Identification of groundwater quality trends in a chalk aquifer threatened by intensive agriculture in Belgium

Jordi Batlle Aguilar · Philippe Orban · Alain Dassargues · Serge Brouyère

Abstract The European Union (EU) has adopted directives requiring that Member States take measures to reach a "good" chemical status of water resources by the year 2015 (Water Framework Directive: WFD). In order to achieve the environmental objectives for groundwater, the identification and reversal of significant upward trends in pollutant concentrations are required. A very detailed dataset available for the Hesbaye chalk aquifer in Belgium is used to evaluate tools and to propose efficient methodologies for identifying and quantifying nitrate trends in groundwater. Results indicate that the parametric linear regression and the non-parametric Mann-Kendall tests are robust; however, the latter test seems more adequate as it does not require verification of the normality of the dataset and it provides calculated nitrate trends very comparable to those obtained using linear regression. From a hydrogeological point of view, results highlight a general upward trend in the whole groundwater basin. The extrapolation of the trend analysis results indicates that measures have to be taken urgently in order to avoid further major degradation of groundwater quality within the next 10–70 years. However, a good groundwater quality status cannot be expected in the Hesbaye aquifer for the 2015 EU WFD deadline.

Résumé L'union européenne (EU) a adopté une directive imposant aux états membres d'atteindre le "bon" état chimique des ressources en eaux pour l'année 2015 (Directive cadre sur l'eau- DCE, 2000). Pour réaliser ces objectifs environnementaux pour les eaux souterraines, il est nécessaire d'identifier et inverser les tendances des concentrations en contaminants significativement à la hausse. Un jeu de données très détaillé, disponible pour la nappe aquifère de Hesbaye, un aquifère crayeux en Belgique, est utilisé pour évaluer des outils et proposer des méthodologies efficaces d'identification et de quantification des tendances en nitrates dans les eaux souterraines sur base d'une procédure statistique en trois étapes. Les résultats indiquent que la régression linéaire paramétrique et le test non paramétrique de Mann-Kendall sont robustes; cependant, ce dernier test semble plus adéquat car il ne requiert pas de vérifier la normalité du jeu de données et il produit des tendances en nitrates calculées très proches de celles obtenues avec la régression linéaire. Du point de vue hydrogéologique, les résultats montrent une tendance générale à la hausse dans l'ensemble du bassin hydrogéologique. L'extrapolation des résultats de l'analyse de tendance montre que des mesures doivent être prise sans tarder pour éviter une dégradation majeure des eaux souterraines dans les 10 à 70 prochaines années. Cependant, un bon état chimique des eaux souterraines ne peut déjà plus être attendu pour la date limite de 2015 prévue dans la DCE.

Resumen La Unión Europea ha adoptado directivas que instan a todos los Estados Miembros a tomar mesuras con el fin de alcanzar un "buen estado" químico de los recursos hídricos en vistas al año 2015 (Directiva 2000/60/CE -WFD). Con el fin de alcanzar los objetivos medioambientales relativos a las aguas subterráneas, son necesarias la identificación e inversión de las tendencias a la alza de las concentraciones de contaminantes. Detalladas series temporales correspondientes al calcáreo acuífero de de Hesbaye, en Bélgica, son el centro del presente estudio, con el fin de evaluar los métodos existentes y proponer pautas metodológicas eficientes para la identificación y cuantificación de las tendencias en las concentraciones de nitratos en las aguas subterráneas, usando un procedimiento estadístico basado en tres pasos básicos. Los resultados obtenidos instan a concluir que tanto el procedimiento paramétrico y no paramétrico, regresión lineal y el test de Mann-Kendall respectivamente, son suficientemente robustos; sin embargo, éste último se muestra más adecuado por el mero hecho que no necesita una previa verificación de la normalidad de la serie de

Received: 10 September 2006 / Accepted: 13 June 2007
Published online: 13 July 2007

J. Batlle Aguilar · P. Orban · A. Dassargues · S. Brouyère
Department of ArGEnCo,
University of Liège,
Building B52/3,
B-4000, Sart Tilman, Belgium

S. Brouyère (✉)
Aquapôle ULg,
University of Liège,
Building B52/3,
B-4000, Sart Tilman, Belgium
e-mail: Serge.Brouyere@ulg.ac.be
Tel.: +32-4366-2377
Fax: +32-4366-9520

 DOI 10.1007/s10040-007-0204-y

datos, obteniendo valores de tendencias totalmente comparables y a acorde con aquellos obtenidos mediante la regresión lineal. Desde un punto de vista hidrogeológico, los resultados demuestran una generalizada tendencia a la alza de la concentración en nitratos de las aguas subterráneas del acuífero estudiado. Asimismo, una extrapolación de los resultados de tendencias obtenidos indica que toda una serie de mesuras necesitan ser tomadas urgentemente con el fin de evitar una mayor degradación de la calidad química de las aguas subterráneas para los futuros 10–70 años. Paradójicamente, un "buen estado" químico de las aguas subterráneas es difícilmente imaginable para el año 2015, fecha límite propuesta por la Directiva WFD.

Keywords Groundwater monitoring · Nitrate · Trend analysis · Belgium · Water Framework Directive

Introduction

Environmental problems related to arable land have a long history in Europe, and they have accelerated in recent decades associated with changes to landscapes, vegetation and animal communities. This has mainly resulted in increased adoption of external inputs such as fertilizers and pesticides (Stoate et al. 2001). Strebel et al. (1989) highlighted nitrate pollution in Western Europe, the main source of nitrogen being agricultural leaching caused by excess inputs of fertilizers and manure, this being especially evident in north-western Europe (Iversen et al. 1998).

Agrochemicals that pose the greatest threat to human health are nitrate fertilizers and pesticides (Bouman et al. 2002; Gardner and Vogel 2005). The high nitrate concentrations in some groundwaters used for human consumption are causing increasing concern (Rajagopal and Tobin 1989). Diffuse (non-point) nitrate contamination originating from agriculture is a worldwide challenge which has been widely documented throughout the world (e.g. Mitchell et al. 2003; Mohamed et al. 2003; Thorburn et al. 2003; Oren et al. 2004; Liu et al. 2005). Many studies have shown the relation between agricultural practices and diffuse contamination of groundwater (e.g. Hudak 2000; Spalding et al. 2001; Harter et al. 2002; Johnsson et al. 2002; Lake et al. 2003). In contrast, point sources of nitrate contamination are more related to urbanized areas and septic tanks (Hantzsche and Finnemore 1992; Aravena et al. 1993; Aravena and Robertson 1998).

As a response to this threat, the European Union (EU) adopted in 1991 the Nitrate Directive 91/676/EEC (EU 1991) requiring that Member States take measures to minimize agricultural nitrate sources in nitrate-contaminated zones. More recently, the "Water Framework Directive 2000/60/EC" (EU 2000) was published, stating that a "good" status of groundwater is required for all EU Member States and specific measures have to be adopted to prevent and control pollution of groundwater. In order to achieve the environmental objectives for groundwater, the Water Framework Directive 2000/60/EC requires Member States to identify and reverse any significant upward trend in the concentration of pollutants and to achieve a good groundwater status by the end of 2015.

For EU Member States, a series of recommendations were made for groundwater quality trend assessment and data aggregation (Grath et al. 2001), proposing parametric (linear regression) and non-parametric (Mann-Kendall) methods, owing to their capability to detect different types of patterns of change and their robustness. Using very detailed nitrate datasets, an efficient methodology for trend analysis derived from the general guidelines of the EU Water Framework Directive is tested and evaluated in the Hesbaye chalk aquifer in the Geer watershed basin in Belgium. This is a representative northwest European chalky aquifer, from the point of view of its geology and spread of nitrate contamination (Downing et al. 1993). This analysis allows one to define the minimal requirements in terms of field data acquisition and processing. It also shows the limitations of the regulations in containing the problem and the fact that the nitrate problem is probably more acute than expected in the Geer basin. The study also suggests that many other cultivated groundwater basins in Europe have a similar problem because of the expected delay between changes in manure application at the soil surface and changes in groundwater quality and so reversal of nitrate trends.

Methodology for statistical trend analysis

The methodology used in this research mostly follows the work of Grath et al. (2001) who proposed particular algorithms and techniques for the identification of pollutant trends in groundwater. As suggested by Hirsch et al. (1991), a three-step procedure is considered (Fig. 1): (1) normality test of the dataset; (2) trend detection; and (3) trend estimation. This three-step procedure is described here after. Results of the three-step procedure are presented in Table 1.

Normality of the dataset

As will be discussed further in this report, the application of a normality test to the datasets is a priori necessary, in order to select the trend detection method to be applied (parametric or non-parametric). For this purpose, the Shapiro-Wilk test (Shapiro and Wilk 1965), hereafter called SW-test, or the Shapiro-Francia test (Shapiro and Francia 1972), hereafter called *SF*-test, was used depending on the number of records of the dataset (Conover 1980; Helsel and Hirsch 1995). Both tests have been widely used to test the normality of environmental datasets (Kumagai et al. 1997; Bonett and Seier 2002; Henderson 2005; Zhang and Wu 2005). They are generally considered as the most powerful tests of normality (Stephens 1974; Gan and Koehler 1990; Bonett and Seier 2002; Mudholkar et al. 2002).

In order to corroborate the results obtained using the SW and SF-tests, the D'Agostino's test (D'Agostino

 DOI 10.1007/s10040-007-0204-y

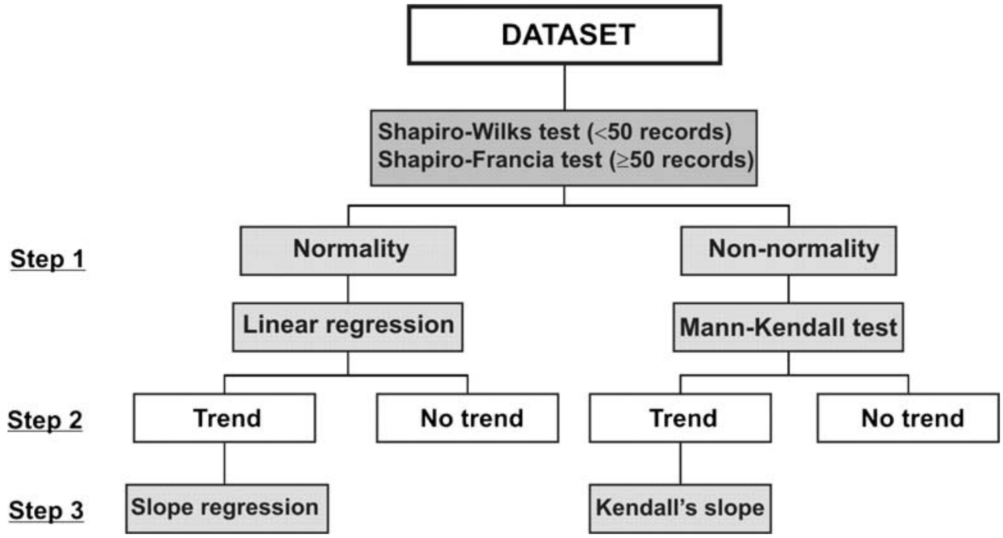

Fig. 1 A three-step procedure is adopted for nitrate trend analysis: (1) normal/non-normal distribution data; (2) trend detection; (3) trend estimation

1970a, b), hereafter called the D-test, has also been applied. It is also a very popular and commonly used test (Mudholkar et al. 2002). Generally, the D-test results have corroborated those obtained with the SW-test or the SF-test.

Trend detection

For normally distributed datasets, a simple linear regression is applicable as a tool for trend detection. Examples of application of this technique in groundwater studies include the works of Hanson (2002), Valverde Ramírez et al. (2005) and Zhang et al. (2005). Trend detection is based on the calculation of the correlation coefficient r, also called Pearson's r, which is a quantitative measure of correlation between time t and concentration C:

$$r = \frac{\sum_{i=1}^{n} (t_i - \bar{t})(C_i - \bar{C})}{\sqrt{\sum_{i=1}^{n} (t_i - \bar{t})^2 \sum_{i=1}^{n} (C_i - \bar{C})^2}} \tag{1}$$

In accordance with Carr (1995), three ranges of correlation degrees have been considered:

- Strong correlation for r values ranging between 0.8 and 1 (or −0.8 and −1)
- Moderate correlation for r values ranging between 0.5 and 0.8 (or −0.5 and −0.8)
- Weak correlation for r values ranging between 0.1 and 0.5 (or −0.1 and −0.5)
- No correlation for r values ranging between −0.1 and 0.1

For non-normally distributed datasets, trend detection is performed using the non-parametric Mann-Kendall test (Mann 1945; Kendall 1975), hereafter called MK-test. Examples of use of the MK-test for detecting trends in hydrological and hydrogeological time series data include the works of, e.g. Hirsch et al. (1982), Taylor and Loftis (1989), van Belle and Hughes (1984), Yu et al. (1993), Lee and Lee (2003), Kahya and Kalayci (2004), Zhang et al. (2005).

The MK-test has the advantage that it does not assume any distribution for the data and it has similar power as parametric methods (Serrano et al. 1999). The MK-test determines whether a trend is present or not with an indicator (T) based on the calculation of differences between pairs of successive data:

$$T = \sum_{i=1}^{n-1} \sum_{j=i+1}^{n} sgn(C_j - C_i), \tag{2}$$

where:

$$\mathrm{sgn}(C_j - C_i) = \begin{Bmatrix} 1 & \text{if} & C_j - C_i > 0 \\ 0 & \text{if} & C_j - C_i = 0 \\ -1 & \text{if} & C_j - C_i < 0 \end{Bmatrix}, \tag{3}$$

where C_j and C_i are concentration data at different time i and j, with $j>i$ and n is the size of dataset.

A 95% significance level has been used for the trend detection test (Helsel and Hirsch 1995), corresponding to a threshold value of T_{thresh}=1.65. It is thus considered that a trend is present for values of $T>T_{thresh}$. The MK-test being non-parametric, it is not possible to define various degrees of trend robustness, as performed with linear regression.

Trend estimation

The trend magnitude is expressed in units of increment of nitrate concentration per year (mg/L NO_3^- per year).

 DOI 10.1007/s10040-007-0204-y

Table 1 Trend test results for the observation points (groundwater) used in the statistical analysis using the Shapiro-Wilk (*SW*), Shapiro-Francia (*SF*) and D'Agostino's (*D*) tests

Sampling point characteristics				Normality test		Linear regression			Mann-Kendall test		
Sample point code	Date start	Date end	Number of records	SW/SF-test	D-test	*r*	Corr. Degree	Linear slope (mg/L NO_3^- per year)	Comparison level (*T*)	95% sl	H (mg/L NO_3^- per year)
H-1	11/05/1994	30/12/2003	126	N-N	N-N				8.64	Upward	0.48
H-2	05/01/1994	30/12/2003	123	N-N	N	0.38	W	0.26	3.93	Upward	0.24
H-3	06/01/1958	26/11/2002	101	N-N	N	0.84	S	0.33	9.95	Upward	0.36
H-4	27/06/1994	11/12/2002	131	N-N	N-N				0.43	No trend	–
H-5	05/01/1994	30/12/2003	123	N-N	N-N				2.78	Upward	~0.00
H-6	11/05/1992	05/08/2003	14	N-N	N-N				1.31	No trend	–
H-7	16/06/1958	12/12/2002	75	N	N	0.89	S	0.58	8.94	Upward	0.59
H-8	13/08/1976	12/12/2002	57	N-N	N-N				5.55	Upward	0.54
H-9	06/12/1957	24/10/2002	223	N-N	N-N				4.97	Upward	0.09
H-10	06/12/1957	02/02/2000	245	N-N	N-N				7.69	Upward	0.39
H-11	26/11/1998	25/07/2002	14	N-N	N-N				−1.58	No trend	–
H-12	26/11/1990	04/12/2002	112	N-N	N-N				5.50	Upward	0.32
H-13	16/05/1990	04/12/2002	116	N-N	N-N				3.83	Upward	0.31
H-14	16/01/1991	04/12/2002	234	N-N	N	0.53	M	0.47	4.42	Upward	0.45
H-15	07/10/1988	04/12/2002	347	N-N	N-N				−0.49	No trend	–
H-16	16/04/1996	28/05/2002	9	N	N	0.00	NC	0.00	0.00	No trend	–
H-17	12/11/1957	12/12/2002	35	N-N	N-N				2.23	Upward	0.16
H-18	06/07/1976	12/12/2002	54	N-N	N-N				7.16	Upward	0.48
H-19	31/12/1982	16/12/2002	172	N-N	N	0.14	W	0.15	2.08	Upward	0.07
H-20	15/08/1988	15/12/2002	329	N-N	N	0.58	M	0.73	10.14	Upward	0.61
HF-17	01/02/1986	25/01/2001	107	N-N	N	0.69	M	0.77	11.44	Upward	0.81
HF-18	07/08/1996	08/11/2000	9	N	N	0.09	NC	0.00	0.00	No trend	–
HF-21	29/05/1985	28/01/2005	137	N-N	N-N				1.58	No trend	–
HF-22	17/06/1981	10/01/2001	133	N-N	N-N				10.37	Upward	0.33

(*r* correlation coefficient; *H* Sen's slope estimator; *N-N* non-normally distributed dataset; *N* normally distributed dataset; *W* weak correlation; *M* moderate correlation; *S* strong correlation; *NC* no correlated data; *sl* significance level)

DOI 10.1007/s10040-007-0204-y

For normally distributed datasets, the magnitude of the trend is estimated using the slope of the linear regression equation (USEPA 2000):

$$C = a + bt, \quad (4)$$

where a is the y-intercept (t=0) of the line and b the slope.

For non-normally distributed datasets, the trend magnitude is based on calculation of the Sen's slope estimator (Hirsch et al. 1991), which is the nonparametric alternative to the linear regression slope. The Sen's slope is obtained as follows. First, one computes the slopes (b_{ij}) for all pairs of successive data:

$$b_{ij} = \frac{C_j - C_i}{t_j - t_i}, \quad (5)$$

where C_i and C_j are nitrate concentrations at time t_i and t_j, respectively.

Finally, the value of the Sen's slope estimator (H) is the median of those slopes (USEPA 2000):

$$\mathrm{b} = \mathrm{median}\left(\mathrm{b}_{ij}\right) \quad (6)$$

Trend analysis as applied to the Geer Basin

The Hesbaye aquifer, located in the Senonian chalk formations of the Geer basin, is an important groundwater resource for drinking water supply for the city of Liège and its suburbs, where around 600,000 people live and consume about 30 million m^3 of water per year (Brouyère et al. 2004a). The land use is dominated by agriculture, covering about 65% of the catchment area, the remaining space being divided between 15% of pasture, 13% of housing and 7% of forests (Broers et al. 2005). As shown further on, a very detailed nitrate dataset is available for this aquifer. These elements make it a very representative example of groundwater resources at risk in the sense of the EU Water Framework Directive.

Geographical, geological and hydrogeological context

The Geer River is a tributary of the Meuse River downstream of the city of Liège. The basin, located in the northern part of the Walloon Region in Belgium (Fig. 2), extends over about 480 km^2, with altitudes ranging between 80 m in the northeast and 206 m in the southwest, with a relatively flat topography. The Geer basin corresponds mostly to the unconfined part of the Hesbaye aquifer. However, the chalk formation dips northward and continues out of the basin. Because of that, the chalk aquifer extends also outside of the hydrological basin. Most of the aquifer is thus unconfined except in the northern part of the basin, where semi-confined conditions prevail on the north bank of the Geer River and outside of the Geer basin.

The geology is made of a succession of Mesozoic and Cenozoic layers that dip northward, with a slope gradient between 1 and 1.5%. From top to bottom, the geology consists of (Fig. 3):

- A Quaternary loess of variable thickness, up to 20 m
- A maximum of 10 m of flint conglomerate, a highly heterogeneous geological formation made of dissolved chalk residues (flints, sand, clay and locally phosphate)
- Locally, several metres of Tertiary sand deposits, mostly in the north of the basin, where they take the place of the flint conglomerate
- Cretaceous chalks forming the main groundwater reservoir, showing depths ranging from a few metres in the south up to 100 m in the north-eastern part of the basin. In most of the area, this layer is divided into two main units by a thin layer of hardened chalk called the "Hardground". This low permeability layer is not continuous, and where there is discontinuity, the hydraulic connectivity between the two main parts of the chalk aquifer can be enhanced.
- Several metres of smectite clay of low hydraulic conductivity, considered as the base of the aquifer

The mean hydraulic gradient in the aquifer is north-oriented (Fig. 4), ranging from 0.01 in the south to 0.003 in the north, close to the Geer River (Dassargues and Monjoie 1993). Groundwater levels are at depths ranging from 10 m to more than 60 m below the land surface.

The aquifer is exploited by two subsurface galleries and pumping wells owned by water companies, local industries and agricultural settlements. The northern gallery was dug in the chalk at a mean depth of 60 m. The southern gallery was dug at a mean depth of 30 m. These 40 km of galleries play a key role in the shape of the piezometric surface, acting in most part as a drain.

As highlighted by various groundwater studies in the basin, the chalk formation shows clear dual-porosity characteristics (Biver 1993; Hallet 1998; Brouyère 2001; Brouyère et al. 2004b). The large total porosity of the chalk (30–50%) allows for an important water storage capacity and the fissure porosity (about 1%) drains groundwater stored in the chalk matrix and provides the fastest pathways for transport.

Description of the nitrate dataset

The nitrate dataset used in this study comes mainly from the Nitrate Survey Network (NSN) established by the Walloon Region water authorities. In this network, existing boreholes, springs, galleries and traditional wells have been selected as monitoring points, where sampling and water analyses are carried out regularly, providing a spatial distribution of nitrate concentrations in the aquifer; in the best cases, data are available from 1957. Collected data are stored in a database of the Walloon Region water authorities. Each new dataset from the NSN is periodically imported into a hydrogeological database (Gogu et al.

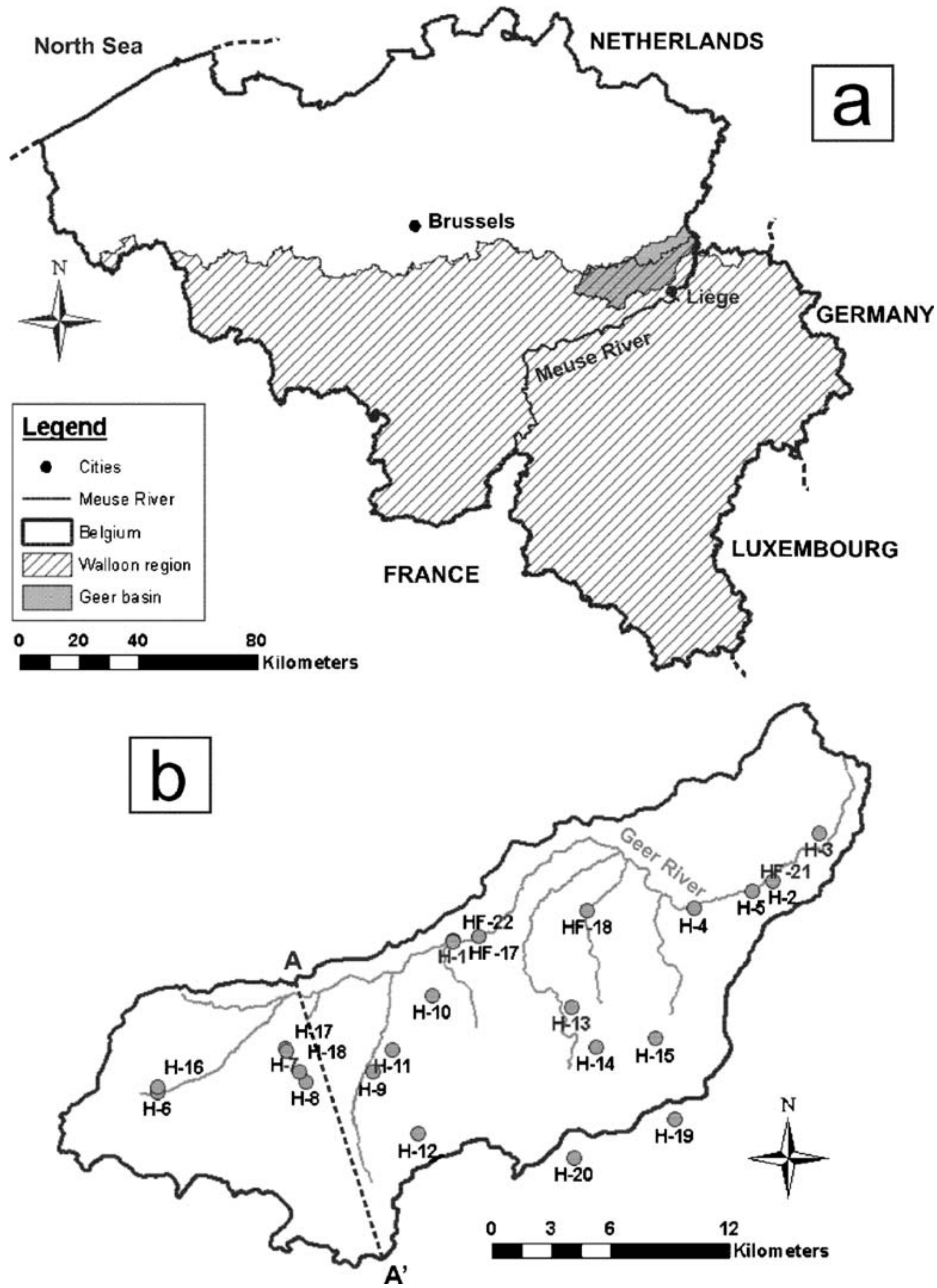

Fig. 2 **a** Geer basin location (*A–A′*: geological cross section, see Fig. 3); **b** Location of sampling points used in the trend analysis

2001; Wojda et al. 2005) developed and managed by the Hydrogeology Group of the University of Liège (HGULg).

The official NSN dataset has been extended using complementary points coming from other sources such as private owners or the VMW (Vlaamse Maatschappij voor Watervoorziening), the Flemish water supply company managing pumping wells in the north western part of the Geer basin and in the confined extension of the chalk aquifer in the Flemish region.

Finally, the network in the Geer basin nitrate study consists of 57 groundwater sampling points, among which 24 have been considered as containing suitable records for trend analysis (i.e. a minimum of 10 nitrate records over time). Importantly, however, there is a lack of knowledge on the position of the screens in most of the wells. Because of that, it was assumed that the deepest wells (generally the public water supply wells) were most probably screened in the totality of the aquifer. For the non-public water-supply wells, a similar criterion was used for the cases where knowledge on the screen position was not available. The remaining 33 points could not be used for trend analysis because of either a too small

 DOI 10.1007/s10040-007-0204-y

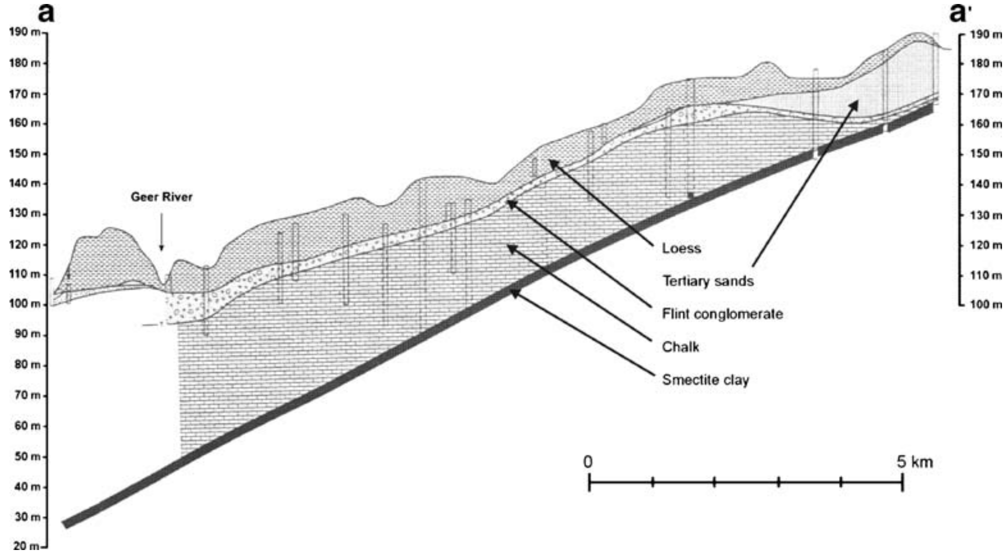

Fig. 3 Geological cross-section in the west part of the Hesbaye aquifer (see Fig. 2 for location of the cross section in the Geer basin; modified from Brouyère et al. 2004b)

number of records or a lack of nitrate detections (mostly in the north-western part of the basin corresponding to the confined zone). However, they were considered as useful for estimating the present distribution of nitrate in the basin.

Figure 5 shows the location of selected sampling points, with some representative time-nitrate concentration plots. Sample points H-19 and H-20 correspond to water reservoir tanks where groundwater drained by the galleries is collected for further distribution by gravity flow. Even if they are reservoir tanks and not sampling points in the aquifer, these two points have been considered in the statistical trend analysis because they constitute amalgamation points, representative of most of the groundwater in the Hesbaye aquifer, at least of the large portion of the aquifer drained by the subsurface galleries.

In the north-west, outside of the Geer watershed basin, nitrate is often below detection limit in groundwater, the time-concentration plot for HF-1 being illustrative of the confined zone of the Hesbaye aquifer. The absence of

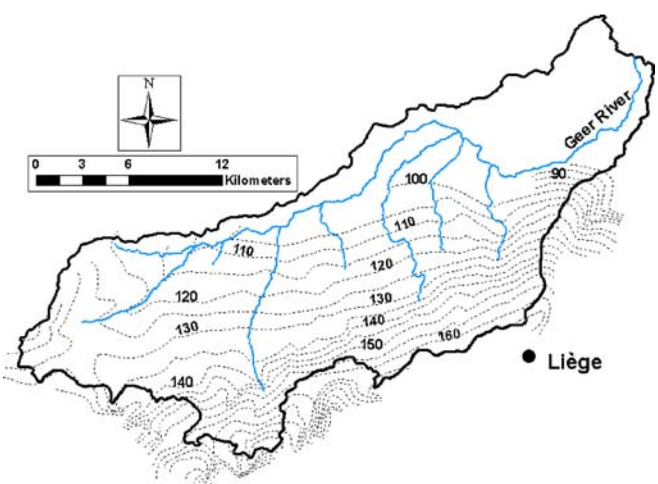

Fig. 4 Piezometric map of the Geer basin, based on a groundwater survey campaign in 1984 on the Hesbaye aquifer (in metres asl)

nitrate in this part of the basin may have two explanations: denitrification processes in this part of the aquifer where prevailing confined conditions are favourable for anoxic reducing conditions, or the occurrence of very old, uncontaminated groundwater. This point will be discussed later.

The first columns of Table 1 summarize time periods of nitrate records for the 24 sampling points selected for trend analysis. In many locations, concentrations are approaching the drinking limit of 50 mg/L (e.g. H-12, H-18 and H-20), sometimes above (e.g. H-10 and H-15) (Fig. 5). As indicated by Hallet (1998), the unconfined part of the aquifer is characterized by nitrate concentrations frequently over 45 mg/L. EPIC (Erosion-Productivity Impact Calculator) simulations (Dautrebande and Sohier 2004) suggest that 88% of the groundwater nitrate contamination originates from diffuse agricultural sources, while domestic, point-sources are responsible for the remaining 12%. The EPIC simulations take into account statistical data on agricultural land use and spreading of domestic waste from the sewage system.

Some of the datasets exhibit clear periodic variations in nitrate concentrations (e.g. H-15 in Fig. 5). As discussed by Brouyère et al. (2004b), such periodic variations are explained by groundwater level fluctuations in the variably saturated dual-porosity chalk. In principle, nitrate spread over the land surface progressively infiltrates through the soil and the unsaturated zone and migrates slowly downward through the unsaturated chalk matrix. Under low groundwater level conditions, the nitrate contamination front in the unsaturated chalk can be disconnected from the saturated zone and nitrate concentrations in the aquifer tend to diminish because of dispersion and mixing processes. When groundwater levels rise, the contamination front is quickly reached and leached. The contamination source is then re-activated and nitrate concentrations are likely to increase rapidly in the saturated zone. Generally, the periodic variations of nitrate concentrations are better observed at observation

 DOI 10.1007/s10040-007-0204-y

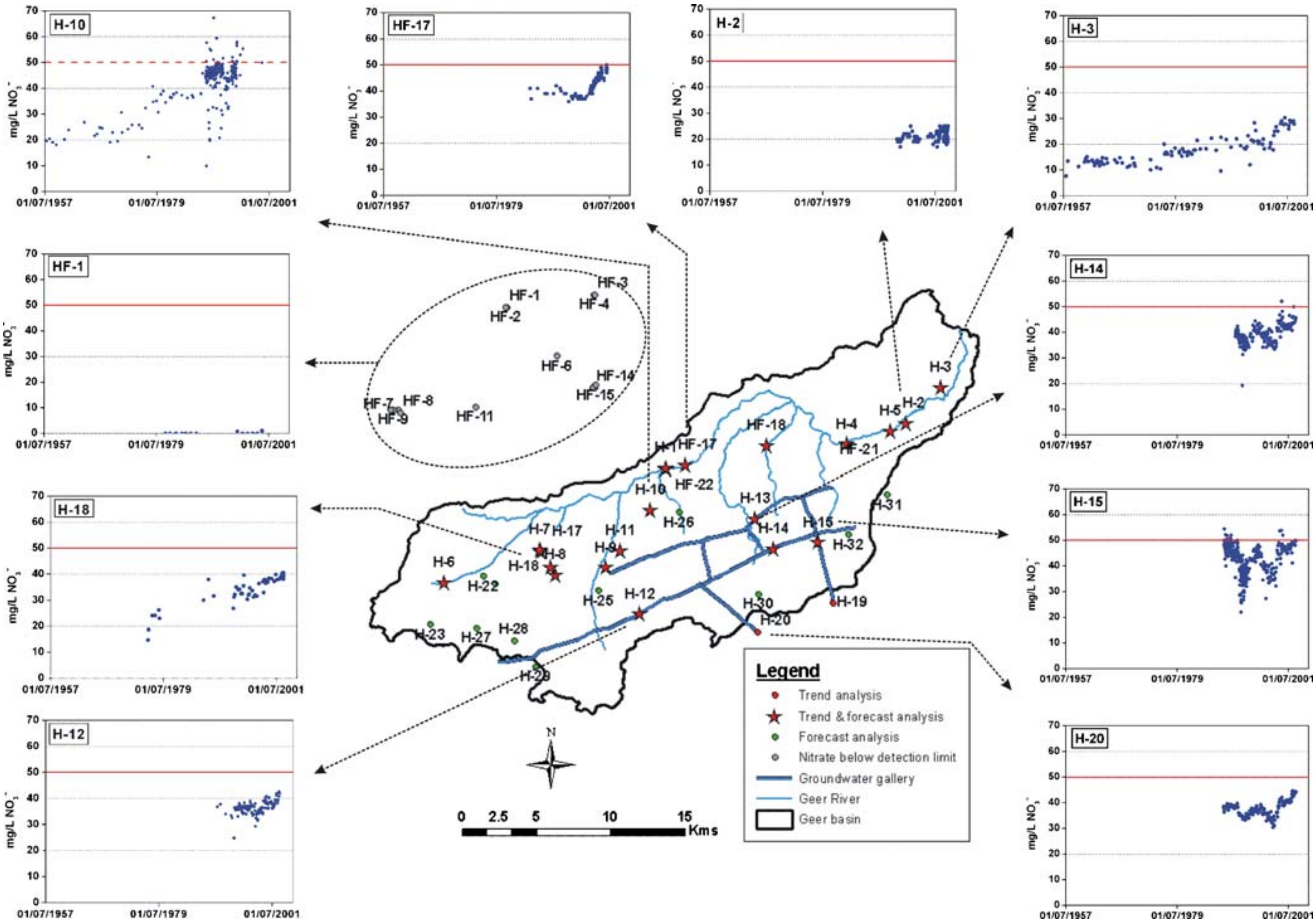

Fig. 5 Sampling locations in the unconfined part of the Hesbaye aquifer (inside the Geer basin) and the confined part of the Hesbaye aquifer (northwest of the Geer basin), and nitrate concentration as a function of time at selected locations

wells screened in the shallowest part of the chalk aquifer, which confirms the explanation given here above.

In the subsequent statistical analysis, the seasonal effect of such periodic variations has not been considered explicitly because of the difficulty in defining a clear periodicity which is related to pluri-annual variations in the precipitation regime. Neglecting the seasonality (or pluri-annual periodicity) is not a problem in trend detection because the datasets integrate several periods and the global trend emerges. Considering only a reduced period of time (about 1 year) would of course lead to dubious conclusions because the general trend is less likely to be observed during a shorter observation window.

Point-by-point trend estimation

Table 1 summarizes the results obtained in terms of statistical trend analysis for the 24 sampling locations selected in the Geer basin. A total of 17 sampling points are characterized by an upward trend (71% of the points), the annual increase of nitrate concentration ranging between 0.3 and 0.8 mg/L. The remaining points (7 out of 24) that do not show any evidence of upward or downward trend, generally correspond to sampling points with limited nitrate records, irregularly distributed in time. One can notice that four of these datasets (H-6, H-11, H-9 and HF-18) consist of between 9 and 14 records, distributed over 8 years. These results imply that in the Geer basin and similar systems, a minimum of 20 samples (the more the better), distributed over a period of 10 years (frequency: two samples/year) is required in order to perform effective statistical trend analysis. Three sample points (H-4, H-15 and HF-21) from those seven do not show evidence of a trend, although they have a sufficient number of records. In the case of the sample point H-15, for example, where a high number of records exist, one could think that the absence of trend detection is masked by the very marked pluri-annual periodicity in nitrate concentrations.

Concerning the points where results were contradictory in terms of normality tests, both the parametric linear regression and the non-parametric Sen's slopes were used to estimate the trend magnitude (see Table 1). Figure 6 shows that there is a very good agreement between slopes calculated using both techniques. Practically speaking, this means that the trend analysis methodology could be simplified by using the non-parametric method (Mann Kendall and Sen's slope estimator) for both normally and non-normally distributed datasets, without performing first the normality test (Hirsch et al. 1991). On the other hand, the SW/SF-test indicates that all datasets are non-normally

 DOI 10.1007/s10040-007-0204-y

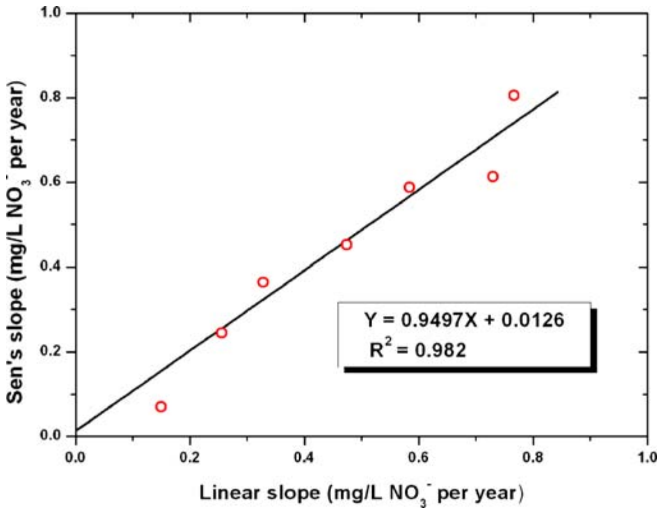

Fig. 6 Correlation between simple linear regression slopes and Sen's slope values

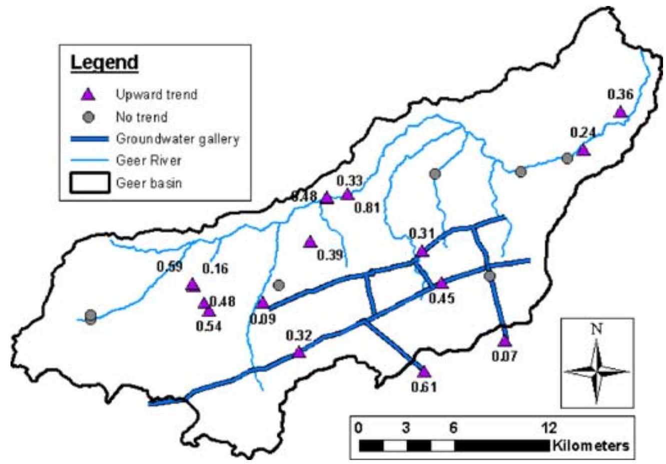

Fig. 7 Spatial distribution of nitrate trends in the Geer basin groundwater, with nitrate upward trend values in mg/L NO_3^- per year

distributed except in three cases: H-7, H-16 and HF-18 (see Table 1). Among these, only one (H-7) shows a trend for which the trend slope estimation is similar using parametric and non-parametric methods. Based on this example, one could thus conclude that the linear regression estimator is also good for non-normally distributed data. However, there are not enough data (just H-7) on which to validate such a conclusion.

Figure 7 gives a regional overview of nitrate trends in the Geer basin. Results indicate a general upward trend in the whole basin. However, as mentioned earlier, the unconfined part of the chalk aquifer, presenting high nitrate concentrations, has to be distinguished from the confined part of the aquifer (north of the watershed basin), where nitrate concentrations have been either below detection limit or very low. Figure 7 also indicates that the most significant upward trends are mainly located in the centre of the basin, where agriculture is more developed (Broers et al. 2005). One could think that subsurface galleries may act as a barrier to the migration of nitrate towards the north, explaining, in this way, the absence of nitrate in the confined zone. This is not likely to be the case because, even if the galleries do modify groundwater flows in the aquifer, they do not produce a reversal of groundwater flow at their vicinity. Furthermore, several points in the basin located to the north of the northern gallery (i.e. H-10 and HF-17) show high concentrations in nitrate and important upward trends (see Fig. 5 and Table 1), which confirms that the aquifer is also contaminated in this region.

Trend extrapolation

Presently, nitrate concentrations in groundwater are generally still lower than the drinking water limit of 50 mg/L, but from time to time, this threshold may already be exceeded. This is particularly the case in the southern subsurface gallery, which is at a shallow depth and thus more vulnerable to nitrate contamination. When exceedance occurs, the local water company mixes groundwater coming from the southern gallery with less contaminated groundwater coming from different points of the northern gallery in order to reduce the nitrate concentration of the supplied water to below the drinking water limit. Based on trend results obtained in the previous section, it is likely that this mixing procedure will be progressively less of an option in the future.

As mentioned by Grath et al. (2001), statistical techniques by themselves do not allow identification of trend reversal because they do not consider possible changes in manure application at the soil surface. However, in a context such as in the Geer basin, i.e. where there is a thick unsaturated zone, there is a long delay between changes at the land surface (i.e. reduction in manure-spreading) and the observed impacts in the aquifer (i.e. trend reversal). At first glance, this might lead to the conclusion that time is still available for taking adequate measures to effect a trend reversal. However, this oversimplified judgment assumes that measures taken to reduce the quantity of nitrate leaching to groundwater would have an immediate effect on groundwater quality. In fact, before reaching groundwater, nitrate must travel through the thick unsaturated formations (from 10 to 70 m) over a long period. Brouyère et al. (2004b) have estimated the mean vertical nitrate velocity across the unsaturated zone in the Geer basin to be approximately 1 m/year. Practically speaking, this means that measures taken today will have an observable effect in the groundwater with a delay of 10–70 years. Compared to that, the travel time of nitrate in the saturated zone to the pumping wells and galleries is extremely small, as it is dominated by fast preferential migration through the intense fracture network in the chalk (Hallet 1998; Brouyère 2001). Because changes in agricultural practices have started recently, one can anticipate that the upward trends observed in groundwater presently are not likely to reverse until many years into the future. Based on this assumption, a "simple" trend extrapolation is relevant in

DOI 10.1007/s10040-007-0204-y

Table 2 List of sampling points used for the calculated remaining time before exceeding the 50 mg/L NO_3^- drinking water standard. *ExcC* currently exceeds the drinking water standard

Sample point code	Mean concentration (mg/L NO_3^-)	Years to breakthrough limit	Expected year of exceedance
H-1[a]	43.05	14.50	2015
H-2[a]	22.11	116.21	2117
H-3[a]	27.68	62.00	2063
H-4[b]	28.19	90.90	2092
H-5[b]	19.4	127.5	2128
H-6[b]	9.28	75.4	2076
H-7[a]	37.83	20.6	2022
H-8[a]	38.32	21.6	2023
H-9[a]	27.48	250.2	2251
H-10[a]	49.90	0	ExcC
H-11[b]	34.97	38.5	2039
H-12[a]	38.10	37.2	2038
H-13[a]	40.00	32.3	2033
H-14[a]	43.12	15.3	2016
H-15[b]	46.69	7.4	2008
H-16[b]	5.57	82.3	2082
H-17[a]	40.84	15.5	2016
H-18[a]	38.09	24.8	2026
H-19[a]	47.00	4.9	2006
H-20[a]	40.69	15.3	2016
H-22[c]	100.00	0	ExcC
H-23[c]	82.00	0	ExcC
H-24[c]	35.00	27.80	2031
H-25[c]	43.8	13.80	2017
H-26[c]	62.7	0	ExcC
H-27[c]	33.20	31.10	2034
H-28[c]	14.50	71.00	2074
H-29[c]	39.2	27.00	2030
H-30[c]	32.00	36.00	2039
H-31[c]	28.40	54.00	2057
H-32[c]	64	0	ExcC
HF-17[a]	46.47	4.4	2004
HF-18[b]	23.00	60.00	2061
HF-19[b]	22.20	61.80	2067
HF-21[b]	19.20	128.3	2129
HF-22[a]	44.17	17.70	2018

[a] Points used in the statistical trend analysis with a calculated trend value

[b] Points used in the statistical trend analysis without trend evidence

[c] Points not used in the statistical trend analysis

order to estimate the time remaining before groundwater is unusable for public water supply.

A rough estimation of the time remaining before the threshold concentration of 50 mg/L would be reached in various parts of the chalk aquifer, has been calculated based on a point-by-point extrapolation of current nitrate contamination levels using nitrate trend estimates as obtained in this research. To do so, the present contamination level has been considered at 36 of the 57 available sampling points (excluding the area to the northwest of the basin, where nitrate concentrations are mainly below detection limit) based on a point-by-point calculation of mean nitrate concentration measured over the period 1999–2003, a period for which the nitrate dataset is well furnished. The resulting nitrate distribution is considered to be representative of year 2001, as the mean year of the period 1999–2003. Then, using the calculated slope value at the nearest available point, an estimation of the time remaining to reach the drinking water limit was performed. The extrapolation exercise was applied to the points where a trend was detected but also to observation points where datasets were not sufficient to perform a trend analysis and to points where no trend was detected in the analysis. In the latter case, one can expect that the absence of a trend at these points is most likely related to the restricted size of the corresponding datasets but nitrate is generally present at high concentrations. One exception is well H-15 where, as mentioned before, the dataset is very detailed and does not show any trend. Anyway, this point has already reached the drinking water limit of 50 mg/L.

Table 2 lists all the points included in this analysis and associated extrapolation results (number of years before reaching the threshold concentration of 50 mg/L NO_3^- and year of occurrence). Figure 8 shows the results of this calculation classified in four categories. Black circles correspond to locations where nitrate concentrations already exceed 50 mg/L NO_3^-. Red, orange and yellow circles correspond to locations where the limit will be reached within 30 years; between 30 and 60 years, and in more than 60 years, respectively. This confirms that the situation is more critical in the central part of the basin where for almost 80% of points, the drinking limit is expected to be reached within 30 years. Sampling points H-19 and H-20 are not represented because they correspond to water reservoirs tanks, not observation points in the aquifer.

Conclusions and perspectives

From a theoretical point of view, the statistical trend analysis, as performed in this research, seems to be robust. This is a result of the two-step procedure, with trend detection first and subsequent trend quantification. Results indicate that the preliminary normality test does not seem to be strictly necessary because the non-parametric MK-test works well with both normally and non-normally distributed datasets and the calculation of the trend amplitude using the Sen's slope estimator consistently

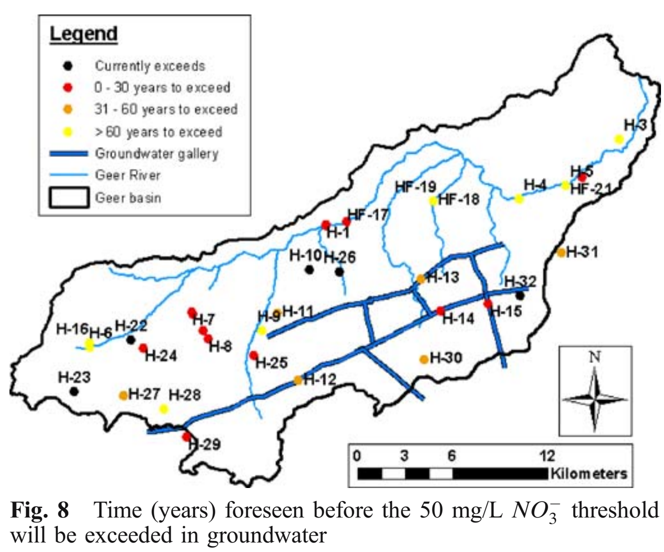

Fig. 8 Time (years) foreseen before the 50 mg/L NO_3^- threshold will be exceeded in groundwater

DOI 10.1007/s10040-007-0204-y

gives results that are comparable to the linear regression estimator. Indeed, it has been shown that in cases where parametric and non-parametric methods can be applied, results are similar for both methods, indicating that non-parametric methods (Mann-Kendall test in this case) are applicable for normally distributed datasets, making this test suitable for the Water Framework Directive application.

However, any statistical trend extrapolation relies on the strong and restrictive assumption that driving factors such as land use, agricultural practices, climate, and the dynamics of solute transport do not change with time. Thus, statistical trend extrapolation must be considered just as a warning procedure and not a forecasting tool. In order to go further in forecasting the evolution of nitrate concentrations in groundwater and in assessing or optimizing mitigation measures, more detailed and elaborated approaches are required such as transfer functions directly relating land-use and agricultural practices to the evolution of nitrate concentrations in groundwater; or even better, integrated mathematical modelling approaches based on observed physical processes of nitrate transport in the unsaturated and saturated zones.

The analysis presented here indicates that the problem associated with the evolution of nitrate concentrations in groundwater of the Geer basin is acute. Indeed, nitrate concentrations are already relatively high, sometimes locally above the drinking water limit; moreover the statistical trend analysis confirms that a general upward trend is observed throughout the basin. Furthermore, no downward trend has been identified, which means that there is no indication of trend reversal, despite the fact that first measures (EU 2000) have been introduced in relation to application of the EU nitrate directive, like control of manure spreading, and more recently the EU Water Framework Directive (Hall 1992). Extrapolation of the results, considering the “worst case scenario” of no significant changes in agricultural practices and land-use, indicates that the concentration threshold of 50 mg/L will be reached in 10–70 years in most of the unconfined aquifer. Measures should thus be adopted as soon as possible, having in mind the fact that, because of the long delay related to nitrate migration in the thick unsaturated zone, they will produce an observable effect within 10–70 years.

Further investigations have been performed in order to improve knowledge and understanding on the dynamics of nitrate in groundwater of the Geer basin. As pointed out by various authors (e.g. Broers 2004; Broers and van der Grift 2004) a strong relation is often observed between groundwater quality and age. In order to accommodate this factor, a groundwater dating campaign is ongoing in the Geer basin, using tritium and CFC-SF_6 (chlorofluorocarbons and sulphur hexafluoride, respectively) as environmental tracers. However, the interpretation of the results is difficult and ambiguous because of the heterogeneity and dual-porosity of the chalk (Weissmann et al. 2002; Cook et al. 2005; LaBolle et al. 2006). Such interpretation should however contribute to a better explanation of the absence of nitrate in the semi-confined area north west of the Geer basin.

Finally, the establishment of a good network to monitor diffuse nitrate groundwater contamination is a difficult challenge in many regions (Smith and Ritzi 1993; Nunes et al. 2004). Further investigations and geostatistical analyses will be performed in the Geer basin in order to propose an optimized monitoring network.

Beside these new field investigations, a regional groundwater model is being developed in the scope of a FP6-IP project (AquaTerra 2004). This model will be used for nitrate trend analysis and forecasting in the Geer basin. For this purpose, nitrate trend results obtained in the present study will be aggregated and used as calibration and validation datasets for the groundwater flow and transport model.

Acknowledgements This work was supported by the European Union FP6 Integrated Project AquaTerra (Project no. 505428) under the thematic priority, Sustainable Development, Global Change and Ecosystems; and by the Walloon Region DGRNE (Direction Générale des Resources Naturelles et Environment de la Région wallonne). The authors gratefully acknowledge F. Delloye from the DGRNE and J. van der Sluys from the VMW (Vlaamse Maatschappij voor Watervoorziening, Flemish region, Belgium) for their support in data acquisition and the two anonymous reviewers whose comments helped to clarify the manuscript significantly.

References

AquaTerra (2004) Integrated modelling of the river-sediment-soil-groundwater system: advanced tools for the management of catchment areas and river basins in the context of global change. Integrated Project of the 6th EU RTD Framework Programme Project no. 505428. AquaTerra. http://www.eu-aquaterra.de/. Cited 23 June 2007

Aravena R, Robertson WD (1998) Use of multiple isotope tracers to evaluate denitrification in ground water: study of nitrate from a large-flux septic system plume. Ground Water 36(6):975–982

Aravena R, Evans MLE, Cherry JA (1993) Stable isotope of oxygen and nitrogen in source identification of nitrate from septic systems. Ground Water 31(2):180–186

Biver P (1993) Etude phénoménologique et numérique de la propagation des polluants miscibles dans un milieu à porosité multiple [Phenomenological and numerical study of miscible contaminant propagation in a multi-porosity medium]. PhD Thesis, University of Liège. Belgium, 389 pp

Bonett DG, Seier E (2002) A test of normality with high uniform power. Comput Stat Data Anal 40:435–445

Bouman BAM, Castañeda AR, Bhuiyan SI (2002) Nitrate and pesticide contamination of ground water under rice-based cropping systems: past and current evidence from the Philippines. Agric Ecosyst Environ 92:185–199

Broers HP (2004) The spatial distribution of ground water age for different geohydrological situations in the Netherlands: implications for groundwater quality monitoring at the regional scale. J Hydrol 299:84–106

Broers HP, van der Grift B (2004) Regional monitoring of temporal changes in ground water quality. J Hydrol 296:192–220

Broers HP, Visser A, Dubus IG, Baran N, Morvan X, Normand M, Gutiérrez A, Mouvet C, Batlle Aguilar C, Brouyère S, Orban P, Dautrebande S, Sohier C, Korcz M, Bronder J, Dlugosz J, Odrzywolek M (2005) Report with documentation of reconstructed land use around test sites. Deliverable T2.2, Integrated Project FP6 no. 505428, AquaTerra , Tübingen, Germany, 64 pp

Brouyère S (2001) Etude et modélisation du transport et du piégeage des solutés en milieu souterrain variablement saturé [Study and modelling of transport and retardation of solutes in

 DOI 10.1007/s10040-007-0204-y

variably saturated media]. PhD Thesis. Faculté des Sciences Appliquées. Laboratoire de géologie de l'ingénieur, d'Hydrogéologie et de Prospection géophysique. Université de Liège, Liège, Belgium, 640 pp

Brouyère S, Carabin G, Dassargues A (2004a) Climate change impacts on ground water resources: modelled deficits in a chalky aquifer, Geer basin, Belgium. Hydrogeol J 12:123–134

Brouyère S, Dassargues A, Hallet V (2004b) Migration of contaminants through the unsaturated zone overlying the Hesbaye chalky aquifer in Belgium: a field investigation. J Cont Hydrol 72:135–164

Carr JR (1995) Numerical analysis for the geological sciences. Penitence-Hall, Englewood Cliffs, NJ, 592 pp

Conover WL (1980) Practical nonparametric statistics. Wiley, New York, 493 pp

Cook PG, Love AJ, Robinson NI, Simmons CT (2005) Ground water ages in fractured rock aquifers. J Hydrol 308:284–301

D'Agostino RB (1970a) Geary's test revisited. Psychol Bull 74:138–140

D'Agostino RB (1970b) Transformation to normality of the null distribution of g_1. Biometrika 57:679–681

Dassargues A, Monjoie A (1993) The chalk in Belgium. In: Downing RA, Price M, Jones GP (eds) The hydrogeology of the chalk of north-west Europe, chap 8. Clarendon, Oxford, UK, pp 153–269

Dautrebande S, Sohier C (2004) Projet de recherche PIRENE: Rapport final [PIRENE research project: final report]. University of Gembloux, France, 120 pp

Downing RA, Price M, Jones GP (1993) The hydrogeology of the chalk of north-west Europe. Clarendon, Oxford, UK, 300 pp

EU (1991) Council Directive of 12 Decembeer 1991 concerning the protection of waters against pollution caused nitrates from agricultural sources (91/676/EEC), EU, Brussels, 13 pp

EU (2000) Directive 2000/60/CE of the European Parliament and of the Council of 23 October 2000 establishing a framework for Community action in the field of water policy, EU, Brussels, 172 pp

Gan FF, Koehler KL (1990) Goodness-of-fit tests based on P-P probability plots. Technometrics 32:289–303

Gardner KK, Vogel RM (2005) Predicting ground water nitrate concentration from land use. Ground Water 43(3):343–352

Gogu CR, Carabin G, Hallet V, Peters V, Dassargues A (2001) GIS-based hydrogeological databases and ground water modelling. Hydrogeol J 9:555–569

Grath J, Scheidleder A, Uhlig S, Weber K, Kralik M, Keimel T, Gruber D (2001) The EU Water Framework Directive: statistical aspects of the identification of ground water pollution trends, and aggregation of monitoring results. Final report. Austrian Federal Ministry of Agriculture and Forestry, Environment and Water Management, Vienna (Ref.: 41.046/01-IV1/00 and GZ 16 2500/2-I/6/00) and Grant Agreement Ref.: Subv 99/130794, European Comission, Brussels, 63 pp

Hall DW (1992) Effects of nutrient management on nitrate levels in ground water near Ephrata, Pennsylvania. Ground Water 30 (5):720–730

Hallet V (1998) Étude de la contamination de la nappe aquifère de Hesbaye par les nitrates: hydrogéologie, hydrochimie et modélisaion mathématique des écoulements et du transport en milieu saturé [Contamination of the Hesbaye aquifer by nitrates: hydrogeology, hydrochemistry and mathematical modeling]. PhD Thesis, University of Liège. Belgium, 361 pp

Hanson CR (2002) Nitrate concentration in Canterbury ground water: a review of existing data. Report R02/17 ISBN 1-86937-457-6, Environment Canterbury, Christchurch, UK, 87 pp

Hantzsche NN, Finnemore EJ (1992) Predicting ground-water nitrate-nitrogen impacts. Ground Water 30(4):490–499

Harter T, Davis H, Mathews MC, Meyer RD (2002) Shallow ground water quality on dairy farms with irrigated forage crops. J Cont Hydrol 55:287–315

Helsel DR, Hirsch RM (1995) Statistical methods in water resources. Techniques of Water Resources Investigations, Book 4, Chapter A3, US Geological Survey, Reston, VA, 510 pp

Henderson AR (2005) Testing experimental data for univariate normality. Clin Chim Acta 366(1–2):112–129

Hirsch RM, Slack JR, Smith RA (1982) Techniques of trend analysis for monthly water quality data. Water Resour Res 18 (1):107–121

Hirsch RM, Alexander RB, Smith RA (1991) Selection of methods for the detection and estimation of trends in water quality. Water Resour Res 27(5):803–813

Hudak PF (2000) Regional trends in nitrate content of Texas ground water. J Hydrol 228:37–47

Iversen TM, Grant R, Nielsen K (1998) Nitrogen enrichment of European inland and marine waters with special attention to Danish policy measures. Environ Pollut 102(S1):771–780

Johnsson H, Larsson M, Martensson K, Hoffmann M (2002) SOILNDB: a decision support tool for assessing nitrogen leaching losses from arable land. Environ Model Softw 17(6):505–517

Kahya E, Kalayci S (2004) Trend analysis of streamflow in Turkey. J Hydrol 289:128–144

Kendall MG (1975) Rank correlation methods. Griffin, London, UK

Kumagai S, Kusaka Y, Goto S (1997) Log-normality of distribution of occupational exposure concentrations to cobalt. Ann Occup Hyg 41(3):281–286

LaBolle EM, Fogg GE, Eweis EB (2006) Diffusive fractionation of 3H and 3He in groundwater and its impact on groundwater age estimates. Water Resour Res 42:W07202. DOI 10.1029/2005WR004756

Lake IR, Lovett AA, Hiscock KM, Betson M, Foley A, Sünnenberg G, Evers S, Fletcher S (2003) Evaluation factors influencing groundwater vulnerability to nitrate pollution: developing the potential of GIS. J Environ Manage 68:315–328

Lee J-Y, Lee K-K (2003) Viability of natural attenuation in a petroleum-contaminated shallow sandy aquifer. Environ Pollut 126:201–212

Liu GD, Wu WL, Zhang J (2005) Regional differentiation of non-point source pollution of agriculture-derived nitrate nitrogen in ground water in northern China. Agric Ecosyst Environ 107:211–250

Mann HB (1945) Nonparametric tests against trend. Econometrica 13:245–259

Mitchell RJ, Scott Babcock R, Gelinas S, Nanus L, Stasney DE (2003) Nitrate distributions and source identification in the Abbotsford-Sumas aquifer, northwester Washington State. J Environ Qual 32:789–800

Mohamed MAA, Terao H, Suzuki R, Babiker IS, Ohta K, Kaori K, Kato K (2003) Natural denitrification in the Kakamigahara ground water basin, Gifu prefecture, central Japan. Sci Total Environ 307:191–201

Mudholkar GS, Marchetti CE, Lin CT (2002) Independence characterizations and testing normality against restricted skewness-kurtosis alternatives. J Stat Plan Inference 104:485–501

Nunes LM, Paralta E, Cunha MC, Ribeiro L (2004) Ground water nitrate monitoring network optimization with missing data. Water Resour Res 40, W02406. DOI 10.1029/2003WR002469

Oren O, Yechieli Y, Böhlke JK, Dody A (2004) Contamination of ground water under cultivated fields in an arid environment, central Arava Valley, Israel. J Hydrol 290:312–328

Rajagopal R, Tobin G (1989) Expert opinion and ground-water quality protection: the case of nitrate in drinking water. Ground Water 27(6):835–847

Serrano VL, Mateios VL, García JA (1999) Trend analysis of monthly precipitation over the Iberian Península for the period 1921–1995. Phys Chem Earth (B) 42(2):85–90

Shapiro SS, Francia RS (1972) An approximate analysis of variance test for normality. J Am Stat Assoc 67:215–216

Shapiro SS, Wilk MB (1965) An analysis of variance test for normality (complete samples). Biometrika 52:591–611

Smith RT, Ritzi JRW (1993) Designing a nitrate monitoring program in a heterogeneous, carbonate aquifer. Ground Water 31(4):576–584

Spalding RF, Watts DG, Schepers JS, Burbach ME, Exner ME, Poreda RJ, Martin GE (2001) Controlling nitrate leaching in irrigated agriculture. J Environ Qual 30:1184–1194

Stephens MA (1974) EDF statistics for goodness of fit and some comparisons. J Am Stat Assoc 69:730–737

 DOI 10.1007/s10040-007-0204-y

Stoate C, Boatman D, Borralho RJ, Rio Carvalho C, de Snoo GR, Eden P (2001) Ecological impacts of arable intensification in Europe. J Environ Manage 63(3):337–365
Strebel O, Duynisveld WHM, Böttcheer J (1989) Nitrate pollution of ground water in western Europe. Agric Ecosyst Environ 26 (3–4):189–214
Taylor CH, Loftis JC (1989) Testing for trend in lake and ground water quality time series. Water Resour Bull 25(4):715–726
Thorburn PJ, Biggs JS, Weier KL, Keating BA (2003) Nitrate in ground waters of intensive agricultural areas in coastal Northeastern Australia. Agric Ecosyst Environ 94:49–58
USEPA (2000) Practical methods for data analysis. Report EPA QA/G-9 QA00 Version, US Environmental Protection Agency (USEPA), 219 pp
Valverde Ramírez MC, de Campos Velho HF, Ferreira NJ (2005) Artificial neural network technique for rainfall forecasting applied to the Sao Paulo region. J Hydrol 301:146–162
van Belle G, Hughes JP (1984) Nonparametric tests for trend in water quality. Water Resour Res 20(1):127–136
Weissmann GS, Zhang Y, LaBolle EM, Fogg GE (2002) Dispersion of groundwater age in an alluvial aquifer system. Water Resour Res 38(10):161–168
Wojda P, Dachy M, Popescu IC, Ruthy I, Gardin N, Dassargues A, Brouyère S (2005) Appui à la conception de la structure, à l'interfaçage et à l'enrichissement de la base de données hydrogéologiques de la Région Wallonne [Support to structuring, interfacing and developing the Walloon Region Hydrogeological Database]. GeoMAC (Hydrogeology)-Walloon Region, University of Liège, Belgium, 48 pp
Yu Y-S, Zou S, Whittemore D (1993) Non-parametric trend analyses of water quality data of river in Kansas. J Hydrol 150:61–80
Zhang J, Wu Y (2005) Likelihood-ratio tests for normality. Comput Stat Data Anal 49(3):709–721
Zhang Q, Liu C, Xu C-y, Xu Y, Jiang T (2005) Observed trends of annual maximum water level and streamflow during the past 130 years in the Yangtze River basin, China. J Hydrol 324:255–265

 DOI 10.1007/s10040-007-0204-y