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AquaTerra

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

Thematic Priority: Sustainable development, global change and ecosystems

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Dissemination Level		
PU	Public	**X**
PP	Restricted to other programme participants (including the Commission Services)	
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CO	Confidential, only for members of the consortium (including the Commission Services)	

SUMMARY

The current document provides i) an overview of the scientific results obtained within the TREND2 module of the AquaTerra project; and, ii) a comparative assessment of the methodologies which were deployed to assess trends in water quality for the various case studies studied (the Dutch part of the Meuse basin, the Brévilles' catchment and the Geer catchment). The methodologies used in TREND2 were: i) age-dating techniques; ii) standard statistical trends analysis methodologies; iii) innovative techniques based on the combination of fuzzy logic and artificial neural networks; iv) transfer-function approaches; v) the deployment of complex deterministic models. Although efforts to simulate the transfer of water and contaminants at the various sites are continuing, the comparative assessment undertaken within TREND2 allows useful conclusions to be drawn already with regard to the merits and shortcomings of the various trends analysis approaches within an operational context.

MILESTONES REACHED

T2.11: Comparison of statistical and physically-deterministic methods of trend assessment and extrapolation in terms of data-requirements, costs and accuracy.

The present deliverable should be of particular interest to decision-makers, to EUPOL scientists and, more generally, to those looking for a position paper on the merits and shortcomings of trends analysis approaches. The document reports on the conclusions drawn from the application of selected trends analysis approaches to contrasting EU catchments within the context of AquaTerra TREND2.

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1 Introduction to TREND 2

1.1 Background and objectives

The implementation of the EU Water Framework Directive (2000/60/EU) and the draft Groundwater Directive asks for specific methods to detect the presence of long-term anthropogenically induced upward trends in the concentration of pollutants in groundwater. Specific goals for trend detection have been under discussion during the preparation of the recent draft of the Groundwater Directive. The draft Directive defines criteria for the identification and reversal of significant and sustained increasing trends in concentration and for the definition of starting points for trend reversal. Figure 1 illustrates the trend reversal concept, as communicated by EU Commission Officer Mr. Ph. Quevauviller. The figure shows how the significance of trends is related to threshold concentrations which should be defined by the member states.

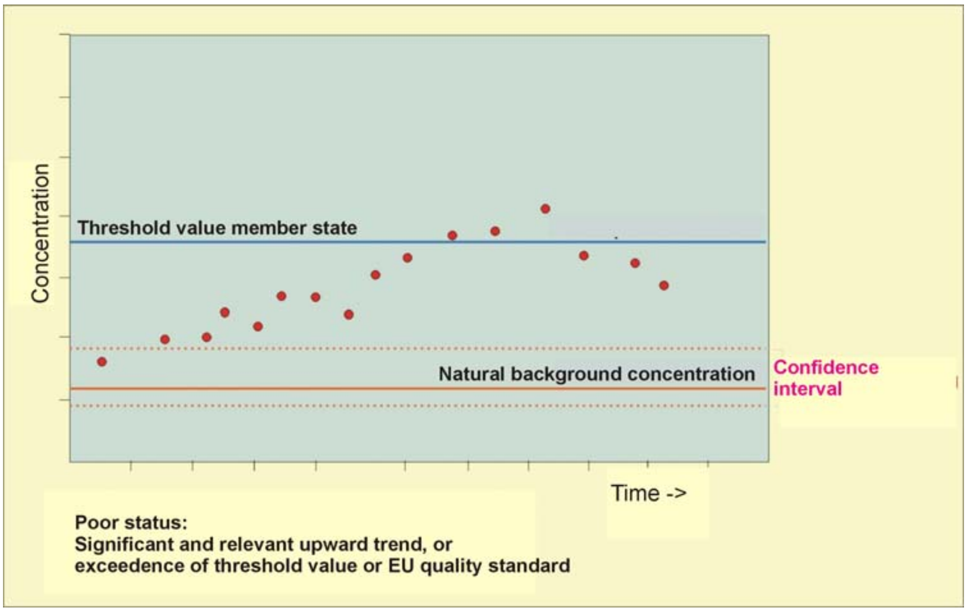

Figure 1 Trend reversal concept of the draft EU Groundwater Directive.

Trends should be reversed when concentrations increase up to 75% of the threshold concentration. Member states should reverse trends which present a significant risk of harm to associated aquatic ecosystems, directly dependent terrestrial ecosystems, human health, whether actual or potential, of the water environment, through the program of measures referred to in Article 11 of the Water Framework Directive, in order to progressively reduce pollution of groundwater. Thus, there is a direct link between trends in groundwater and the status and trends in related surface waters. This notion is central to the overall objectives of the AQUATERRA research project.

> Working hypothesis 1:
> Groundwater quality is of utmost importance to the quality of surface waters. Establishment of trends in groundwater is essential for prediction and evaluation of measures taken within the Framework Directive and the draft Groundwater Directive.

Accordingly, the work package TREND-2 of Aquaterra is dedicated to the following overall objectives.

1 Development of operational methods to assess, quantify and extrapolate trends in groundwater systems. The methods have been applied and tested at various scales and in various hydrogeological situations. The methods applied are related to the trend objectives of the Water Framework Directive and draft Groundwater Directive. In addition to the Description Of Work (DOW), it was our ambition to link changes in groundwater quality to changes in surface water quality.

2 Linking changes in land use, climate and contamination history to changes in groundwater chemistry. We define a temporal trend as '*a change in groundwater quality over a specific period in time, over a given region, which is related to land use or water quality management',* according to Loftis 1991, 1996.

It should be noted that trends in groundwater quality time series are difficult to detect because of (1) the long travel times involved, (2) possible obscuring or attenuating effect of physical and chemical processes, (3) spatial variability of the subsurface, inputs and hydrological conditions and (4) short-term natural variability of groundwater quality time series and (5) costs associated with groundwater sampling in time and particularly in space (i.e. it costs money to establish new piezometers and wells to access groundwater). The TREND 2 package was dedicated to the development and validation of methods, which overcome many of these problems.

> Working hypothesis 2:
> Detection of trends in groundwater is complicated by spatial variations in pressures, in flow paths and groundwater age, in chemical reactivity of groundwater bodies, and by temporal variations due to climatological factors. Methods for trend detection should be robust in dealing with historic and actual atmospheric deposition

Groundwater pollution is caused by both point and diffuse sources. Large scale groundwater quality, however, is mainly connected to diffuse sources, so that the TREND 2 project concentrated on trends in groundwater quality connected to diffuse inputs, notably nutrients, metals and pesticides. Although trends in groundwater quality can occur at large scales, linking groundwater quality to land use and contamination history requires analysis at smaller scale, i.e. groundwater subsystems. Thus, the approach zooms in on groundwater system analysis around observation locations. Results are to be extended to large scale monitoring via upscaling Alternative integral sampling methods (i.e. looking at river water chemistry as a groundwater proxy or remote sensing methods) can be developed.

1.2 General methods used in TREND 2

Research activities within TREND 2 have focussed on the following issues:

1 *Inventory of monitoring data of different basins and sub-catchments*. The inventory focused on observation points with existing long time series. The wells were located in agricultural areas, because pesticides and nutrients are the main concern in trend detection for the Water Framework Directive. Additional information was collected about historical land use changes and related changes in the input of solutes into the groundwater system.

2 *Development of suitable trend detection concepts.* Trend detection concepts included both statistical approaches (classical parametrical and non-parametrical methods, hybrid techniques) and conceptual approaches (time-depth transformation, age dating)

3 *Methods for trend aggregation for groundwater bodies.* The Water Framework Directive demands that trends for individual points are aggregated on the spatial scale of the groundwater bodies. The project has attempted to provide robust methods for trend aggregation.

4 *Trend extrapolation.* Trend extrapolation has been based on statistical extrapolation methods and on deterministic modelling. Both 1D and 3D model may be applied to predict future changes and to compare these with measured data from time series.

5 *Recommendations for monitoring.* Results from the various case studies will be used to outline recommendations for optimizing monitoring networks for trend analysis

1.3 TREND 2 case studies

The following case studies have been selected for testing the methodologies (Table 1). Statistical trend extrapolation have been performed on all the selected case studies. Deterministic modelling was limited to the Dommel and the Geer catchment in the Meuse basin, and the Brévilles catchment.

Table 1: Case studies in TREND 2

Basin	Contaminants	Trend extrapolation	Institutes
Meuse			
Dommel upper tributaries	Nitrate, sulfate, Ni, Cu, Zn, Cd	Statistical and deterministic modelling	TNO/UU
Noord-Brabant region	Nitrate, sulfate, Ni, Cu, Zn, Cd	Statistical	TNO/UU
Wallonian catchments: • Néblon • Pays Herve • Hesbaye • Floodplain Meuse	Nitrate	Statistical	ULg
• Geer catchment	Nitrate	Statistical and deterministic modelling	ULg
Brévilles			
Brévilles catchment	Pesticides	Statistical and deterministic modelling	BRGM
Elbe			
• Czech subbasins	Nitrate	Statistical	IETU
• Schleswig-Holstein	Nitrate		

These cases have different spatial scales and different hydrogeological situations. Details on the various cases are provided in previous TREND 2 deliverables: T2.1 (description of cases), T2.2 (historical land use and contaminant inputs), and T2.5 (model input data).

1.4 Contents of the current report and timing

The current document provides i) an overview of the scientific results obtained within the TREND2 module of the AquaTerra project; and, ii) a comparative assessment of the methodologies which were deployed to assess trends in water quality for the various case studies studied (the Dutch part of the Meuse basin, the Brévilles' catchment and the Geer catchment). It should be noted that although modelling efforts are still underway in the various institutes involved, the experience gained allows the drawing of conclusions already regarding the merits and shortcomings of the various methodologies investigated for trends analysis. The present document was produced slightly later in time than initially estimated. This is due to modelling activities of TREND2 partners having being delayed in response to the late delivery of climate change meteorological series by the HYDRO work package.

1.5 Structure of the report

The first chapter is an introduction chapter recalling the importance of trends analysis in the current legislative context. The 3 subsequent chapters provide a summary of the efforts deployed in applying the various trend analysis techniques at the study sites (Dommel, Geer, Brévilles). The last chapter attempts to make a comparative assessment of the various techniques, which were deployed within the context of TREND2.

1.6 Glossary

CFC	Chlorofluorocarbons, used to date groundwater
apparent groundwater age	Groundwater travel time estimated from transient tracer concentration
Holt's two parameter methodology	A classical method for analysis of trends in statistics
KIWA	Dutch water research agency
MT3DMS	Three-dimensional multi-species transport model
Oxidation capacity (OXC)	Weighted sum of nitrate and sulphate concentrations
Kendall-τ correlation coefficients	Non-parametric correlation coefficient
Kendall-Theil trend slopes	Non-parametric method to determine trend slope
deterministic model	Model which implements a comprehensive description of phenomena using sets of equations
VMW (Vlaamse Maatschappij voor Watervoorziening)	Flemish water supply company
normality tests	Tests to check a given set of data for similarity to the normal distribution
Hybrid Finite Element Mixing Cell technique (HFEMC)	New technique developed by HG-ULg for groundwater flow and solute transport modelling
SUFT3D	Groundwater model
EPIC-Grid model	Soil model for water and nitrate fluxes
ARIMA	Statistical analysis used commonly to assess trends in time series (autoregressive integrated moving average)
effective precipitation	Part of the precipitation which effectively contribute to groundwater recharge
Deterministic modelling	Modelling activities which rely on series of equations describing each of the physical, chemical and biological phenomena of importance by a series of numerical equations
transfer function approach	Approach originating from the theory of signal processing in electronics which relies on the calibration of numerical functions expressing the transformation (e.g. reduction, delay) of an input signal to result in an output signal

2 Context and objectives of the present report

The effective transfer of contaminants, may they be of industrial, natural or agricultural origin, to surface water and groundwater represents a major threat to the long-term sustainability of water resources across the European Union and elsewhere. This has been recognized at the legislative level by the introduction by the European Commission of the Water Framework Directive, which proposes challenging objectives with regard to the long-term quality of water resources in the EU. Monitoring programmes to assess the status and evolution of the quality of European water bodies are being established and large amounts of physical and chemical data are becoming increasingly available to support the management of water resources. Most of monitoring programmes in place are aimed at evaluating whether water quality standards are met, for instance to decide whether pollutant concentrations exceed regulatory thresholds. Although programmes are designed to some extent to capture some of the inter-annual or intra-annual variability in concentrations and fluxes, the costs associated with running these programmes means that programmes are often not optimized to assess or detect trends in water quality. These trends aspects are currently receiving much attention within the context of new environmental directives to answer questions such as:

- is the water quality improving or degrading?
- Is the current water quality status likely to change in the future?
- How much time do we need to achieve 'good ecological status'?

The TREND2 module of the AquaTerra project is dedicated to the application of various trends analysis and detection techniques to a range of catchments to come up with informed statements regarding the merits and shortcomings of various trends analysis techniques in both a research and operational context. The current document provides i) an overview of the scientific results obtained within the TREND2 module of the AquaTerra project; and, ii) a comparative assessment of the methodologies which were deployed to assess trends in water quality for the various case studies studied (the Dutch part of the Meuse basin, the Brévilles' catchment and the Geer catchment). Much of the underlying science supporting our conclusions has been reported in earlier AquaTerra deliverables and the document therefore attempts to refer to these deliverables as much as possible in an effort to focus on the more innovative aspects of the science and the more practical inferences. The methodologies used in TREND2 were:

i) age-dating techniques;
ii) standard statistical trends analysis methodologies;
iii) innovative techniques based on the combination of fuzzy logic and artificial neural networks;
iv) transfer-function approaches;
v) the deployment of complex deterministic models.

3 Applications of trend analysis techniques in the Brévilles agricultural catchment in France (BRGM)

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The Brévilles spring is the main outlet of a small agricultural catchment in the village of Montreuil-sur-Epte, situated some 70 km west of Paris. The Brévilles catchment has been the subject of detailed characterization efforts to study the transfer of water and agricultural contaminants to the Brévilles spring within the FP5 PEGASE and the FP6 AquaTerra projects. The main context of the catchment is the contamination of groundwater resources and the Brévilles spring by the herbicide atrazine and its main degradation products.

As part of TREND2, the time series of water and pesticide fluxes and concentrations were used to assess the merits and shortcomings of the following techniques for studying trends in water quality contamination: i) age-dating techniques; ii) standard statistical trends analysis methodologies; iii) innovative techniques based on the combination of fuzzy logic and artificial neural networks; iv) transfer-function approaches; v) the deployment of complex deterministic models. Methodological developments were undertaken as part of (iii) and (iv) and much effort has been put into (v) since the catchment was first instrumented at the end of the 20th century.

3.1 The Brévilles catchment

The Brévilles catchment has been described in detail in a number of AquaTerra deliverables and the present paragraph will therefore only contain a summary description. The reader is referred to the following AquaTerra BASIN and HYDRO deliverables for additional information (http://www.attempto-projects.de/aquaterra/51.0.html) : BASIN 1.2, BASIN 1.5, BASIN 1.6, HYDRO 2.1, HYDRO 2.1bis, HYDRO 2.2, HYDRO 2.4.

The Brévilles experimental catchment is a small watershed situated 70 km west of Paris, at the boundary between the Ile de France and Normandie regions. Agricultural activities in the catchment are dominated by crop production (wheat, maize). The study area was instrumented in 1999 as part of the EU PEGASE project to investigate water, major ions and pesticide fluxes through the soil and the subsoil. Monitoring activities have been conducted on a two-week return period in seven piezometers and the Brévilles spring. Two soil types are broadly encountered on the catchment: i) a moderately deep, silty-loamy to silty-clayey neoluvisol (45% of the area); and, ii) a thin, stony, silty clay loam soil (55% of the area). From a geological perspective, the catchment is essentially made of tertiary material: a carbonaceous formation from Lutetian age and sandstones from Bartonian age. Quaternary sediments are present in the form of silts on the upper part of the catchment and colluvions at the foot of hills. Below the chalk formations the 'Cuise' sands (upper part of Ypresian) are located with a thickness of 8-20 m, which constitute the main aquifer layer in the hydrogeological catchment. The basement of the aquifer is made of ca. 10 m of 'Sparnacian clays' which directly overlay several hundreds of meters of chalk of Cretaceous age. Hydrogeologically speaking, the Cuise sands aquifer is not fully saturated except to the west of the basin due to the dip in geological layers being stronger than the hydraulic gradient of the water table. The aquifer is generally unconfined (although local semi-confinement may occur) and outcrops on the western and southern slopes of the aquifer giving birth to a number of springs, including the Brévilles spring, the main aquifer outlet. Seven piezometers were drilled in 2001 and cross the water table down to the Sparnacian clays. The depth of the water table was found at 8-42 m depending on their location in the basin. Within the AquaTerra project further piezometers were established to a field of a total of 20 piezometers.

The data used within TREND2 at Brévilles focused on concentrations measured for nitrate, calcium, atrazine and its first metabolite deethylatrazine in the spring water and the seven piezometers in the catchment for the period extending from 1999 to mid-2006. Additional piezometers were drilled in 2005 as part of the AquaTerra project, but results originating from these new drillings were not included in the TREND2 work as they did not lead to significant historical time series.

3.2 The application of trends analysis techniques to the Brévilles dataset

3.2.1 Age-dating techniques

A literature review was undertaken to investigate the potential usefulness of age-dating techniques for fractured systems such as the Brévilles hydrogeological catchment (Deliverable T2.3). The major issue for age dating water in fractured aquifers is that the implications of matrix diffusion processes for apparent groundwater ages measured with environmental tracers are not well understood. It is nevertheless clear that groundwater flow through vertical fractures can result in rapid water movement, and cause very young apparent groundwater ages at considerable depth. Groundwater sampling in fractured rocks creates additional difficulties because of the heterogeneous distribution of the fractures. Great precautions should therefore clearly be taken for the selection of the sampling points.

The use of CFC for apparent age-dating at Brévilles (Deliverable T2.3) gave ages compatible with the hydrogeological functioning of Brévilles aquifer. The heterogeneity in apparent ages obtained is consistent with other observations made at the site suggesting a great variability in water flow, which has been tentatively attributed to a high fracture density and variability in geological characteristics. In addition to the presence of groundwater mixture and matrix diffusion phenomena, the interpretation of the CFC data at Brévilles is made difficult by the presence of a thick unsaturated zone (0-35 m) as it is usually considered that errors in excess of 8-12 years are expected for unsaturated zone thickness exceeding 30 m. Although the combination of information gathered for various environmental tracers is expected to be of interest in some instances, the heterogeneity observed at the site, the presence of a thick and fractured unsaturated zone raises doubts over the potential interpretation of the data, even if sophisticated reservoir models and alternative age-dating techniques were used.

3.2.2 Standard statistical techniques for trends analysis

The Holt's two parameter methodology was applied to the Brévilles dataset for the forecasting of atrazine, deethylatrazine, calcium and nitrate fluxes in spring water (Deliverable T2.4). Although the exponential smoothing process used in the Holt's methodology is a predictive technique which is used widely, its use to forecast fluxes in spring water is an ill-posed problem, i.e. an error on the estimation of one given parameter may induce very different forecasts, without being able to certify a value is more likely than another. In all cases, the forecast strongly depends one particular parameter whose incidence on the fitting of the model is very low, which is a typical characteristic of ill-posed problems. Clearly, in the case of the Brévilles spring data, more information has to be taken into account into the model to produce a reliable forecast. Moreover, the uncertainty of predicted fluxes and concentrations has to be estimated according to the external conditions responsible for the transfer from the soils to the spring. For these reasons, it is believed that conventional statistics are somewhat limited and that more advanced techniques such as those which were deployed at a later stage (see below) should be preferred.

3.2.3 Development of an innovative methodology combining fuzzy logic and artificial neural networks for trends analysis

Methodological developments were undertaken to allow uncertainty in predicted trends to be assessed by combining possibilistic regression approaches with artificial neural networks (Deliverable T2.4). The new methodology was tested using time series for atrazine, deethylatrazine and spring discharge data from the Brévilles case study. Various case studies were considered based on the level of information fed to the system for training purposes. These were: i) Time of measurement (week number); ii) Measurement month number; iii) the annual supply of atrazine in the fields; and, iv) precipitation data. The methodology was found to be strongly influenced by assumptions built in neural networks and in parameters, which is due to some extent to the limited availability of measured ("training") data. The most appropriate results were expected for cases where informative data such as precipitation would be fed into the training process. Predictions were found to be extremely peaky, suggesting that the system does allow for little buffering of the rainfall data. Although the methodology has the potential to improve on classical non-linear regression using neural network schemes by providing estimates of the imprecision on the regression, application to the Brévilles data showed that it was inadequate in practice for time series typical of monitoring programmes. A limitation factor is also that the technique does not allow for existing knowledge to be fed into the system.

3.2.4 Use of a transfer-function approach using the TEMPO software

TEMPO is a software which allows time series to be modelled based on transfer function approaches. The system requires very few input data and has been shown to provide predictions of interest in the past with regard to water quantity issues. As part of the TREND2 module, methodological developments were undertaken to allow the application of TEMPO to the prediction of fluxes and concentrations of degrading pollutants such as pesticides. Initial methodological and results are presented in Deliverable T2.4, while the latest results have been submitted by Pinault & Dubus for publication in the Journal of Contaminant Hydrology. The methodology is based on signal transfer analysis, which originates from the field of electronics and has been used in hydrology for a number of years. The main advantage of TEMPO is that it requires very little data to operate, typically monitoring data and information of effective precipitation (i.e. the water actively contributing to the recharge of groundwater).

TEMPO was found to provide a good fit to the Brévilles data for water, tracer and pesticide variables, even if the calibration period was limited and in any case less than the expected response time of the system. The generation by TEMPO of historical information regarding former atrazine application on the catchment was found to match existing knowledge drawn from farmer surveys. Based on these promising results, TEMPO was used to predict trends in water quality based on a large number of randomly-generated climatic series. Predictions for these different statistical realizations were found to be consistent and suggest i) a steady decrease of atrazine concentrations at the spring (Figure 2); and, ii) a stability of its degradation product DEA at levels of ca. 0.6-0.7 µg/l over the foreseeable future (Figure 3). Although TEMPO cannot capture very abrupt changes in concentrations, the software appears to provide a very good fit to the overall trends.

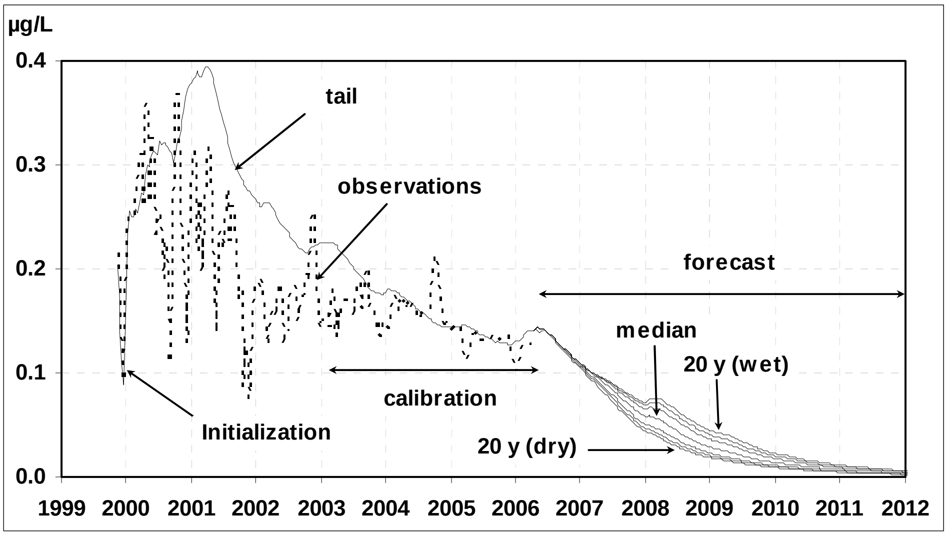

Figure 2. Predictions of the TEMPO software for concentrations of atrazine at the Brévilles spring.

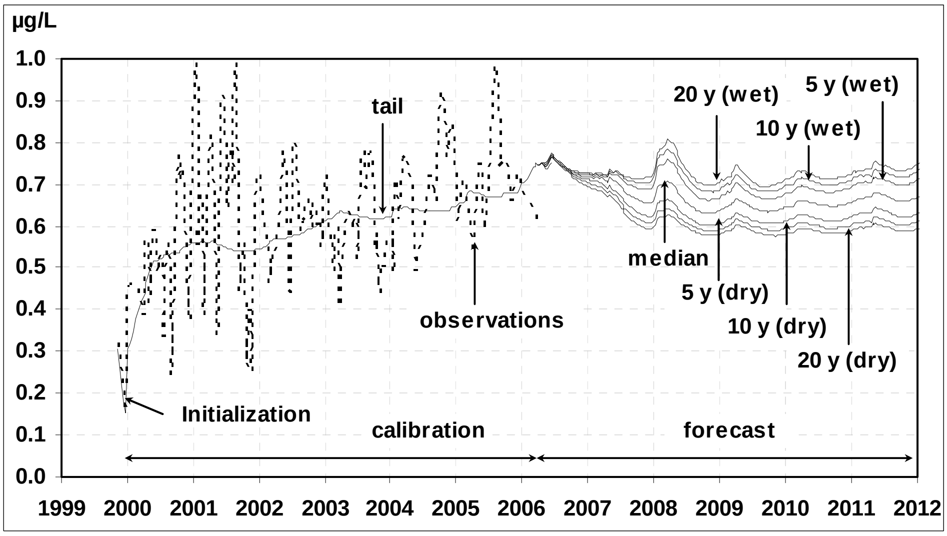

Figure 3. Predictions of the TEMPO software for concentrations of the atrazine metabolite DEA (De-Ethyl Atrazine) at the Brévilles spring.

3.2.5 Deterministic modelling using a combination of two leaching models

Initial modelling efforts within AquaTerra were aimed at creating a 3D unsaturated/saturated zone modelling of the Brévilles hydrogeological system. The complexity of the system under study revealed such that this was not considered an appropriate approach and an alternative methodology based on the combination of a 1D unsaturated (MACRO) and a 2D saturated (MARTHE) zone model was therefore developed. The input data, the modelling approach and first results have been presented in earlier TREND2 deliverables, in particular Deliverable T2.5, and T2.7.

Although modelling efforts are still underway (Deliverable T2.10 presents the latest modelling results available), inferences regarding the merits and shortcomings of the modelling can already been drawn. The modelling results at this stage are promising with regard to the capability of the modelling to describe the system under study. However, the modelling relies on numerous supporting data and extensive calibration activities, in contrast to the deployment of the TEMPO software presented above. Predictions regarding future trends in pesticide concentrations at Brévilles will be presented once calibration activities yield results considered to be adequate. Those will be based on predicted meteorological time series originating from the HYDRO module.

3.3 Merits and shortcomings of each trends analysis technique deployed at Brévilles

A total of five approaches to trends analysis were deployed at Brévilles to simulate and predict water fluxes and concentrations of agricultural pollutants (nitrate, atrazine and its metabolite DEA) in piezometers and the spring.

Approaches based on the application of classical statistics to the Brévilles dataset were found to be inadequate for predicting trends in the catchment due to that fact that they rely on examination of the time series only and do not account for external factors such as pollutant inputs on the catchment or rainfall amounts.

Methodological developments were undertaken to combine artificial neural networks with fuzzy logic in a possibilistic regression framework to allow uncertainty in predictions to be assessed. Although the methodology is attractive, its implementation in the real world is difficult and subject to significant subjectivities. Also, the methodology does not allow for existing knowledge about the system to be integrated, thereby reducing its potential usefulness.

The trends analysis software TEMPO which relies on transfer functions provided a good fit to the data as well as interesting insights into the functioning of the Brévilles groundwater system. The main advantages of the approach are that it requires very few data to operate and it is relatively quick to deploy. The main inputs to TEMPO are the monitoring data supporting the trends analysis and information regarding the main drivers of the system (typically precipitation data in the field of hydro(geo)logy) which can usually be readily obtained.

Deterministic modelling based on the coupling of the MACRO and MARTHE software was undertaken. The calibration activities of the combined models are expected to provide valuable information into the functioning of the system under study and can be considered a valuable resource tool. However, the extensive calibration activities required to achieve a good fit to the data mean that the modelling problem is probably too time intensive and associated solutions and parameter estimations are probably non-unique and therefore subject to much uncertainties. This is likely to affect the confidence, which should be attributed to results coming out of the modelling exercise. Also, the achievement of a successful fit to the data requires a very significant investment in data collection in the field (to characterize environmental and hydrological conditions in detail) and in modelling activities. Although this significant investment can be justified within the context of research programmes aimed at furthering the understanding the transport of contaminants to water resources, its justification is more questionable for operational goals such as those of the Water Framework Directive. Such detailed and complex studies can clearly not be deployed for each of the several thousands of water bodies across Europe.

4 Applications of trend analysis techniques in the Netherlands (TNO)

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4.1 Supporting dataset

For the work performed by TNO/UU in the Work package Trend 2 of AquaTerra (Trends in groundwater quality), the following data were used. Several sources of data were used to compile the historical concentrations in recharging groundwater for the province of Brabant: estimates of atmospheric deposition by the meteorological service and several individual studies, economic figures on land use and the amount of livestock present in the province from the national statistics bureau, several individual studies into the composition of manure (T2.2). 34 time series of concentrations of major cations and anions and selected heavy metals were available, for the period 1980 or 1991 to present, sampled from monitoring wells with short (2 m) monitoring screens at 2 depth levels: shallow (8 – 10 m) and deep (20 - 25 m) below surface (T2.3: §2.3). A new data set of groundwater dating samples was collected and analysed. It consisted of 34 samples for $^{3}H/^{3}He$ analyses, CFCs (11, 12 and 113) and SF_6. (T2.3). The three-dimensional reactive transport model used in this project is the Integrated Transport Model, developed by TNO and KIWA. (Vander Grift et al. 2001) It combines a stationary groundwater flow model (MODFLOW) with a three-dimensional multi-species transport model (MT3DMS). (T2.5)

4.2 Work undertaken and main results

4.2.1 Reconstructing historical concentrations in recharging groundwater (T2.2)

Trends in recharge concentrations of agricultural contaminants were derived from historical records of land use and manure practices. The estimates were not precise and were only able to predict trends on a larger temporal scale for the entire province of Brabant. In general, these figures show an upward trend in agricultural contaminants related to the application of manure, such as for example nitrate, sulphate, and zinc, up to the 1980s, and a decreasing trends starting in the 1990s. (T2.2: section 2.8)

4.2.2 Concentration-depth, concentration–time and groundwater age-depth profiles (T2.3)

Concentration-depth and concentration-time data were analysed for trends. By constructing conservative chemical indicators, such as oxidation capacity, geochemistry is removed as a complicating factor for trend detection. Oxidation capacity (OXC) is the weighted sum of nitrate and sulphate concentrations and behaves conservatively when denitrification in the subsurface is caused by pyrite oxidation. (T2.3: section 2.4)

In homogeneous recharge areas, conservative contaminants or conservative chemical indications (such as OXC), the trends in recharge concentrations will appear as (somewhat distorted) vertical profiles of concentrations, because travel time may be assumed to increase with depth and geochemistry does not affect the downward transport. LOWESS smooth through the depth profiles may reveal the trend present in the large scatter of data. (T2.3: section 2.5)

Concentration-time graphs are useful when the depth of the monitoring screens can be used to filter out some variation in groundwater travel times, for example by separating "old" (deep) from "young" (shallow) groundwater. A LOWESS smooth through all time series gives an idea of the trend in the groundwater quality, but is very sensitive to missing cases and should ideally only be applied to complete datasets. (T2.3: section 2.6)

Trends were determined on the individual time series, providing statistical significance (in the form of Kendall-τ correlation coefficients of Kendall-Theil trend slopes) and significant trends were found in the conservative chemical indicators OXC and the sum of cations: increasing in the deep screens and decreasing in the shallow screens. (T2.3: section 2.7)

Combining concentration depth profiles with (significant) trends in time gives the best insight into the status of the groundwater body and the present-day trends in groundwater quality.

For a more thorough discussion on trend detection and a concise summary table we refer to AquaTerra Deliverable T2.3, section 2.8.

4.2.3 Groundwater dating (T2.3: section 2.9)

We analyzed the data of groundwater age samples of $^{3}H/^{3}He$, CFCs and SF_6 from 34 screens, by comparing the estimated ages as well as the internal checks for consistency they provide. $^{3}H/^{3}He$ provides a check on the quality of the data, as well as the undesired subsurface processes (such as degassing) that might have affected the sampled groundwater, by simultaneous measurement of ^{4}He and Ne. Also, the history of ^{3}H in precipitation provides a check on the groundwater age estimate. CFCs and SF_6 provide an internal check by means of the ratio between the individual CFC species and the SF_6 concentration. These do not clearly show the effect of degassing, contamination or degradation of one of the tracers, but inconsistency does indicate poor data quality.

We found that although $^{3}H/^{3}He$ suffers from degassing, this can be properly corrected for (Visser *et al.*, 2007a) and it provides the most accurate and reliable groundwater age estimate. The cost of the analysis of a $^{3}H/^{3}He$ sample is about 450 EUR.

4.2.4 Demonstrating trend reversal (T2.4: Chapter 2)

Groundwater dating – for example by $^{3}H/^{3}He$ – is an excellent novel tool to detect trends in aggregate monitoring data for larger groundwater bodies. By transposing monitoring time series and relating measured concentrations to the time of recharge we could demonstrate trends and trend reversal in groundwater quality – indicated by the concentrations of nitrate, oxidation capacity and sum of cations – in the Dutch part of the Meuse basin (Visser *et al.*, 2007b). Also the concentration - recharge time relationship may be used to validate the estimated historical concentrations in recharge concentrations and improve the methods by which it was obtained and a more accurate extrapolation of present day trends may be obtained using future land use predictions. Because the travel time at some monitoring locations may be as high as 50 years, changing land use practices to reduce the pressure of agriculture on the groundwater system will not be observed before present day groundwater reaches these monitoring screen. As a result, improvement of groundwater quality in the entire groundwater body . will take several decades.

4.2.5 Physically-deterministic determination and extrapolation of time trends (T2.10: Chapter 2)

A physically-deterministic model was used to determine trends in groundwater quality and predict future. The physically-deterministic model was built for a part (34.5 × 24 km) of the groundwater body (Figure 4.1). A deterministic model can actually predict future trends, rather than extrapolating present day trends. For example, the deterministic model predicted an increasing trend in the zinc concentration in the shallow monitoring screens between 2010 and 2020 caused by the retarded breakthrough of the zinc contamination front. However, no trend was found in the monitoring data to date, nor was a trend predicted for the period up to present by the model. Only a deterministic model may be able to predict such future trends.

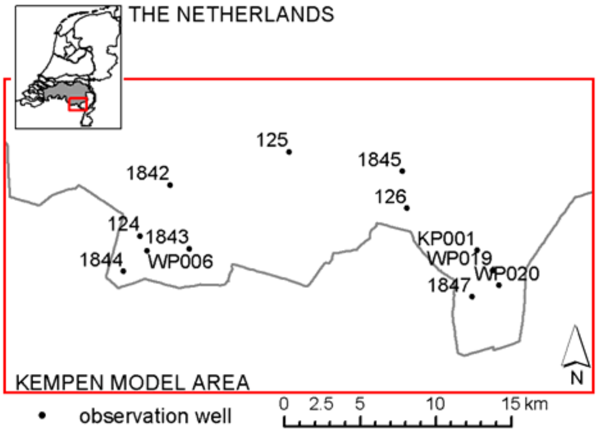

Figure 4.1: Location and extend of the model area, as well as monitoring locations within the model area.

4.3 Merits and shortcomings of each trends analysis technique deployed by TNO

For Work package Trend 2 of AquaTerra, TNO and UU worked on determining trends in groundwater quality in the sandy groundwater body “Maas” within the Meuse catchment. The groundwater body is an unconsolidated granular aquifer with a rather homogeneous geology, consisting of periglacial sands and delta deposits with more clay rich sediments. Although the fine scale geology may be quite heterogeneous, on the larger scale this can be considered a homogeneous sandy groundwater body. Groundwater flow mostly occurs in local and regional drainage systems with a shallow (1-5 m) unsaturated zone. In areas with less drainage and deeper infiltration the net precipitation contributes to recharge of larger and deeper groundwater systems.
In this system, little mixing of older and younger water occurs. Samples taken from monitoring wells with short monitoring screens (2 m) in this groundwater system probably have a distinct travel time.
We used an extensive data set of time series since 1991 of concentrations of agricultural contaminants from 15 locations screened at 2 depths. The data set showed large year-to-year variation, thus complicating trend detection. This size of data set was needed to smooth the year-to-year variation and also to provide measurement support over 4 to 5 decades in the past.

The first and easiest analysis of groundwater quality data is the detection of trends of concentration with (sampling) time. This can be performed, either visually on the entire data

set, or statistically on individual time-series, to be aggregated to a groundwater body average trend.
The wells selected for this project are located in regional recharge areas with little to no drainage within a few kilometres from the monitoring well. In these circumstances, the travel time of groundwater may be assumed to increase with depth, and concentration depth profiles will provide more insight into the downward transport of diffuse contamination. Selecting monitoring wells in recharge areas is recommended also because the shallow screens will then provide an early warning of the downward movement of contaminants into the groundwater system. Combined with statistical trends with time, concentration-depth profiles can provide the best overview of the status of the groundwater body as well as the trends in groundwater quality.
The data-driven trend analysis may be extended by incorporating groundwater travel times. The groundwater dating methods today are best suited for groundwater systems like the one under study, having a thin unsaturated zone and little mixing, for example by fractures, large heterogeneities or long sampling screens. Nevertheless, problems that may be encountered using groundwater dating in these types of aquifers are:

- regional contamination of CFCs
- degradation of CFCs in anoxic environments
- degassing of $^3H/^3He$, CFCs and SF_6 (caused by denitrification and N_2 production, or other geochemical processes producing large amounts of gas)

We recommend using $^3H/^3He$ dating in well suited groundwater systems, because it has the best internal checks on data consistency; or as many groundwater age tracers as possible in more complex systems, each providing possibly more information.

Groundwater dating removes travel time as a complicating factor for trend analysis, and trends in groundwater quality will be determined only by historical land use and geochemistry. In this case, groundwater dating revealed trends and trend reversal in groundwater quality by sorting measurements to the recharge year of the sampled water. We would recommend this method of trend analysis if groundwater dating is possible and time series from monitoring wells are available. Groundwater dating greatly increases the value of existing groundwater quality data sets.
The best data-driven predictions for future (extrapolated) trends in groundwater quality are obtained by combining the relationship between concentrations and recharge time with land use history and predictions of future land use. However, these extrapolated trends are limited to conservative contaminants. Reactive contaminants will show non-linear behaviour that is not reproduced by the linear extrapolation of the concentration - recharge time relationship.

A physically-deterministic model predicts the most reliable future trends in groundwater quality, especially for reactive contaminants. Building a physically-deterministic model for a part of the groundwater body was only possible because a large amount of data on geology, hydrology and land use was available. Physically-deterministic models do not rely on the presence of trends in available data sets, and are capable of predicting the breakthrough of contamination fronts that have not yet reached the monitoring screens in the study area. If a 3D reactive transport model is not possible, combined stream tube and 1D reactive transport modelling may provide a simpler solution. Future work should investigate the possibilities of such an approach, compared to the existing 3D reactive transport (MODFLOW+MT3D) model.

5 Applications of trend analysis techniques in Belgium (ULg)

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5.1 Short description of the dataset

At the beginning of the project, four groundwater basins were selected for nitrate trend analysis in the framework of WP TREND T2 (Figure 4). Nitrate data concentrations used in this study come mainly from the Nitrate Survey Network established by the Walloon Region Government. In this network, boreholes, springs, galleries and traditional wells are considered as monitoring points, where sampling and water analyses are carried out regularly. This network provides a spatial and temporary representation of nitrate contents in the aquifers. Nevertheless, gaps exist in the datasets and in the spatial distribution of monitoring points. During the first months of the project, contacts were also established with the VMW (Vlaamse Maatschappij voor Watervoorziening), the Flemish water supply company, in order to have access to data and to water supply wells in the North of the Hesbaye groundwater body (included in the Geer basin), where the chalk aquifer becomes confined, in continuity with the free part of the aquifer which corresponds to the Walloon Region territory. A complete description of the nitrate dataset is presented in Deliverable T2.1 and T2.2. Most time series selected for this study covers a period of ten years with some of them covering more than forty years. The number of data in the time series of nitrate concentrations can vary greatly, ranging from a few tens to more than a thousand data points. The number of points used for the trend analysis in the different groundwater bodies is summarized in Table 2.

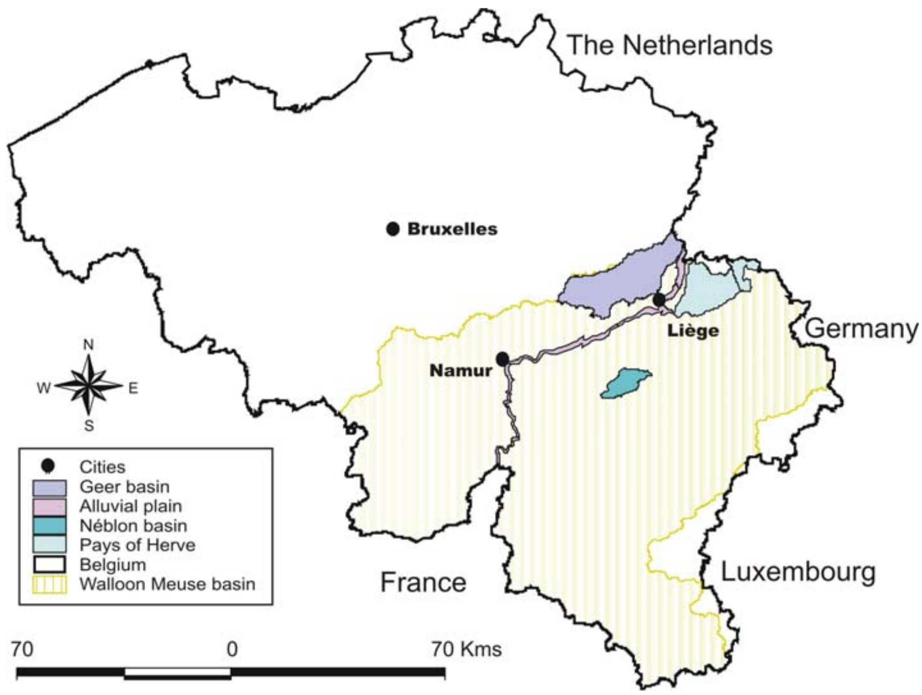

Figure 4: Location of selected groundwater bodies

	Observation points from the nitrate survey network	Other observation points (with nitrate data sets)	Total of observation points
Geer basin	27	5	32
Pays Herve-	38	21	59
Néblon basin	6	0	6
Alluvial plain of the Meuse river	24	23	47

Table 2: Summary of observation points in the selected groundwater bodies.

In the second part of the project, research efforts have focused on one of the groundwater bodies, the Geer basin. This basin, located in the North-East of the city of Liège is a groundwater resource of major importance for the city of Liège and its suburbs. The aquifer is located in a chalky layer of several meters where groundwater is extracted using drainage galleries and wells, mainly by local drinking water supply companies. The chalk is overlain by a thick loess deposit.

Due to the relatively flat topography and to the existence of the loess deposit, the Geer basin is predominantly covered by agriculture: crops cover 66% of the catchment area, the remaining space being divided between pastures (17.5%), housing (10%), forest (2.5%) and others (4%). For many decades, the use of fertilizers in agriculture has allowed to increase the farm yield. Unfortunately, an excessive or inappropriate use of these substances has led to pollution of surface water and groundwater. According to the characterisation report of the Meuse district in the Walloon region (DGRNE, 2005), the Geer basin aquifer is the most affected groundwater body by agricultural pressures in the Walloon region. Moreover, according to Dautrebande and Sohier (2004), the respective contributions of diffuse agricultural and dispersed domestic sources in the Geer basin regarding to nitrogen losses are 88% and 12% respectively. It is thus important to understand the impact of agriculture practices on groundwater. HG-ULg has been involved for years in different projects to study this aquifer. A detailed description of the Geer basin, available datasets and a synthesis of the previous studies can be found in the Deliverable R3.16.

5.2 Presentation of the work done and key results

Work done by HG-ULg project can be divided in two parts. Firstly, HG-ULg has developed a methodology using statistical approaches to infer trends in nitrate groundwater quality for four selected groundwater basins of the Meuse River. Secondly, HG-ULg has developed a groundwater flow and solute transport model as a prediction tool for trend analysis in one of these sub-basins, the Geer Basin.

5.2.1 Statistical trend analysis

A three-step consistent and rigorous approach (Figure 5) has been proposed and applied for trend detection and quantification in groundwater quality (nitrate) datasets, based on a three steps statistical analysis methodology (Deliverable T2.4; Batlle Aguilar J., Orban P., Dassargues A. and Brouyère S. (2007). Identification of groundwater quality trends in a chalk aquifer threatened by intensive agriculture in Belgium. Hydrogeology Journal 15(8): 1615-1628.):

- the first step is to evaluate whether the dataset is normally distributed;
- the second step consists in detecting whether a trend exists in the dataset;
- the third step is the actual trend estimation or quantification.

This methodology has been applied for the Geer basin, but also for three other groundwater bodies in the Walloon part of the Meuse basin. Some general observations can be drawn from the point-by-point results obtained for the four groundwater bodies selected in the Walloon part of the Meuse basin. The statistical approach seems robust and able to discriminate between “clear” and “weak” trends.

For some datasets, results of the normality tests were not univocal. However, whatever the trend detection and quantification method applied to these datasets, conclusions were very similar. Even if, from a pure statistical point of view, the normality of the dataset is a factor to be considered for selecting one or another trend analysis technique, from a practical point of view, the result of the analysis is not so sensitive to the distribution of the dataset.

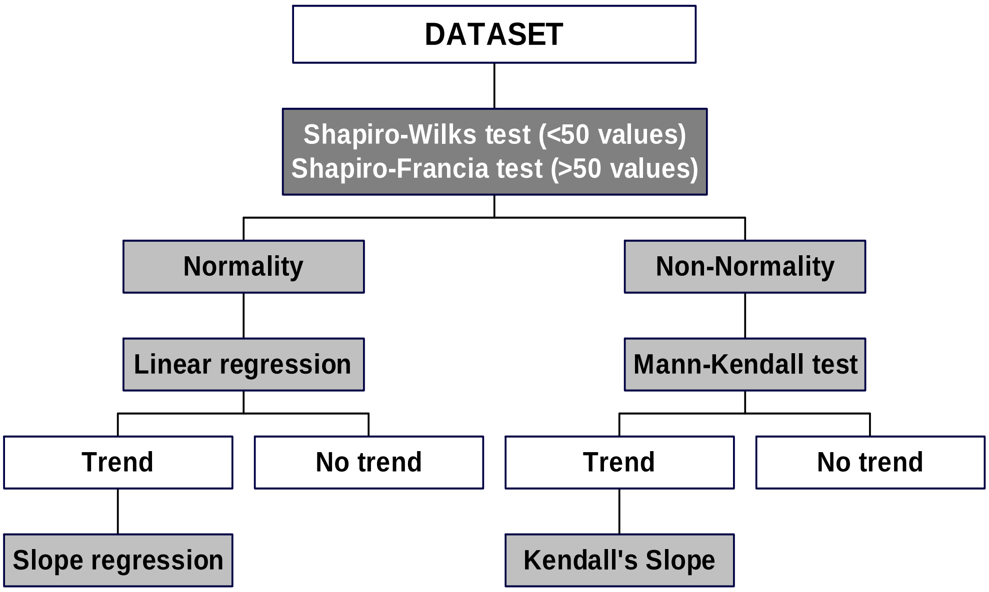

Figure 5: A three step procedure is adopted for trend analysis of nitrate concentrations in the selected groundwater bodies: 1) normal/non-normal distribution data; 2) trend detection; 3) trend estimation.

The statistical analysis provided point-by-point estimates of nitrate trends, in the form of slopes expressed in mg NO3/year (increase or decrease). In the Geer basin, a general upward trend is observed in the entire basin. However, two zones can be distinguished in the basin: the Southern part corresponding to the unconfined part of the chalk aquifer where high nitrate concentrations are observed, and the Northern part corresponding to the confined part of the aquifer, where nitrate has not been detected (or at very low concentrations only) (Figure 6).

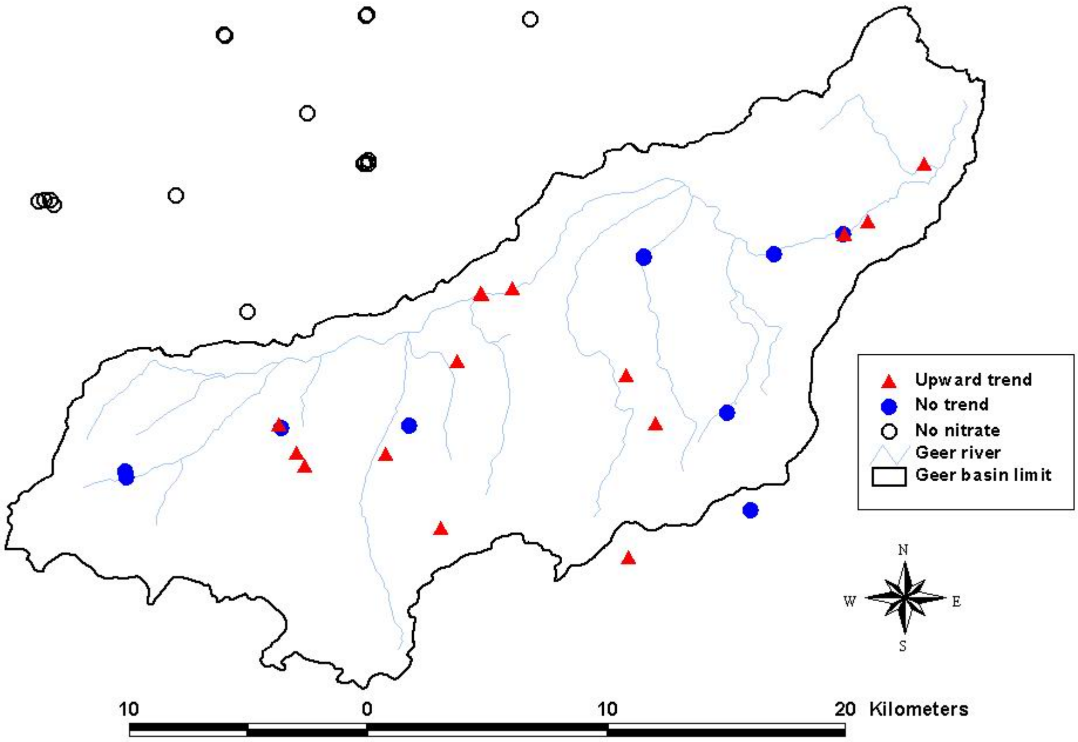

Figure 6: Spatial trend distribution in the Geer basin.

Results obtained with the “pure” statistical analysis might be enough and appropriate to estimating the short term evolution of groundwater quality (few years), particularly for the Geer basin overlaid by a thick unsaturated zone that lead to important buffer effects in the evolution of nitrate concentrations in the aquifers. In order to use the methodology developed by TNO, efforts have been made to link the nitrate concentrations in the groundwater and the age of groundwater using tritium data recently acquired (Deliverable T2.8). However, the computation of the age of water is not easy due to the heterogeneity of the chalk, the variable thickness of the unsaturated zone…

However, end-users and decision makers such as regional authorities and water companies are more interested in the long-term evolution of groundwater quality (tens of years). At such a time scale, the main driver of nitrate trends in groundwater becomes land use. In that context, the major disadvantage of using a “pure” statistical trend analysis is the fact that it is not able to consider variations in land use and functional relations between land use and groundwater quality.

5.2.2 Deterministic modelling

In a second step, HG-ULg has developed, in collaboration with WP BASIN, a spatially distributed physically-based deterministic groundwater flow and solute transport model for the Geer basin, at regional scale. A new concept for large-scale solute transport modelling, approach, the Hybrid Finite Element Mixing Cell technique (HFEMC) developed by HG-ULg and implemented in the 3D simulator SUFT3D (Deliverable R3.18) has been used to develop this model. Available data and conceptual choices made to develop the model were presented in Deliverable R3.16, R3.18, T2.8 and T2.10.

As mentioned in deliverable T2.8, water age is of crucial importance for the understanding of diffuse pollutions. The spatial distribution of groundwater age is indeed a key factor determining the distribution of solutes in groundwater. In parallel of the study of the nitrate pollution in the Geer basin, HG-ULg has taken samples for tritium analysis.

Two datasets were used for the calibration of the transport model, one corresponding to tritium data acquired during winter 2004-2005 (deliverable T2.8), the second to trends in nitrate groundwater quality (Deliverable T2.4). The advantage of tritium is that the concentration in the infiltration can be easily estimated because it is essentially a function of latitude. Nitrate concentrations in the infiltration are more difficult to determine as they are function of land use and there time evolutions (Deliverable T2.2). As spatially distributed estimation of nitrate fluxes as computed with the EPIC-Grid model (UHAGX, Deliverable T2.2) are not yet available, a simplified uniform scenario of input was defined.

The computed nitrate concentrations are of the same order of magnitude as the observed one (Example on Figure 7). The general increasing trend is reproduced by the model. A more detailed comparison between the modelling results and the statistical trend analysis results is not so relevant since the nitrate input applied to the model remains relatively arbitrary, at least affected by a large uncertainty. Nevertheless, results presented here prove however the capability of the model to reproduce nitrate concentrations and their time evolutions.

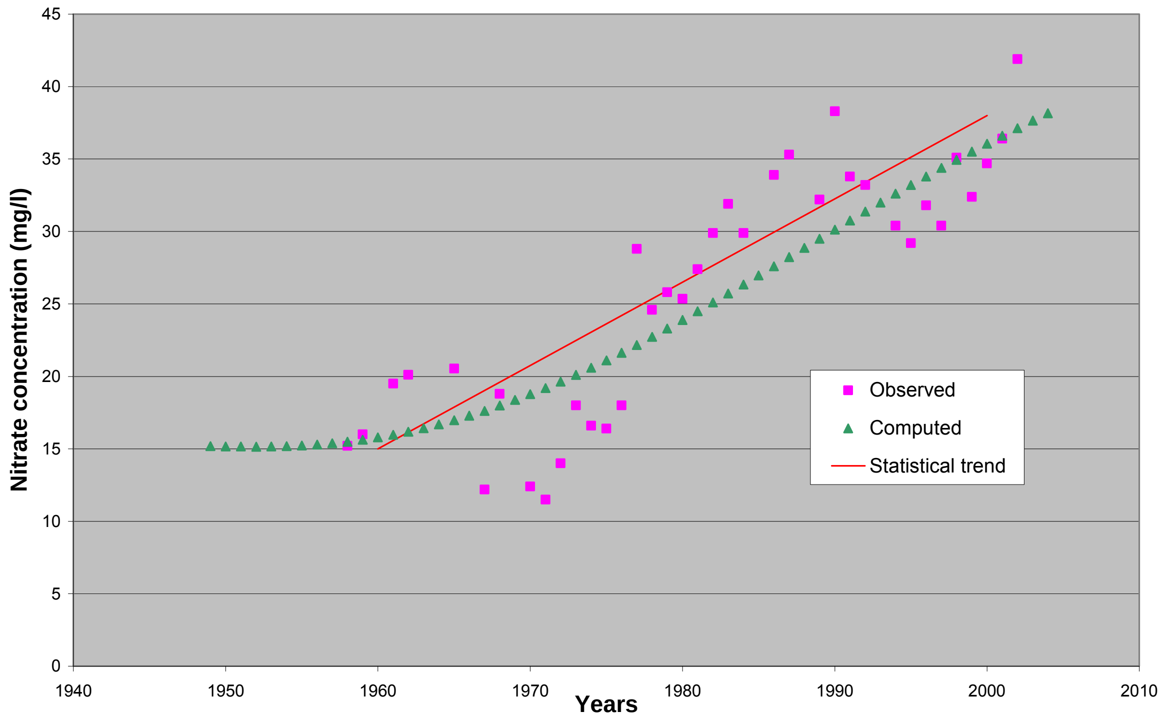

Figure 7: Comparison between the computed and observed time evolution of nitrate concentration in the well H7

Two contrasting scenarios of nitrate input (from 1950 to 2050) have been tested to assess the time evolution of nitrate concentrations in the groundwater (Deliverable T2.10). In Scenario 1, the nitrate concentrations increase from 1950 until the beginning of the 1990s and then remain constant. In Scenario 2, the nitrate concentrations in the infiltration increase from 1950 until the beginning of the nineties and then are set to zero after 2010. As an example, prediction of nitrate concentrations computed for three points located along a North-South oriented transect (for more details see Deliverable T2.10) in the Geer basin are presented Figure 8.

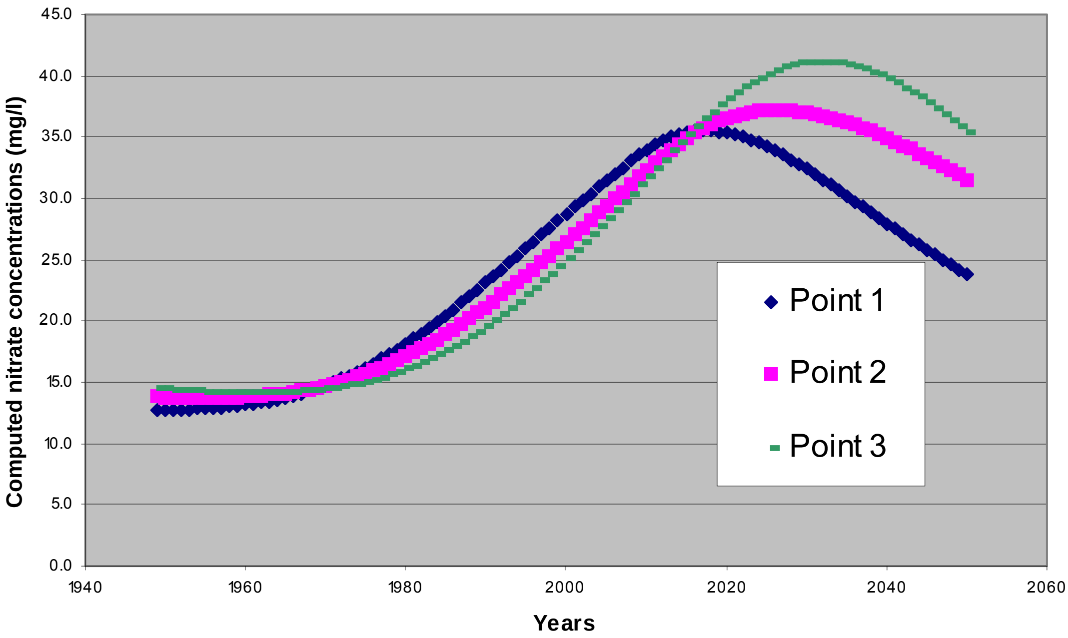

Figure 8: Time evolution of the nitrate concentrations computed with Scenario 2.

5.3 Discussion regarding the merits and shortcomings of each methodology deployed by ULg

The strengths and the weakness of the two trend analysis methodologies developed by HG-ULg are analysed with regard to (1) data requirement, (2) time consumption, (3) potential for trend prediction, (4) reproducibility of the methodology.

5.3.1 Data requirement

For statistical trend analysis, only time series of nitrate concentrations in groundwater are required. However, in case of "seasonal" (period) effects, the time series have to be long enough to include data representing more than one period of variation.

On the contrary, the development of a spatially distributed physically-based groundwater flow and solute transport model requires large and detailed datasets including information on the geometry of the aquifer, hydrodynamic and hydrodispersive parameters, time series of water levels and nitrate concentrations and fluxes of water and solute to groundwater. Such detailed information is available only for groundwater bodies that are characterized in detail and studied for years.

5.3.2 Time consumption

Once the time series of nitrate concentration are collected, time and efforts required to statistically identify and quantify trends are low due to the simplicity of such techniques. On the contrary, time and efforts required for developing a reliable groundwater flow and solute transport model are more important. Meshing procedure and implementation of the data can be time consuming. Time needed for calibration can also be important depending on the required degree of confidence in the model.

5.3.3 Potentials for trend prediction

Results provided by the statistical trend analysis might be sufficient and appropriate for assessing objectively trends in the dataset and for estimating the short term evolution of groundwater quality (few years), particularly for basins overlain by a thick unsaturated zone that lead to important buffer effects in the evolution of nitrate concentrations in the aquifers.
However, the interest of the different end-users is more focused on the long term evolution of groundwater quality (tens of years) and geological and hydrogeological factors are not the only drivers of nitrate trends in groundwater: land use is also a key factor. The major disadvantage of using a “pure” statistical trend analysis is that it is not able to consider variations in land use and functional relations between land use and groundwater quality. For long-term evaluation of nitrate trends, more advanced techniques are required, such as mechanistic modelling relating land use and physical and biogeochemical processes occurring in the underground to groundwater quality.

On the contrary, capability of the groundwater flow and solute model to reproduce long-term temporal evolution of nitrate concentrations in groundwater has been proven. The only requirement is to have reliable predictions of the temporal evolution of nitrate fluxes to the groundwater and a reliable model. As illustrated in the previous section, the model is also able to predict trend reversal.

5.3.4 Reproducibility of the methodology

The statistical methodology of trend analysis can be applied on any kind of time series of data long enough to include more than one period of periodic variation.
As the development of the model requires more data and thus more costs for data acquisition, modelling is more adapted in well-studied basins or in basins of high ecological or economic importance.

6 Discussion

A total of five approaches to trends analysis were applied at various sites in France, the Netherlands and Belgium as part of the TREND2 work package of the AquaTerra project. The experience gained through these various activities allows conclusions to be drawn regarding the general applicability of the various approaches for assessing trends in concentrations of pollutants in water resources in the European Union. The discussion below on the comparative assessment of the various approaches used in TREND2 is supported by Table 3.

One of the main important points drawn from the comparative assessments is that there is **no unique approach, which enables the satisfactory description and prediction of trends across widely-differing catchments**. This is due to differences in catchment characteristics and data availability, but also to differences in expertise of scientists with one particular approach or the other. The most striking example within TREND2 was the potential use of age-dating techniques which proved suitable for age-dating -and hence trends forecasting- in the Netherlands where subsoil materials are generally fairly homogeneous while age-dating was considered to be of little operational use in the fractured aquifer of Brévilles where mixing of water of different ages occur.

'Classical' time series statistics such as ARIMA were applied to the various data series in TREND2. The advantages of these statistics is that i) they do not require the collection of data other than those constituting the series under study; and, ii) they help objectify the detection of trends. Results in terms of overall description of data were mixed with groups obtaining relatively good fit to their data (ULg) while others could not obtain satisfactory fits (BRGM). This is believed to be due to differences in the characteristics of the hydrogeological systems under study. The general feeling among the various groups involved was that classical statistics were of little operational use given that they did not account for variables, which are known to significantly affect pollutant concentrations in water systems such as timing of pollutant inputs and meteorological characteristics.

The non-inclusion of driving factor in the analysis is considered a major limiting factor for both the description and the prediction of trends in water quality. However, classical time series statistics may still have a role to play at an early stage of trends investigations since they are cost-effective and could be used to decide on the need to proceed further with field investigations and mechanistic modelling.

Innovative approaches combining fuzzy logic and artificial neural networks were developed and applied to the Brévilles dataset. The rationale for such combination is the derivation of information regarding the uncertainty in trends prediction. Although the approach is of potential interest, its practical applicability was found to be of limited value for systems where monitoring data spanning over a few years only are used. The fact that neural networks are used means that no inference regarding the functioning of the system can be made and that no information regarding pre-existing knowledge can be fed into the system. For example, the fact that applications of atrazine were discontinued from 1999 in the Brévilles catchment could not be integrated into the analysis. Given these limitations and the lack of overall applicability of these new developments, the approach has not been listed in Table 3.

Age dating techniques were used by all TREND2 partners. In contrast to statistical analyses described above, the methodology requires the availability of a monitoring system allowing the robust collection of water samples at different depths, the sampling of water aliquots, their analysis in the laboratory and the gathering of information regarding pollutant inputs to the system. Although the establishment of monitoring networks allowing such sampling is costly if started from scratch, the availability of existing networks, e.g. in the Netherlands, means that the methodology can be applied economically in some instances.

A range of age-dating tracers were deployed by TREND2 partners: CFCs, tritium and $^3H/^3He$. Difficulties in the practical deployment of the methodology for CFCs were experienced by two of the partners. Although care was taken in the sampling and handling of the resulting samples, suspicion regarding contamination of the samples due to local, high, atmospheric mixing ratios were expressed by two partners. In addition, degradation of CFCs was evidenced under anoxic (pyritic) conditions in the aquifer of the Brabant catchment in the Netherlands.

A distinction was made within TREND2 between those geological systems which are fairly homogeneous and those with heterogeneous flows resulting from fractures and fissures. Age-dating techniques revealed very useful for trends prediction in the former case, e.g. when applied in the Netherlands. In contrast, the mixing of waters in fractured systems such as those encountered in large areas across France means that the age measured is a composite of various ages and that no real information regarding true age and future trends in water quality can be made unless mixing ratios can be disentangled. The extrapolation potential is therefore strongly dependent on the type of hydrogeological systems: from excellent in homogeneous systems to limited in heterogeneous settings.

Beyond the demonstration of trends in groundwater quality, age-dating using tracers in homogeneous groundwater systems is also a good tool to calibrate and test groundwater flow models (see below). The sampled concentration of tritium can be compared to predicted concentrations, or groundwater ages determined by $^3H/^3He$ can be compared directly to groundwater ages calculated by the model, either using particle tracking or direct simulation. These tests validate the accuracy of the numerical groundwater flow model.

A **transfer function approach** was deployed by two TREND2 partners in France and Belgium using the TEMPO software. The approach is intermediate between statistical and deterministic approaches in that it relies on the calibration of a transfer function, which expresses the delay in transfers of water and pollutants in the systems considered. The main advantage of the methodology is that it is very data parsimonious as it only relies on the data series under study, key driving variables (usually precipitation or effective precipitationin the field of hydrology) and on information about the historical inputs to the system. The approach is known to perform well in homogeneous settings and proved its worthiness in heterogeneous fractured aquifers through its application in two of the TREND2 case studies.

The TEMPO software is easy to use and could be potentially deployed throughout Europe to give information regarding future trends in water pollution in water bodies within the context of the Water Framework Directive.

Deterministic modelling activities were undertaken in France, Belgium and the Netherlands as part of TREND2. Interim results by the three research groups enable the drawing of information regarding the merits and shortcomings of the deployment of complex hydro(geo)logical models within a trends analysis context.

One of the main issues associated with deterministic modelling is the need to have a detailed characterization of the system under study available in terms of meteorology, soils, subsoils, hydro(geo)logy and use of pollutants on the catchment. This typically requires very significant financial and time investments in the field, which usually spans over several years. Although there is no dispute as to the usefulness of intensive catchment modelling activities from a research point of view, the 'return on investment' from an operational perspective is mainly dependent on whether a model can be successfully fitted to the data and whether the model has shown potential for supporting extrapolation and management activities. A distinction was therefore made in Table 3 between modelling efforts which provide successful fit to measured data and trends, and those where the fit to the data is considered to be beyond the standard for operational activities.

Some of the great advantages of undertaking deterministic modelling activities is that it brings together various sources of information collected in the field and that it allows the furthering of the understanding of the catchment functioning using both measured and predicted information. In some instances, the modelling successfully integrates the factors driving the behaviour of water flow and pollutant transfers and can therefore support robust extrapolation activities in space and time. Another advantage of deterministic modelling is that deterministic models are potentially capable of predicting future trends, rather than extrapolating trends in present-day datasets. They may predict trends which do not yet occur at present, for instance due to delayed breakthrough of sorbing contaminants.

The fact that the deterministic models rely on detailed measured data in the field can become a negative aspect when temporal extrapolations are being made as one has often little knowledge regarding the possible future evolutions of the key variables driving the model predictions. This can be exemplified for pesticides for which there is considerable uncertainty on: i) the future distribution of crops in the EU, which is likely to be driven by economical, strategic, political, environmental and phenomenological aspects; and, ii) the future types of products which will be on the market in, say, 20 years; iii) climatic characteristics in specific locations in Europe. It should however be noted that these limitations are not specific to deterministic modelling approaches.

Because of the simplifications that are needed to make large complex models feasible, concentrations are predicted with a large uncertainty at a short time scale and such models are therefore not well suited for predicting year-to-year variations. They are however capable of demonstrating trends in groundwater quality on longer time scales (decades), and also excellent tools to predict future trends in concentrations of contaminants, for example nitrate, metals or pesticides. Overall, it is concluded that the deployment of complex deterministic models provides useful insight into the functioning of the system and is therefore the most suitable approach in a research context. However, the very large financial, human resources and time investments associated with the collection of data and their integration into an overarching modelling exercise means that the deployment of deterministic models for operational analysis of trends across the EU is beyond reach. Such modelling activities should concentrate on areas of high ecological, sustainability or economical importance within the context of the Water Framework Directive.

	Purely statistical approaches	Transfer function approaches	Age dating	Deterministic modelling with poor fit to the data	Deterministic modelling with good fit to the data
Prerequisite	Collection of monitoring data in the field	Collection of monitoring data in the field + Collection of information on the input flux (rainfall, and either inputs or land use)	Collection of monitoring data in the field + information on the evolution of the input function + analysis of tracers in samples	Collection of monitoring data in the field + Heavy effort in collection of additional information (other piezometers, pumping and tracer tests, geophysics, soil mapping)	Collection of monitoring data in the field + Heavy effort in collection of additional information (other piezometers, pumping and tracer tests, geophysics, soil mapping)
Associated cost magnitude (on top of the data collection effort)	None	1 (surveys if not already available - purchasing of met data)	10	100 (geophysics, additional piezometers, soil mapping)	100 (geophysics, additional piezometers, soil mapping)
Understanding of the system?	No understanding of the system	Functional understanding of the system (identification of the key factors and understanding of their influence)	Functional understanding of the system	Detailed data on the system, but lack of overall understanding of the functioning of the system (exemplified by the lack of fit of the deterministic model)	Potential detailed understanding of the system under study
Extrapolation potential	Poor	Good	Good	Poor	Good
Potential universality to all systems	Potentially	Potentially	Limited. only applies to homogeneous systems.	Potentially	Potentially
Potential for operational use	-	++	+	-	+
Knowledge about the functioning of the system	-	++	+	++	+++

Table 3. Summary table comparing the strengths and weaknesses of each of the trends analysis methodologies deployed within the TREND2 module.

7 Summary and conclusions

A total of five approaches to trends analysis were applied in catchments in France, the Netherlands and Belgium as part of the TREND2 work package of the AquaTerra project. The experience gained through these various activities allows several conclusions to be drawn regarding the general applicability of the various approaches for assessing trends in concentrations of pollutants in water resources in the European Union.

1. 'Classical' time series statistics were applied to the various data series in TREND2. The general feeling among the various groups involved is that classical statistics are of little operational use given that they do not account for variables which are known to significantly affect pollutant concentrations in water systems such as the timing of pollutant inputs or meteorological characteristics. They may still have a role to play at an early stage of trends investigations since they are cost-effective and could be used to decide on the need to proceed further with field investigations and mechanistic modelling.

2. In groundwater systems with little heterogeneity, dating groundwater has proven a valuable tool to reveal trends in groundwater quality. By relating measured concentrations to the year of recharge of the water sample, travel time is removed as a complicating factor for trend detection. Trends found when relating concentrations to recharge year are then only determined by trends in historical inputs of contaminants and - for reactive contaminants - geochemical processes. The age-dating approach is however of little value in heterogeneous groundwater systems where mixing between waters of different ages occur. In addition, the approach relies on the availability of monitoring networks allowing the collection of samples at various depths and involves significant expenses for groundwater dating.

3. Transfer function approaches - which integrate information on key driving variables of the system under study - have been demonstrated to successfully describe the variations in time series for nitrate and pesticide concentrations within the context of TREND2. The TEMPO software package is very data-parsimonious and could therefore be readily deployed to assess trends in piezometric levels and pollutant concentrations in selected water bodies across the EU.

4. The deployment of complex deterministic models provides useful insight into the functioning of the system and is therefore the most appropriate approach in a research context. However, the very large financial, human and time investments associated with the collection of data and their integration into an overarching modelling activity means that the deployment of deterministic models for operational analysis of trends across the EU is beyond reach. Such modelling activities should concentrate on areas of high ecological, sustainability or economical importance within the context of the Water Framework Directive.

5. Decision-makers should be reminded that predictions of future trends in pollutant concentrations are subject to significant uncertainties due to i) the methodologies used; ii) the fact that predictions made regarding future inputs of contaminants to water systems or underlying driving variables (e.g. climate) are subject to significant uncertainty themselves.

6. The inherent uncertainty associated with trends detections and predictions in most groundwater systems means that monitoring activities to assess trends in water quality should remain a priority in Europe. Data collected can feed into trends analysis work which can itself provide valuable feedback on the optimisation of the monitoring strategy and on appropriate measures to address water quality concerns.

List of references

Batlle Aguilar, J., P. Orban, A. Dassargues and S. Brouyère (2007). "Identification of groundwater quality trends in a chalk aquifer threatened by intensive agriculture in Belgium." Hydrogeology Journal 15(8): 1615-1628.

Broers H.P, Visser A., Grift, B. van der, Dubus I.G, Baran, N., Morvan, X., Normand, M., Guttierez, A., Mouvet C., Batlle Aguilar J., Brouyère S., Ph. Orban, Dautrebande, S., Sohier, C., Korcz M., Bronder J., Dlugosz J., Odrzywolek M. (2005). AquaTerra Deliverables - Trend 2.2: Report with documentation of reconstructed land use around test sites.

Broers H.P, Visser A., Grift, B. van der, Gourcy L., Dubus I.G., Baran N., Mouvet C. & Gutierrez A. (2005). AquaTerra Deliverables Trend 2.3: Report on concentration-depth, concentration-time and time depth profiles in the Meuse basin and the Brevilles catchment.

Broers H.P, Visser A., Heerdink R, Van der Grift B, Surdyk N., Dubus I.G., Gutierrez A, Orban Ph., Brouyere S., 2007, Report with results of groundwater flow and reactive transport modelling at selected test locations in Dutch part of the Meuse basin, the Brévilles' catchment and the Geer catchment, Deliverable T2.8, AquaTerra project.

Broers H.P, Visser A., Pinault J.-L., Guyonnet D., Dubus I.G., Baran N., Gutierrez A., Mouvet C, Batlle Aguilar J., Orban Ph., Brouyere S., 2005, Report on extrapolated time trends at test sites, Deliverable T2.4, AquaTerra Project.

Broers H.P., Baran N., Batlle-Aguilar J., Bierkens M., Bronder J., Brouyère S., Dlugosz J., Dubus I.G., Gutierrez A., Korcz M., Mouvet C., Slowikowski D. and Visser A., 2004, Documented spatial data set containing the subdivision of the basins into groundwater systems and subsystems, the selected locations per subsystem and a description of these sites, available data and projected additional measurements and equipment., Deliverable T2.1, AquaTerra Project.

Broers H.P., Visser A., Heerdink, R., Van der Grift, B., Surdyk N., Dubus I.G., Amraoui N., Ph. Orban, J. Batlle-Aguilar, P. Goderniaux & Brouyère S. (2008). AquaTerra Deliverables - Trend 2.10: Report which describes the physically-deterministic determination and extrapolation of time trends at selected test locations in Dutch part of the Meuse basin, the Brévilles catchment and the Geer catchment.

Broers H.P., Visser A., Pinault J.-L., Guyonnet D., Dubus I.G., Baran N., Gutierrez A., Mouvet C., Batlle Aguilar J., Ph. Orban & Brouyère S. (2005). Aqua Terra Deliverables Trend 2.4: Report on extrapolated time trends at test sites.

Dautrebande, S. and C. Sohier (2004). Projet de recherche PIRENE, Rapport final., University of Gembloux: 120.

Orban Ph., Batlle Aguilar J., Goderniaux P., Dassargues A., Brouyère S., 2006, Description of hydrogeological conditions in the Geer sub-catchment and synthesis of available data for groundwater modelling, Deliverable R3.16, AquaTerra project.

Orban Ph., Brouyère S., 2006, Groundwater flow and transport delivered for groundwater quality trend forecasting by TREND T2, Deliverable R3.178, AquaTerra project

Visser, A., H. P. Broers, and M. F. P. Bierkens (2007a), Dating degassed groundwater with $^{3}H/^{3}He$, Water Resour. Research, 43, W10434, doi:10.1029/2006WR005847.

Visser, A., H. P. Broers, B. Van der Grift, and M. F. P. Bierkens (2007b), Demonstrating trend reversal of groundwater quality in relation to time of recharge determined by $^{3}H/^{3}He$, Environmental Pollution, 148, 797–807.