- concentration is about twice as high (30 and 17 mg/l). This implies that the Gulp brook is also fed by other nitrate-poor groundwater than the groundwater taken from the selected wells and springs. The sampled wells are not fully representative for all groundwater. No clear zonation in NO_3^- concentrations of the groundwater from the wells and springs could be observed in a cross-section from the plateau to the valley, which would identify discharge areas of groundwater following different flow paths (Fig. 2).

Temporal distribution of nitrate observed in the Gulp brook

In the years 1980-1984 the monthly determined NO_3^- concentrations of the Gulp brook had a positive correlation with the 10-day' average stream flow (Nota et al., 1988). This positive correlation also applies to the period 1992-1993 (Fig. 3). More frequent investigations showed that during short periods, i.e. 22 May - 4 July 1991, when the stream flow was somewhat constant, NO_3^- concentrations did not show much variation (Van Duinen, 1992).

The NO_3^- time-series of the surface water sampled at the two gauging stations (for locations see Fig. 1) are similar. The observation period is rather short to investigate a possible trend in NO_3^- concentrations. The NO_3^- concentrations of surface water of the Gulp brook, which consists predominantly of groundwater, has a remarkably seasonal variability. The proportional distribution of groundwater discharge following different flow paths in dry and wet periods might be an explanation for this phenomenon as will be shown below.

Simulated proportional distribution of groundwater flow following different flow paths

The proportional distribution of groundwater recharge in the chalk area over the three flow routes is calculated

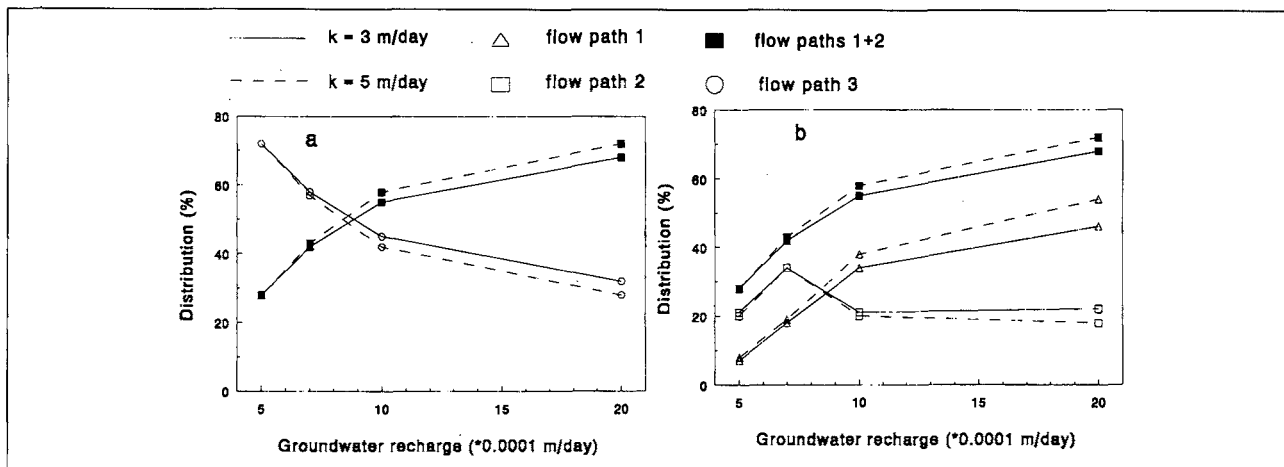


Figure 4. Simulated proportional distribution of the groundwater flow following different flow paths for various groundwater recharge regimes and two permeabilities of the intermediate chalk layer, (a) distribution of groundwater flowing through the chalk (flow paths 1+2) and groundwater flowing through the Vaals Formation (flow path 3), and (b) distribution of groundwater flowing through the permeable intermediate chalk layer (flow paths 1) and groundwater flowing through the less-permeable bottom chalk layer (flow path 2).

using MODFLOW for four different groundwater recharge regimes, i.e. varying from 0.0005 to 0.002 m/day. The simulations were carried out for two different permeabilities of the permeable intermediate chalk layer (layer 1), namely 3 and 5 m/day. The distribution of the recharge over the groundwater flow to the chalk spring (flow paths 1 and 2, see Fig. 2) and groundwater flow to the Formation of Vaals (flow path 3) is given in Fig. 4a.

In dry years (recharge 0.0005 m/day) about 30% of the groundwater recharge flows towards the chalk springs and the remaining part flows towards and through the Vaals Formation. In extremely wet years (groundwater recharge 0.002 m/day), when the intermediate chalk layer is saturated, the opposite occurs, about 70% of the recharge flows towards the chalk springs and only 30% through the Vaals Formation. Under more average conditions (recharge 0.0007-0.001 m/day) the distribution is close to 50%.

The distribution of the groundwater recharge over the two distinguished flow paths in the chalk, i.e. through the permeable intermediate chalk layer (flow path 1) and through the less-permeable bottom chalk layer (flow path 2) is given in Fig. 4b. In dry years the major part of the chalk spring flow (less than 30% of total recharge) comes from the less-permeable bottom chalk layer. This situation changes if recharge is slightly above average (recharge 0.001 m/day). In wet years chalk springs are predominantly fed by groundwater flowing through the more permeable intermediate chalk layer (about two-thirds of spring flow). A nearly doubling of the permeability of the intermediate chalk layer has a

minor influence on the proportional distribution (Fig. 4a and 4b).

The simulated residence times of groundwater following flow path 1, 2 or 3 might be in the ratio of 1:10:100, although no firm evidence (e.g. tracer tests) is available. This implies that the surface water of the Gulp brook might comprise young groundwater from the intermediate chalk layer (flow path 1), medium-aged groundwater from the bottom chalk layer (flow path 2) and old groundwater from the Vaals Formation (flow path 3). The distribution of residence times of the water particles in the Gulp brook depends on groundwater recharge. In dry years the Gulp brook consists of more old water that followed flow path 3 through the Vaals Formation, whereas in wet years young groundwater that flowed through the intermediate chalk layer dominates. For example in wet years about half the surface water of the Gulp brook consists of young groundwater. In dry years the percentage young water drops to less than 10%. Under these conditions a substantial part of the groundwater recharge can relatively quickly flow towards the chalk springs. This shallow young groundwater bypasses deep old groundwater.

CONCLUSIONS AND DISCUSSION

Groundwater simulation showed that the proportional distribution of groundwater flow to the Gulp brook following different groundwater flow paths in the Upper-Cretaceous multiple aquifer system is substantially affected by groundwater recharge. In wet years the

Table1. Hydrogeological characteristics of the Gulp catchment

Time period	Formation	Description
Quaternary		unconsolidated regolith or overburden (1-10 m); mixture of clay with flints and loess; unsaturated; permeability up to 5 m/day
Upper-Cretaceous	Gulpen (GF)	slightly consolidated, light coloured, fine-grained chalk with fissures (max. 40 m); upper part unsaturated; permeability: 1-25 m/day
	Vaals (VF)	saturated; glauconite-containing, unconsolidated clayey silts with thin fractured sandstone layers (max. 40 m); permeability of silts and sandstone layers: 0.02-0.2 m/day and 100-500 m/day, respectively
	Aken (AF)	starts with clayey sediments and proceeds with slightly cemented fine-grained sands (max. 35 m); permeability of sands: 6-8 m/day
Upper-Carboniferous		consolidated shales and sandstones impermeable base

permeable intermediate chalk layer is saturated and a significant part of the groundwater recharge (40-50%, Fig. 4) can follow a short flow path through this chalk layer and quickly feed the Gulp brook. Even under average recharge conditions, when part of the intermediate chalk layer is unsaturated, half the groundwater recharge flows relatively fast through the less-permeable bottom chalk layer (Fig. 4), which bypasses groundwater flowing through the Vaals Formation. So, the Gulp brook is fed by groundwater with different ages, which vary dependent on the groundwater recharge. This might be an explanation for the distinct seasonal patterns of NO_3^- in the surface water of the Gulp. The short groundwater flow routes (paths 1 and 2) are likely to have higher NO_3^- concentrations than the slower one through the Vaals Formation. Groundwater following the short flow paths contains recently leached water from the unsaturated zone that generally is rich in NO_3^- . Currently, in South-Limburg NO_3^- concentrations of soil water above 100 mg/l are not exceptional under agricultural land (Juhász-Holterman et al., 1989; Bosch & Pijpers, 1991). In the well-aerated soils, which are poor in organic matter, denitrification of NO_3^- is low (Schouten et. al., 1986). Deep groundwater that follows the long flow path contains less NO_3^- because it might be infiltrated before the increase of nitrate load by agriculture started some decades ago. Moreover in the dee-

per layers denitrification is likely to occur because the oxygen content decreases and some pyrite occurs in the Vaals Formation (Nota et al., 1988). Denitrification also will occur in the wet valley itself, where organic matter causes transformation of the NO_3^- , which could decrease the NO_3^- contents of the deep groundwater from the Vaals Formation that feeds the Gulp brook there.

The assumption that shallow young groundwater is nitrate-rich is supported by high NO_3^- concentrations of groundwater in most of the observation wells. The wells only penetrate the saturated zone over a few meters, thereby only young groundwater (leakage areas) or a mixture of shallow young groundwater and upcoming deep old groundwater (seepage areas) can be sampled.

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Table 2. Statistics of the NO₃ concentration (mg/l) in the Gulp catchment in 1991 (derived from Van Duinen, 1992).

Type	n	Median	Percentile	
			25th	75th
<i>Groundwater</i>				
All wells and springs	103	30	13	54
All wells	53	39	22	66
• western ¹⁾ wells	41	50	29	74
• eastern ¹⁾ wells	12	11	4	46
All springs	50	22	9	34
• western springs	32	30	13	37
• eastern springs	18	16	7	23
<i>Surface water</i>				
Gulp brook	4	17	17 ²⁾	19 ²⁾

1) western and eastern: west and east from Gulp brook, respectively

2) minimum and maximum, insufficient data to determine percentiles

on an earlier version improved the paper. The discharge data were kindly provided by the Waterschap Roer en Overmaas.

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