Project no. 505428 (GOCE)

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)	
RE	Restricted to a group specified by the consortium (including the Commission Services)	
CO	Confidential, only for members of the consortium (including the Commission Services)	

SUMMARY

The establishment of tools for trend-analysis in groundwater is essential for the prediction and evaluation of measures taken within context of the Water Framework Directive and the Groundwater Directive. Physically-deterministic modelling of groundwater and solutes is one of the methods tested by Trend 2. This report describes the physically-deterministic determination and extrapolation of time trends at selected test locations in Dutch part of the Meuse basin (TNO/UU), the Brévilles catchment (BRGM) and the Geer catchment (ULg).

The following trends were predicted by the physically-deterministic models:

- **in the Dutch part of the Meuse basin, nitrate and OXC concentrations will decrease in the near future under present day land use, but zinc concentrations will increase due to the leaching of zinc currently sorbed to the soil,**
- **in the Geer basin, nitrate concentrations will keep increasing at least up to 2050 under present day land use because of the long travel times in this system. Even when no new nitrate were applied, concentrations would increase over the next decade before decreasing,**
- **in the Brévilles catchment, atrazine concentrations will decrease in the coming years to levels below regulatory concern but the modelling needs further adaptation to ascertain whether this would also be the case for its main metabolite DEA.**

MILESTONES REACHED

T2.10: 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

The calibration of the models was performed based on data of groundwater levels acquired in collaboration with the BASIN sub program as well as climatic data acquired in collaboration with the HYDRO sub program.
These findings are interesting and important for the INTEGRATOR sub program to perform socio-economic analyses of the problems with the present day and future trends of the concentrations of contaminants in groundwater.
Future trends will assist decision makers of EUPOL to anticipate the effects of measures aimed at improving the quality of groundwater and surface water

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1 Introduction to TREND 2 (TNO)

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 upward trends and for the definition of starting points for trend reversal. Figure 1.1 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.

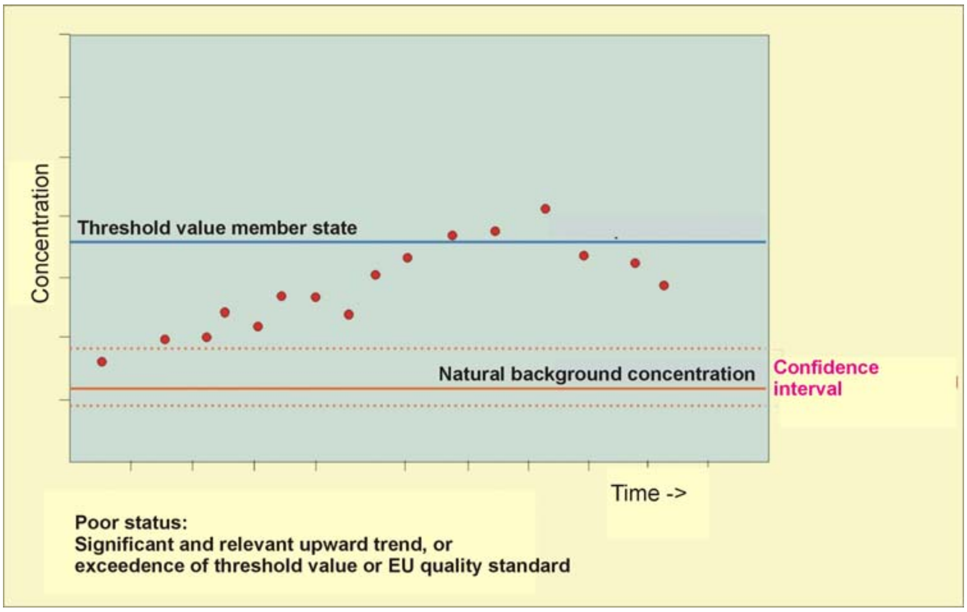

Figure 1.1 Trend reversal concept of the draft EU Groundwater Directive.

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 will be applied and tested at

various scales and in various hydrogeological situations. The methods applied should be related to the trend objectives of the Water Framework Directive and draft Groundwater Directive. In addition to the Description Of Work DOW, it is 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. The TREND 2 package is 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 will concentrate 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 will be extended to large scale monitoring.

1.2 General methods used in TREND 2

Research activities within TREND 2 focus on the following issues:

1 *Inventory of monitoring data of different basins and sub-catchments*. The inventory focuses on observation points with existing long time series. The wells should preferably be located in agricultural areas, because pesticides and nutrients are the main concern in trend detection for the Water Framework Directive. Additional information will be 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 include 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 will focus on robust methods for trend aggregation.

4 *Trend extrapolation.* Trend extrapolation will be 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 1.1). Statistical trend extrapolation will be performed on all the selected case studies. Deterministic modelling is limited to the Dommel and the Geer catchment in the Meuse basin, and the Brévilles catchment.

Basin	Contaminants	Trend extrapolation	Institutes
Meuse			
Dommel upper tributaries	Nitrate, sulphate, Ni, Cu, Zn, Cd	Statistical and deterministic modelling	TNO/UU
Noord-Brabant region	Nitrate, sulphate, 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		Statistical	IETU
Czech sub basins	Nitrate		
Schleswig-Holstein	Nitrate		

Table 1.1: Case studies in TREND 2

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

This report describes the physically-deterministic determination and extrapolation of time trends at selected test locations in Dutch part of the Meuse basin (TNO/UU), the Brévilles catchment (BRGM) and the Geer catchment (ULg).

1.5 Structure of the report

Subsequent chapters each describe the results of the trend determination in the Dommel (Chapter 2, TNO/UU), the Geer (Chapter 3, ULg) and the Brévilles (Chapter 4, BRGM) catchments.

1.6 Submission

This initial due date of 15 November 2007 was postponed by two months to 15 January 2008 because of two unforeseen problems that obstructed the timely submission of this deliverable.

The first problem was that T2.10 relied on meteorological data to be supplied by the HYDRO work package. Accidentally, the submission date for Deliverable T2.10 was planned before these data were due for HYDRO and these data were unavailable before the initial deadline.

The second problem was due to the development of a new hydrological model, a process of which the progress is always hard to foresee. These problems appeared more severe, due to the transition from one Fortran compiler to the next, and more debugging and code testing was necessary to provide scientific confidence in the results.

Both problems were overcome within the 2 months postponement granted by the Scientific Technical Co-ordinator of AquaTerra Dr. Barth, thanks to hard work during these months of the postponement. We are grateful for this solution to the scientific problems, which enabled the TREND 2 team to deliver scientific work of good quality from which valuable conclusions for were drawn.

1.7 GLOSSARY

$^{3}H/^{3}He$	tritium-helium, method to determine accurate groundwater age
EPIC-Grid soil model	Semi-distributed physically based soil model developed by UHAGx
GCM	Global Circulation Model
HFEMC	Hybrid Finite Element Mixing Cell
MACRO	1D soil leaching model
MARTHE	2D saturated zone model
MODPATH	A particle tracking post-processing package for MODFLOW, the U.S. Geological Survey finite-difference ground-water flow model
MT3D	a modular three-dimensional multi-species transport model for simulation of advection, dispersion and chemical reactions of contaminants in groundwater systems
OXC	oxidation capcity; conservative chemical indicator under denitrification by pyrite oxidation
particle tracking	calculation of the movement of imaginary particles by groundwater flow
physically-deterministic model	numeric model using physical principles to predict ahead in time the state of the system, such as pressures, flow velocities or concentrations.
RCM	Regional Climate Model
SUFT3D code	Finite element simulator for Saturated Unsaturated Flow Transport in 3D
tritium	radioactive hydrogen isotope (^{3}H) with a half-life of 12.43 y, used to determine period of recharge of groundwater samples
TU	tritium unit, unit of measure for tritium, corresponds to 1 ^{3}H per 1^{18} atoms
TVD	Total Variation Diminishing scheme, numerical solution to contaminant transport

2 Physically-deterministic determination and extrapolation of time trends at selected test locations in Dutch part of the Meuse basin

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2.1 Introduction

After demonstrating trends and trend reversal in groundwater quality in agricultural recharge areas in Noord-Brabant (the Netherlands), as reported in Deliverable T2.3 and T2.4 and Visser et al (2007), we used a numerical deterministic model to determine trends in groundwater and surface water quality. We modelled groundwater flow with MODFLOW and contaminant transport with MT3D. For a more detailed description of the model and the input data we refer to AquaTerra Deliverable T2.5 and T2.6. Historical trends can be used to validate the numerical model; future trends will assist decision makers of EUPOL to anticipate the effects of measures aimed at improving the quality of groundwater and surface water.
Key to understanding trends in groundwater quality is to know the groundwater travel times towards the monitoring screens. Trends in surface water quality are the product of the travel time distribution at the catchment outlet and the concentration-depth profiles in the groundwater body. Other factors determining trends in groundwater quality are varying inputs and subsurface reactions.

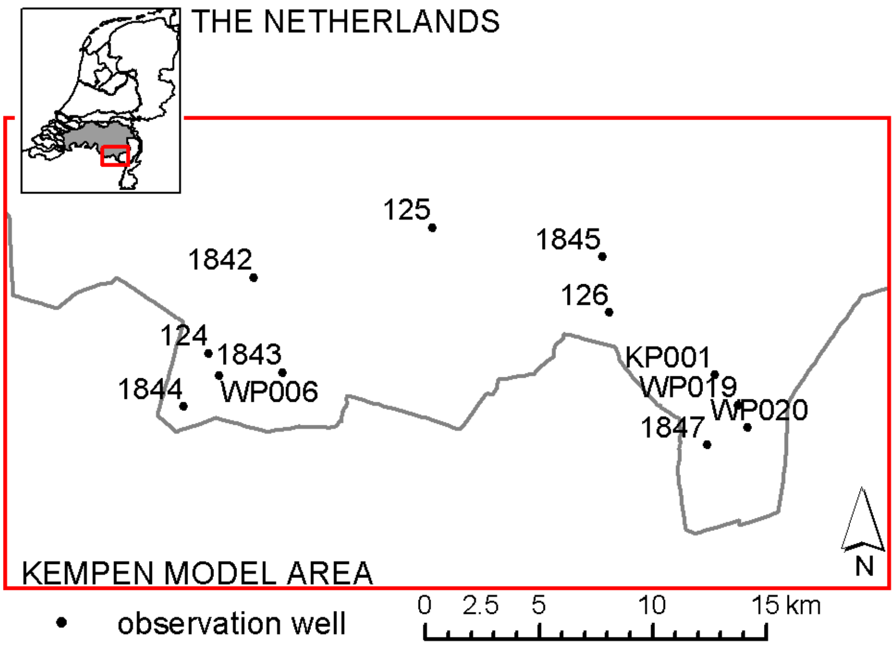

Figure 2.1: Locations of the observations points within the model area

To analyze trends in groundwater quality, we selected 12 locations in recharge areas of the model coinciding with actual monitoring locations of which time series of groundwater quality parameters are available (Figure 2.1). 9 locations were in agricultural land. At these locations we focused at model layers, which represent the depths of the screens of the monitoring wells: *phreatic* (4.5 - 6.5 m below surface), *shallow* (8.5 - 10.5 m b.s.) and *deep* (23.5 - 26.5 m b.s.).

2.2 Travel times at the monitoring screens

We compared vertical profile of groundwater travel times (ages) determined by particle tracking (MODPATH, Pollock, 1994) and direct age simulation (Goode, 1996; MT3D: Zheng and Wang, 1999) (Figure 2.2). Most profiles agree well to the logarithmic age-depth profile defined by the Vogel equation (Vogel, 1967).

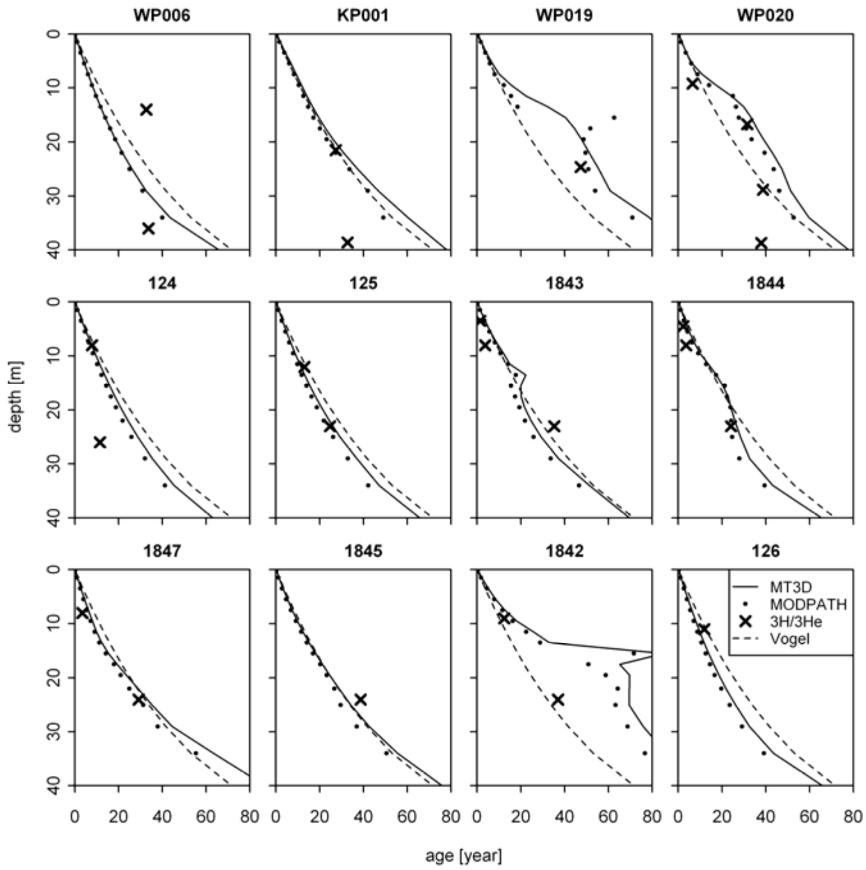

Figure 2.2: Groundwater ages at 9 well locations determined by 3H/3He, in the model determined by direct simulation (MT3D) and particle tracking (MODPATH).

Well locations WP019 and WP020 show the effect of a groundwater sub-catchment upstream of the monitoring well (Figure 2.3). The age profile of the shallow flow pattern follows the logarithmic profile, but flow paths that originate beyond the sub-catchment have significantly greater ages. This effect results in a discrete jump in the age profile calculated with MODPATH (especially WP190), but dispersion in the direct simulation of groundwater age using MT3D smoothens the age profile.

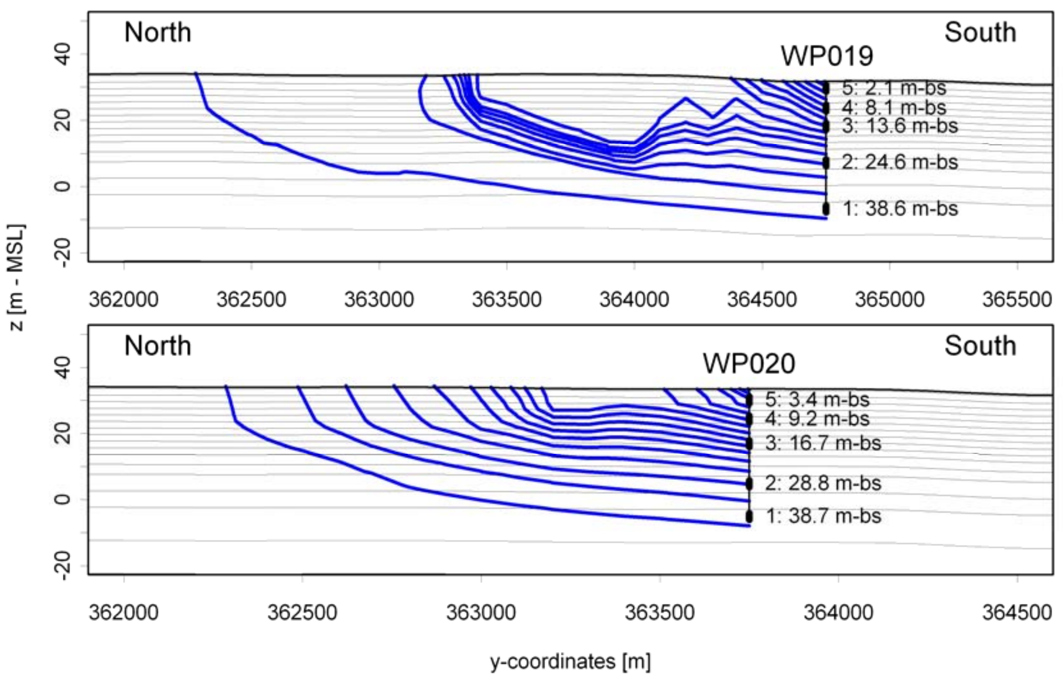

Figure 2.3: Path lines of particles ending at the locations of monitoring wells WP019 and WP020 show a groundwater sub-system upstream of the monitoring well.

Well location 1842 (Figure 2.4) shows the effect of old groundwater residing in a clay layer. Groundwater flow is very slow and nearly vertical. Groundwater ages in this layer are distinctly older than in neighbouring layers, both in the particle tracking and the direct simulation profile.

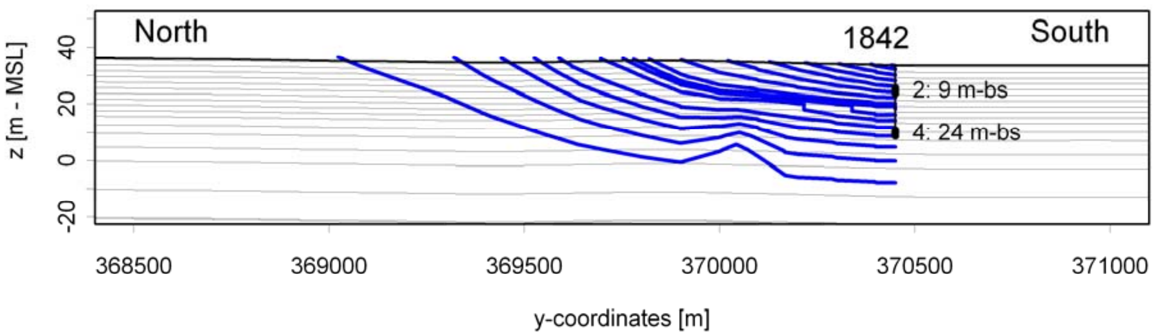

Figure 2.4: Path lines of particles ending at the location of monitoring wells 1842 show slow vertical groundwater flow through a clay layer between the monitoring screens.

Considering the large scale of the model domain and grid, the $^3H/^3He$ ages compare quite well with the modelled ages. Observed ages generally deviate more from the theoretical logarithmic profile then the modelled ages do, probably because the 3D model is only a coarse approximation to the real heterogeneous subsurface.

2.3 Time-series of modelled concentrations

Annual time-series of modelled concentrations at the screen locations in the model (Figure 2.5) show the effect of variations in travel times at the monitoring screen, as well as variation in inputs at the groundwater table and reactions in the subsurface. Oxidation capacity (OXC) behaves as a conservative tracer in the model, and variations in OXC concentrations are only the result of trends in input and variations in travel times. The peak of the concentrations is horizontally shifted by variations of travel times at the monitoring screens. Nitrate was modelled using a first order degradation process, to account for the denitrification by pyrite oxidation in the subsurface of Brabant. This approach was chosen, because the depth of the pyrite containing sediments is unknown. The degradation rate constant was calibrated to concentration-depth profiles (Broers and Heerdink, 2006). The time-series of nitrate concentrations clearly show the effect of the first order degradation: nitrate concentrations in older water from deeper screens are more reduced. Variations in travel times at the same screen depth result in higher or lower concentrations due to more or less degradation, as well as a horizontal shift of the peak concentrations. Concentration time series of heavy metals (zinc) are the result of spatially varying atmospheric deposition at the surface, the complex geochemistry in the unsaturated zone leading to large spatial variations of concentrations at the groundwater table, retardation in the saturated zone, and variations in travel times. As a result of retardation in the saturated zone, the modelled zinc contamination has not yet reached the depth of the shallow monitoring wells.

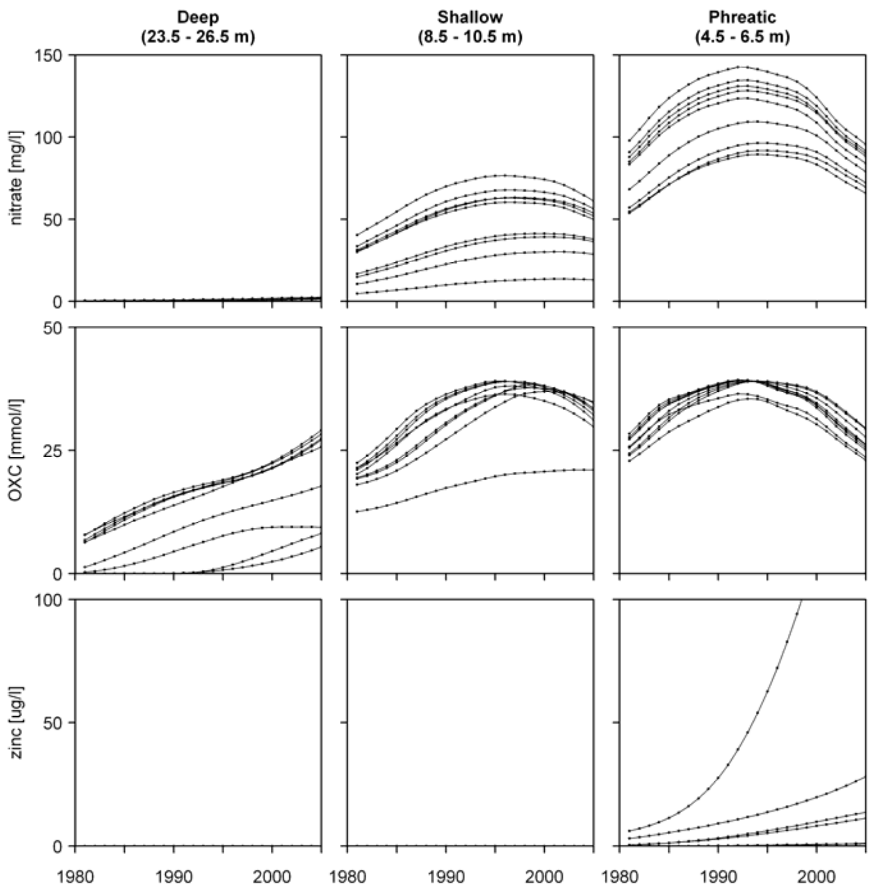

Figure 2.5: Time series of modelled concentrations of nitrate, OXC and zinc at the 9 monitoring locations.

2.4 Trends in modelled concentrations

We determined linear trends in the time-series of modelled concentrations between 1995 and 2005. We found a significant trend with increasing concentrations of nitrate and OXC at the deep screen level. A trend with decreasing concentrations was significant for nitrate and OXC in the *phreatic* screen, and also at the shallow depth for OXC. No significant trends were found for zinc. Because the contaminants are retarded and have not yet reached the screen depth of the phreatic screens at all locations, the trend for all locations combined is not yet significant. Significant trends can be determined, but only for individual locations.

Trends	deep	shallow	phreatic
Nitrate	↑	-	↓
OXC	↑	↓	↓
Zinc	-	-	-

Table 1: Trends in modelled concentrations between 1995 and 2005.

2.5 Vertical profiles of modelled concentrations

Vertical profiles of modelled concentrations plotted for several times and averaged over all well locations (Figure 2.6) illustrate the vertical movement of the contaminants in the groundwater system. Arrows in the profiles indicated significant trends between 1995 and 2005 at the *phreatic*, *shallow* or *deep* monitoring level. Again, OXC (Figure 2.6b) only shows the downward movement of the contaminant front unaltered by subsurface reactions. Nitrate (Figure 2.6a) shows the combined effect of downward movement and first order degradation. The complex combination of processes acting on zinc (Figure 2.6c), downward transport, input concentrations and adsorption to the subsurface, all of them varying in time and space, result in an apparent delayed downward movement of the zinc contamination front.

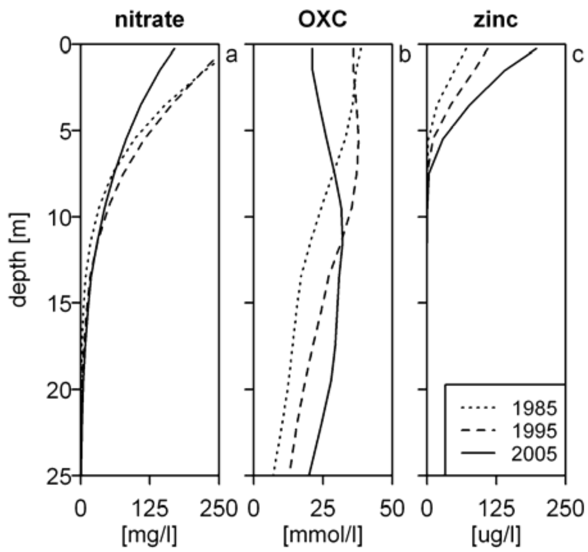

Figure 2.6: Average profiles of the modelled concentrations at the 9 locations in 1985, 1995 and 2005.

2.6 Trend extrapolation

The real value of 3D deterministic models is the capability of predicting future trends and the effect of regulatory measures aimed at improving the groundwater and surface water quality. Here we show the long-term effect of the current land use practices, by running the model with a constant present day input up to 2050 (Figure 2.7). Concentrations of nitrate and OXC decline in the phreatic and shallow screens, because historical inputs were higher than at present. In the deep screens concentrations of nitrate are still close to zero, due to denitrification. Concentrations of OXC may increase or decrease up to 2010, but will generally decrease afterwards, although screens sampling very old water may still show an increase. Contrary to nitrate and OXC, concentrations of zinc will increase in the shallow and phreatic screens in the future due to the leaching of the zinc front from the adsorption complex.

2.7 Future trends

Table 2 summarizes present day and future trends in concentrations of nitrate, OXC and zinc as predicted by the physically deterministic model. Between 2010 and 2020 a significant downward trend was found for OXC at the shallow screen depth. Large variations in travel time at the deep screen obscure clear trends for all data together. Individual time series may show both upward and downward trends. At shallow and phreatic screen depths, the constant input causes no new trends to develop. Leaching of the zinc front from the adsorption complex however causes a new significant upward trend to develop at the shallow screen depth. No other significant trends were found for all data together, but concentrations may increase dramatically at individual locations if the (retarded) contamination front reached the screen depth.

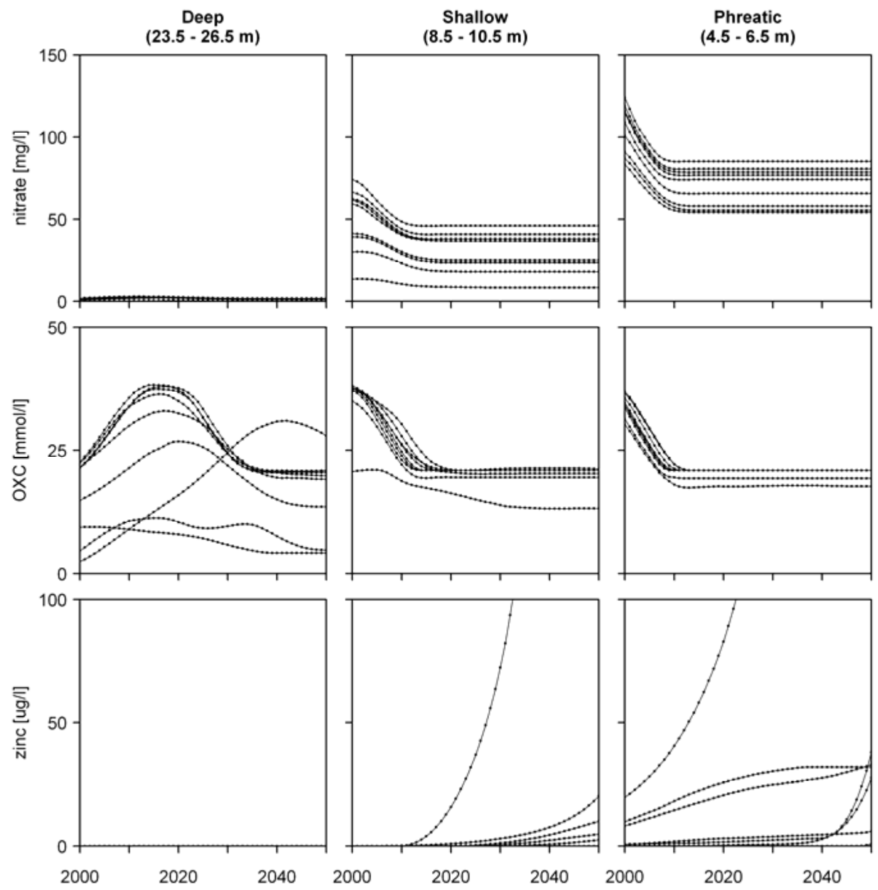

Figure 2.7: Physically deterministic extrapolated time series of concentrations of nitrate, OXC and zinc at the 9 monitoring locations between 2000 and 2050.

Trends	1995 - 2005			2010 - 2020			2040 – 2050		
	deep	shallow	phreatic	deep	shallow	phreatic	deep	shallow	phreatic
nitrate	↑	-	↓	-	-	-	-	-	-
OXC	↑	↓	↓	-	↓	-	-	-	-
zinc	-	-	-	-	↑	-	-	-	-

Table 2: Future trends in groundwater quality

2.8 Conclusions

The numerical deterministic model can reveal variations in groundwater travel times at the monitoring screen caused by clay layers in the subsoil or groundwater sub-catchments. These variations are the key to interpreting variations, in space and time, of concentrations of contaminants at the monitoring screens. Backward particle tracking or direct age simulation to obtain travel time distributions also allows the model to be validated against $^{3}H/^{3}He$ groundwater dating.

Figure 2.7 shows that in the future new trends may develop – for example a trend of increasing zinc concentrations at the shallow screen – which were not observed in modelled time series before 2005. Nonlinear mechanisms such as adsorption and desorption cause these new trends. Extrapolating present day trends into the future may be invalid if nonlinear processes play a role in the behaviour of contaminants in the groundwater system. A physical deterministic approach is then preferred.

3 Physically-deterministic determination and extrapolation of time trends in the Geer catchment (ULg)

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3.1 Introduction

In the framework of the AquaTerra project, HG-ULg has developed a methodology using statistical approaches to infer trends in nitrate groundwater quality for different sub-basins of the Meuse River. Following the development of this methodology, HG-ULg is developing a groundwater flow and solute transport model as a prediction tool for trend analysis in one of these sub-basins, the Geer basin.

A concept for large-scale solute transport modelling, the Hybrid Finite Element Mixing Cell (HFEMC) developed by HG-ULg and implemented in the 3D simulator SUFT3D (Deliverable R3.18) is used to develop a large scale groundwater flow and solute transport model for the hydrological basin of the Geer river (480 km²). In the concept, different mathematical approaches can be used to represent the flow and solute transport phenomena (Table 3.1). For the model developed in this project, groundwater flow is modelled using classical equations based on Darcy's law. The transport of solute is modelled using the distributed mixing cells approach. This approach consists in assuming that water entering in a cell is instantly mixed with the water in that cell. The transport processes that can be considered using the HFEMC approach are advection, degradation, sorption-desorption and dual-porosity related to the presence of immobile water. Hydrodynamic dispersion is not considered in the model as it is directly related to the degree of refinement of the 3D grid. The advantage of the mixing cell approach is the fact that the numerical scheme is unconditionally stable. In this case study, the source of transported solutes (tritium and nitrate, see afterwards) being fully dispersed at the top of the model, willing to quantify and model the "true" hydrodynamic dispersion in the model is unnecessary since dispersion in groundwater corresponds essentially to the dispersion of the source. Further conceptual choices made to develop the model were presented in Deliverable R3.18 and T2.8.

		TRANSPORT		
		Simple Linear Reservoir	***Distributed Mixing Model***	***Advection-dispersion***
FLOW	***Simple Linear Reservoir***	OK	impossible	impossible
	Distributed Linear Reservoir	OK	OK	impossible
	Flow in porous media	OK	USED HERE	OK

Table 3.1. Solutions implemented in the SUFT3D code and restrictions of use. The combined approach used for the model developed in the framework of the AquaTerra project is highlighted in red

During the last months of the project, HG-ULg activities have concerned:

- Calibration of the groundwater flow model;
- Calibration of the groundwater solute transport model;
- Use of the model for trend prediction in nitrate groundwater quality.

3.2 Calibration of the groundwater flow model

The groundwater flow model has been calibrated in steady state using two contrasting piezometric situations assimilated to steady state: one corresponding to high groundwater levels (during the period 1983-1984), the second to low groundwater level (during the period 1991-1992). For this calibration step, all stress factors (pumping, recharge…), prescribed to the model, are assumed constant. The recharge (368 mm/year and 226 mm/year for respectively the period 1983-1984 and 1991-1992) is uniformly distributed and calculated based on groundwater budgets for the corresponding years (Deliverable R3.16). For the period 1983-1984 and 1991-1992, respectively 39 and 50 piezometric measurements were used. The calibration was performed by trial-and-error changing values and spatial distribution of hydraulic conductivities. The values of hydraulic conductivities obtained at the end of the calibration process are summarized in Table 3.2. These values are of the same order of magnitude than those presented in Deliverable R3.16 (10^{-9} to 10^{-7} m/s for the loess and 10^{-5} to 5×10^{-3} for the chalk). The objective of the calibration process was to minimize the difference between observed and computed groundwater levels. The process was ended when each difference between observed-computed groundwater levels was less than 5 meters.

In Figure 3.1, a general quality of the calibration is presented in the form of a scatter plot diagram of observed versus computed groundwater levels for the period 1983-1984.

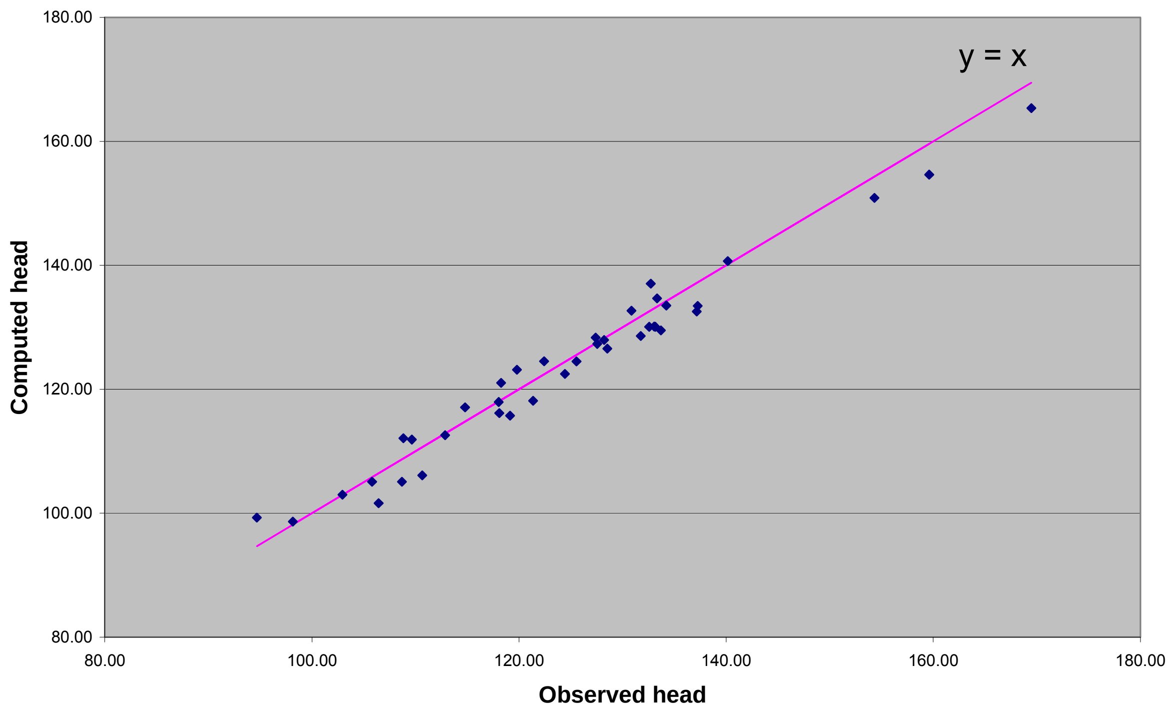

Figure 3.1. Comparison between observed and computed head for the period 1983-1984 (RMSE = 0.4621 m)

	Particularity	Saturated hydraulic conductivity (m/s)	Specific storage (1/m)	Residual water content (/)	Saturated water content (/)	Effective porosity (/)
Bottom chalk						
Material 1		4.0×10^{-5}	1.0×10^{-4}	0.01	0.41	0.005
Material 2		2.0×10^{-5}	1.0×10^{-4}	0.01	0.41	0.01
Material 3		2.0×10^{-5}	1.0×10^{-4}	0.01	0.41	0.01
Material 4		2.75×10^{-5}	1.0×10^{-4}	0.01	0.41	0.01
Material 5	Fractured zone	4.0×10^{-4}	1.0×10^{-4}	0.01	0.41	0.01
Material 6	Fractured zone, Horion-Hozémont Fault	1.0×10^{-3}	1.0×10^{-4}	0.01	0.41	0.01
Material 7	Fractured zone, dry valleys	3.0×10^{-3}	1.0×10^{-4}	0.01	0.41	0.02
Upper chalk (bottom part)						
Material 8		2.0×10^{-4}	1.0×10^{-4}	0.01	0.41	0.005
Material 9		2.0×10^{-4}	1.0×10^{-4}	0.01	0.41	0.01
Material 10		7.0×10^{-5}	1.0×10^{-4}	0.01	0.41	0.01
Material 11		1.5×10^{-4}	1.0×10^{-4}	0.01	0.41	0.01
Material 12	Fractured zone, Horion-Hozémont Fault	1.0×10^{-3}	1.0×10^{-4}	0.01	0.41	0.01
Material13	Fractured zone, dry valleys	3.00×10^{-3}	1.0×10^{-4}	0.01	0.41	0.02
Upper chalk (upper part)						
Material 14		2.0×10^{-4}	1.0×10^{-4}	0.01	0.41	0.005
Material 15		1.0×10^{-4}	1.0×10^{-4}	0.01	0.41	0.01
Material 16		1.0×10^{-3}	1.0×10^{-4}	0.01	0.41	0.01
Material 17		2.0×10^{-4}	1.0×10^{-4}	0.01	0.41	0.01
Material 18	Fractured zone, Horion-Hozémont Fault	1.0×10^{-3}	1.0×10^{-4}	0.01	0.41	0.01
Material 19	Fractured zone, dry valleys	2.70×10^{-3}	1.0×10^{-4}	0.01	0.41	0.02
Material 21		$7.\ 0 \times 10^{-5}$	1.0×10^{-4}	0.01	0.41	0.01
Loess						
Material 20		1.0×10^{-7}	1.0×10^{-4}	0.01	0.44	0.44

Table 3.2. Hydrodynamic parameters and effective porosity as obtained after calibration

3.3 Calibration of the groundwater solute transport model

The solute transport model has been developed to study the long-term temporal evolution and to predict the evolution of nitrate concentrations in the Geer basin aquifer at regional scale. As mentioned in the Deliverable R3.18, the loess layer is considered as a single porosity media while the chalk layers are modelled as dual-porosity media. Values of the parameters of the equation describing the transport processes have been taken from the literature on the Geer Basin (Brouyère 2001; Brouyère *et al.* 2004).

The model is run in steady state for groundwater flow and in transient state for solute transport modelling. The model has been run firstly in steady state to create initial conditions needed for the transient solute transport model.

Two sets of data have been 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). Tritium and nitrates have the same behaviour in groundwater (except the fact that the tritium is degraded due to the radioactive decay). The advantage of tritium is that the concentration in the infiltrating water can be easily estimated because it is essentially a function of latitude. Nitrate concentrations in the infiltrating water are more difficult to determine as they are function of land use and show temporal as well as seasonal evolutions (Deliverable T2.2).

The calibration was performed by trial-and-error changing the values and the spatial distribution of the effective porosity. The values of parameters obtained at the end of the calibration process are presented in Table 3.2.

3.3.1 Calibration of the groundwater transport model using the results of the tritium survey

As mentioned in Deliverable T2.8, HG-ULg has performed a tritium survey during winter 2004-2005. Samples have been taken in the Geer basin and to the north of the basin where the aquifer becomes confined, in continuity with the unconfined part of the aquifer which corresponds to the Walloon Region territory. As the spatial extension of the model is limited to the hydrologic basin of the Geer River, the model does not simulate the concentration in the confined part located to the North of the basin.

The concentrations measured during this survey have been used to calibrate the model. The concentration in the infiltration is uniformly distributed in the basin, equal to the mean tritium concentration measured in the precipitation at the station of Groningen (Figure 3.2). The transport model has been run in transient state for the period 1950-2004. The modelling period was divided in 55 time steps of 1 year. The quality of the calibration procedure has been assessed in three different forms:

- A qualitative comparison of observed versus computed maps of the spatial distribution of tritium in the aquifer at the end of 2004;
- A scatter plot of observed versus computed tritium concentration for the winter 2004-2005;
- Scatter plots of computed tritium concentrations versus time for different nodes located on a same vertical in the mesh describing the aquifer and its surmounting layers.

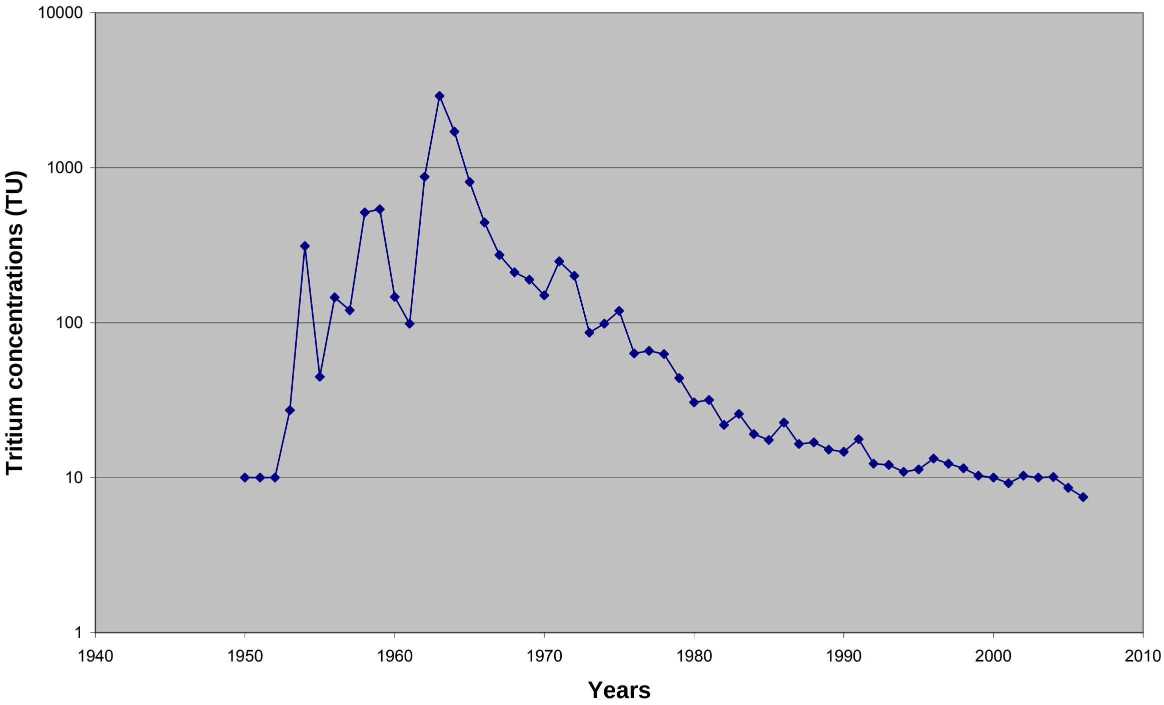

Figure 3.2. Tritium concentration in the precipitation measured at the Groningen station (data provided by Dr P. Maloszewski, GSF Helmholtz Zentrum, Munich)

Computed spatial distribution of the tritium concentrations

A qualitative comparison between the maps respectively of tritium units measured in the Geer basin drawn after the winter 2004-2005 survey (Figure 3.3) and of computed tritium concentration at the top of the chalk in 2004 (Figure 3.4) shows that (1) the concentrations are of the same magnitude and (2) the zones distinguished from the tritium concentrations measured are also observed on the computed map.

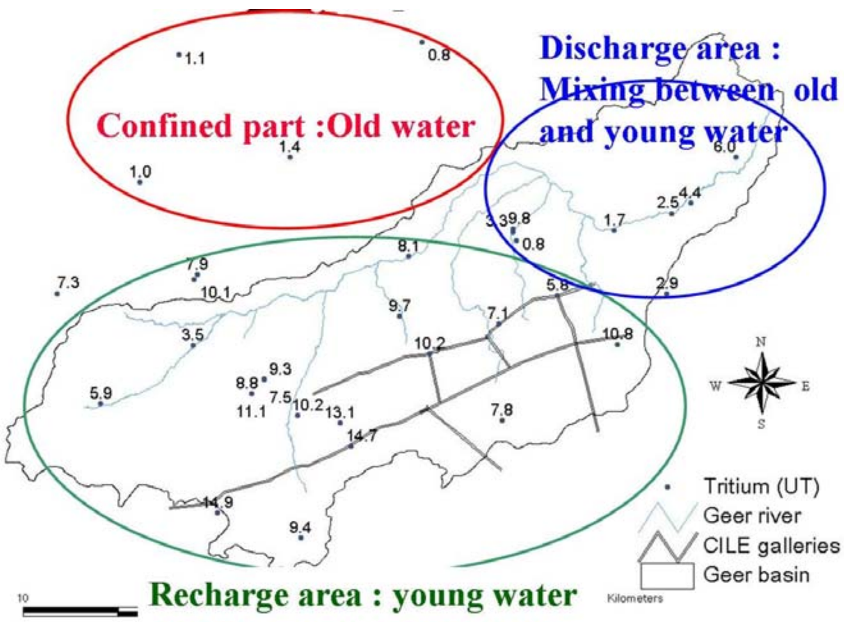

Figure 3.3. Tritium units measured in the groundwater samples from the Geer basin

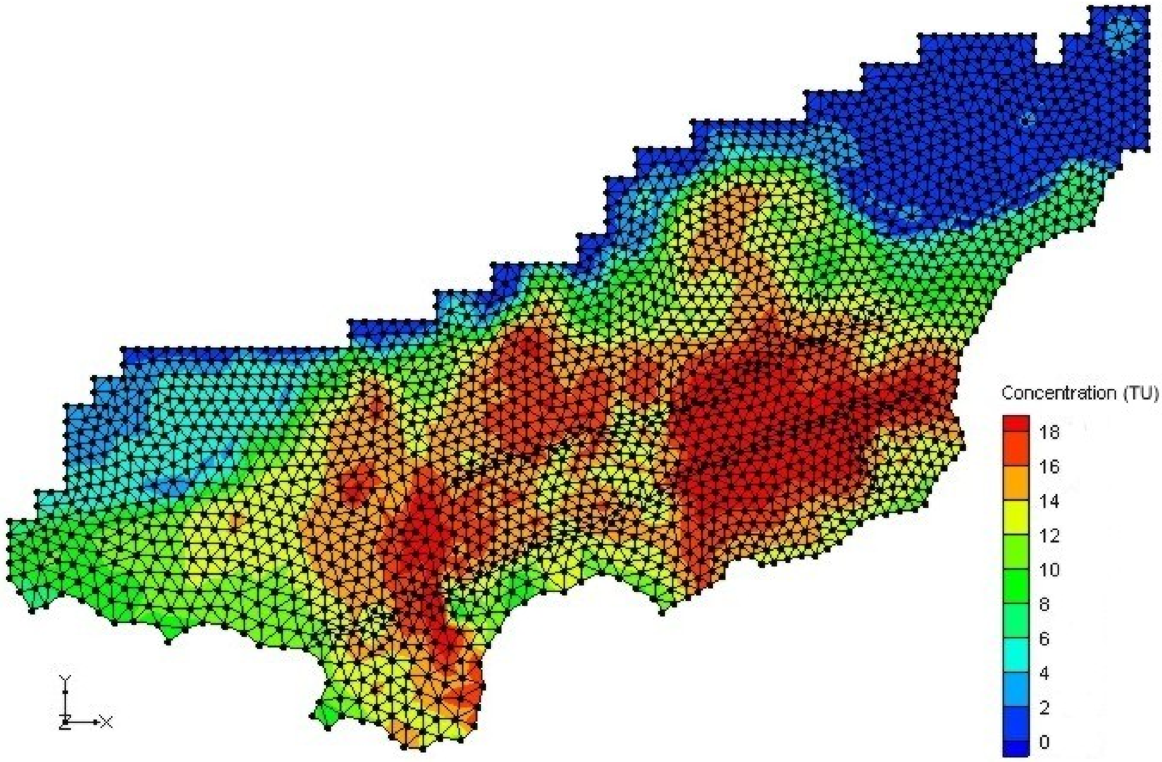

Figure 3.4. Tritium units computed for the year 2004 at the top of the chalk layer (red and blue colors correspond to respectively young and old water)

Scatter plot of observed versus computed concentrations

A scatter plot of observed versus computed concentrations allows checking the general quality of the calibration (Figure 3.5). 25 points where tritium concentrations were analysed were used for this comparison. One difficulty is that most piezometers and wells that were sampled for tritium analysis are screened over the whole thickness of the aquifer (most often several tens of meters). Because of that, tritium samples represent depth-averaged conditions. As the concentrations are computed at nodes and they vary with depth, it is difficult to compare measured concentrations and concentrations computed at a particular node of the 3D mesh. For the wells where the screen depth is known, a vertical weighting of the computed concentrations has been computed on the basis of hydraulic conductivities and the height of the saturated zone in each horizontal layer of model nodes. However, the depths of the wells and the position of the screens are, for the majority of the wells, unknown. These explanations can partly explain the dispersion of points around the line Y = X in Figure 3.5.

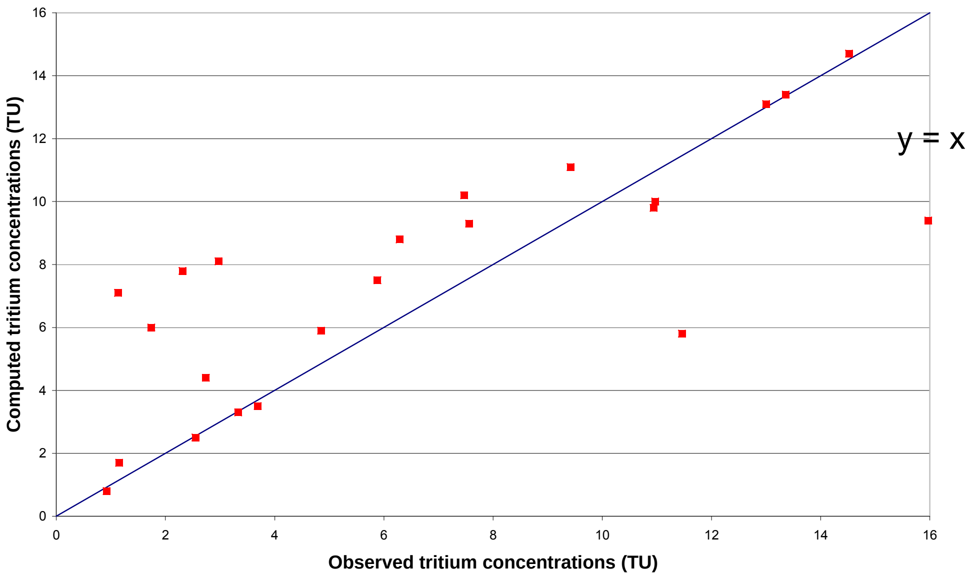

Figure 3.5. Comparison between observed and computed tritium unit for the winter 2004-2005

<u>Vertical variation of computed tritium concentrations</u>

The average velocity of solute in the unsaturated zone (loess and chalk layers) in the Geer basin has been estimated at 1m/year (Brouyère *et al.* 2004). The capability of the model to reproduce this velocity in the unsaturated zone has been assessed by following the propagation of the tritium peak in the unsaturated zone. For example, computed tritium concentrations versus time for different nodes located on a same vertical, shown Figure 3.6, can be used to compute the solute velocity in the unsaturated zone (Table 3.3). The calculated solute velocity in the unsaturated zone confirms the previous estimates of 1m/year, which indicate that the tritium transport model is well parameterized in the unsaturated zone.

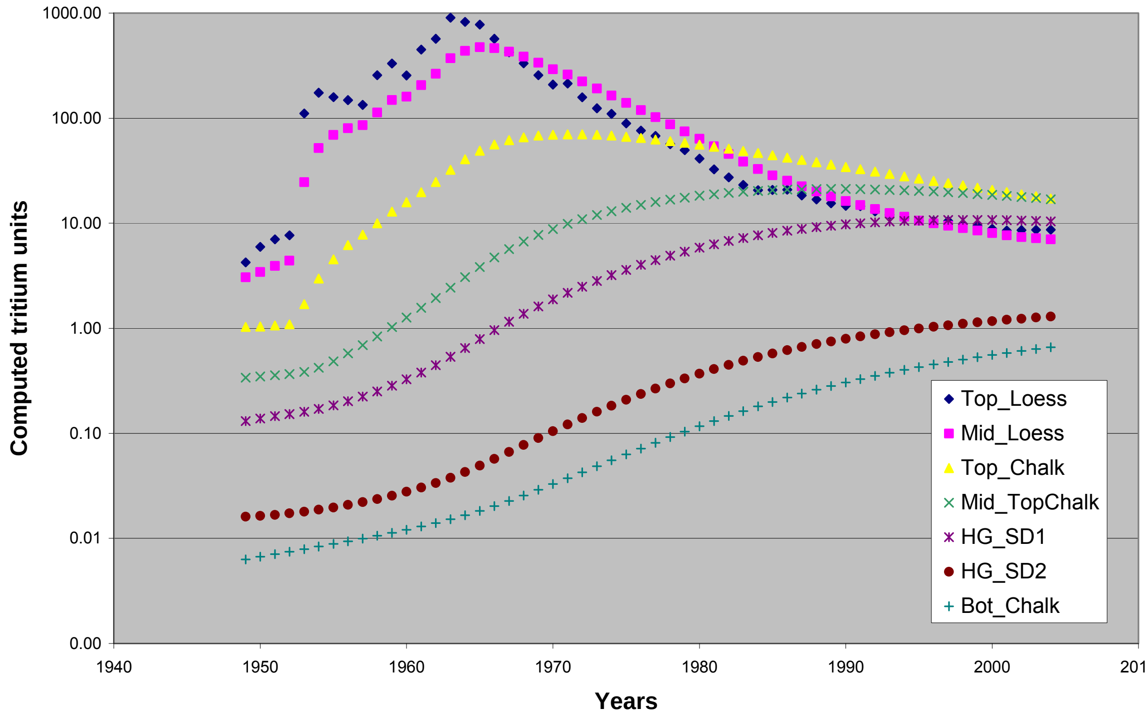

Figure 3.6. Computed temporal evolution of tritium units for the different nodes defining a vertical in the mesh

Node location	Node elevation (m)	Year of tritium peak	Velocity (m/y)
Top loess	132.02	1963	
Mid loess	129.263	1966	0.92
Top chalk	126.507	1971	0.55
Mid of top chalk	111.951	1987	0.90

Table 3.3. Calculated velocity of the tritium peak in the unsaturated zone

3.3.2 Calibration of the groundwater transport model using the time evolution of nitrate concentrations observed in the Geer basin

The capability of the model to reproduce the observed long-term time evolution of nitrate concentrations in the groundwater has been evaluated comparing observed and computed time series of nitrate concentrations. The model has been run in transient state for the period 1950-2004. The modelling period was divided in 55 time steps of 1 year.

Nitrate concentrations in infiltrating water are complex to determine as they are function of the land use and there are temporal evolutions that are difficult to correct for (Deliverable T2.2). Estimation of water and nitrate fluxes to groundwater is computed by the UHAGx (Dr. A Degrée, ir. C.Sohier). These fluxes are evaluated using the EPIC-Grid model for the last 30 years (1970-2000) for the Walloon part of the Meuse Basin, on the basis of a 1km² grid. These fluxes are not yet available because the correspondence between the EPIC-Grid model and the groundwater model has to be improved. As a first step, a simplified uniform scenario of input (Figure 3.7) was defined on the basis of information from the PIRENE project (Dautrebande and Sohier 2004). In this scenario, the concentration in the infiltrating water is assumed to be equal to 15 mg/l at the beginning of the fifties. Nitrate concentrations increase between 1950 and the mid of the eighties to reach a plateau with nitrate concentrations of 80 mg/l. This plateau can be related to the stabilization of nitrogen load on the crops observed in Western Europe.

As an example of the capability of the developed model, observed and computed time series of nitrate concentrations for the point H7, H9 and H18 (for which trends have been detected and estimated, Deliverable T2.4) are shown respectively in Figures 3.8, 3.9 and 3.10. The computed values shown Figure 3.8, 3.9, 3.10 are those computed at the highest node in the unsaturated zone.

During the first part of the Aquaterra project, HG-ULg has developed a methodology to identify and quantify trends in nitrate quality trends. To assess the quality of the calibration, a ‘computed slope’ describing the upward of the computed nitrate concentration was calculated and compared with the slope computed with the statistical tools (Table 3.4). This ‘computed slope’ is determined considering a linear variation between the concentrations computed in 1960 and 1995.

The computed nitrate concentrations are of the same order of magnitude than the observed one. The general upward trend is reproduced by the model but 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. The results presented here proves however the capability of the model to reproduce the nitrate concentrations and their temporal evolutions.

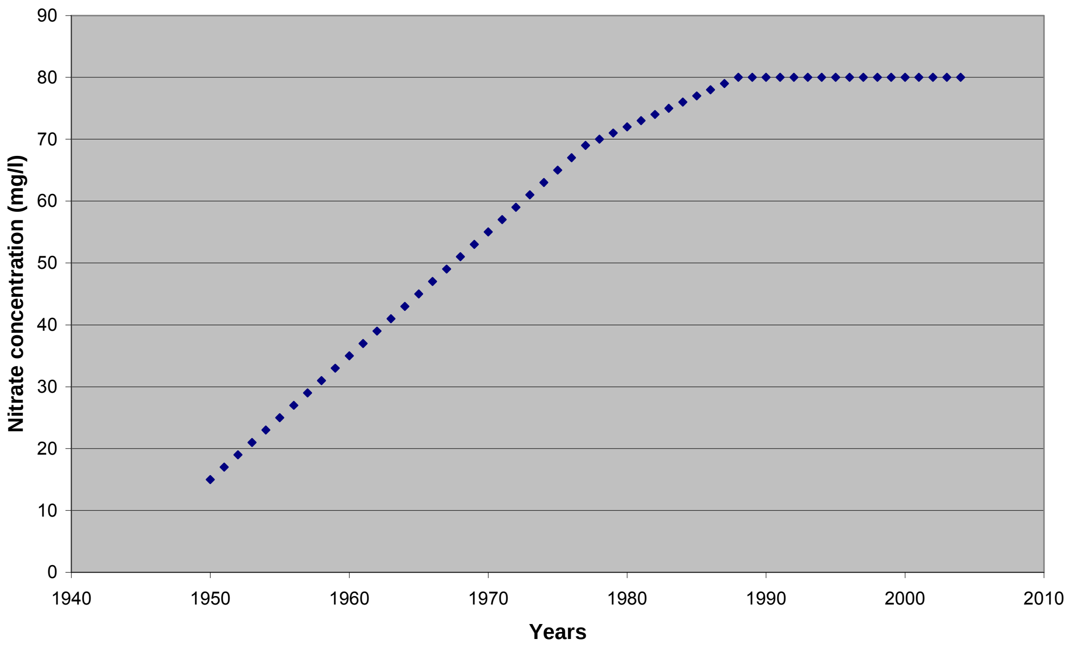

Figure 3.7. Simplified assumed input function for nitrate concentration in the infiltration

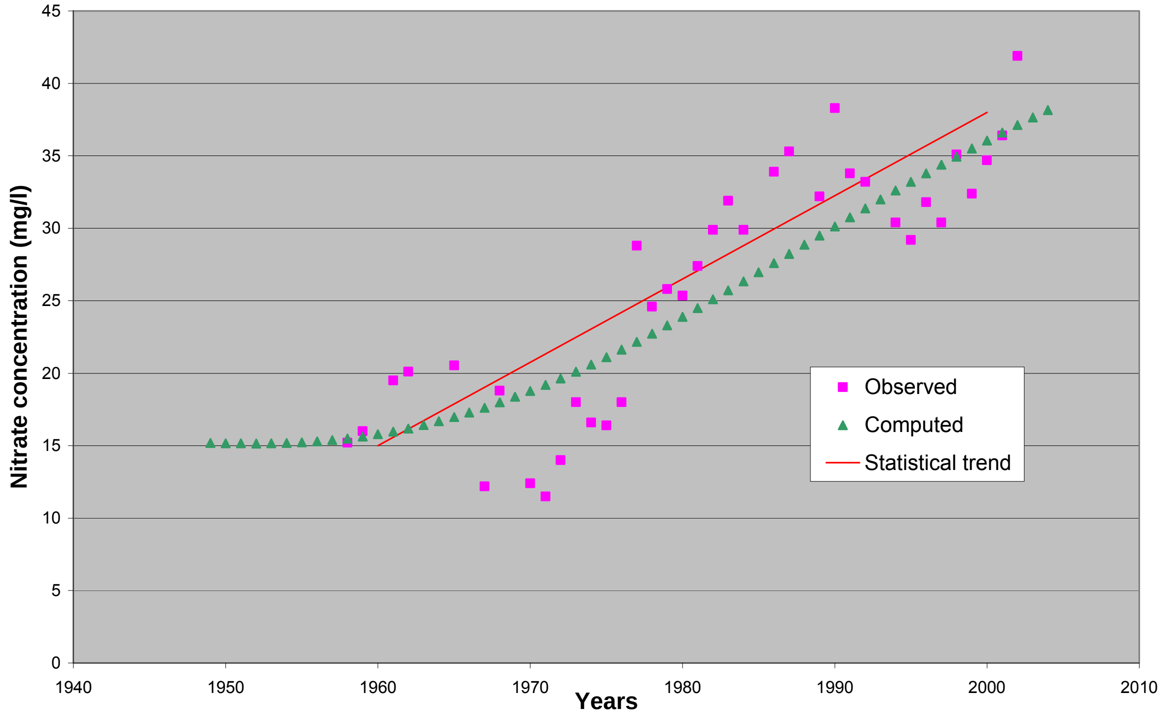

Figure 3.8. Comparison between the computed and observed time evolution of nitrate concentration in the well H7?

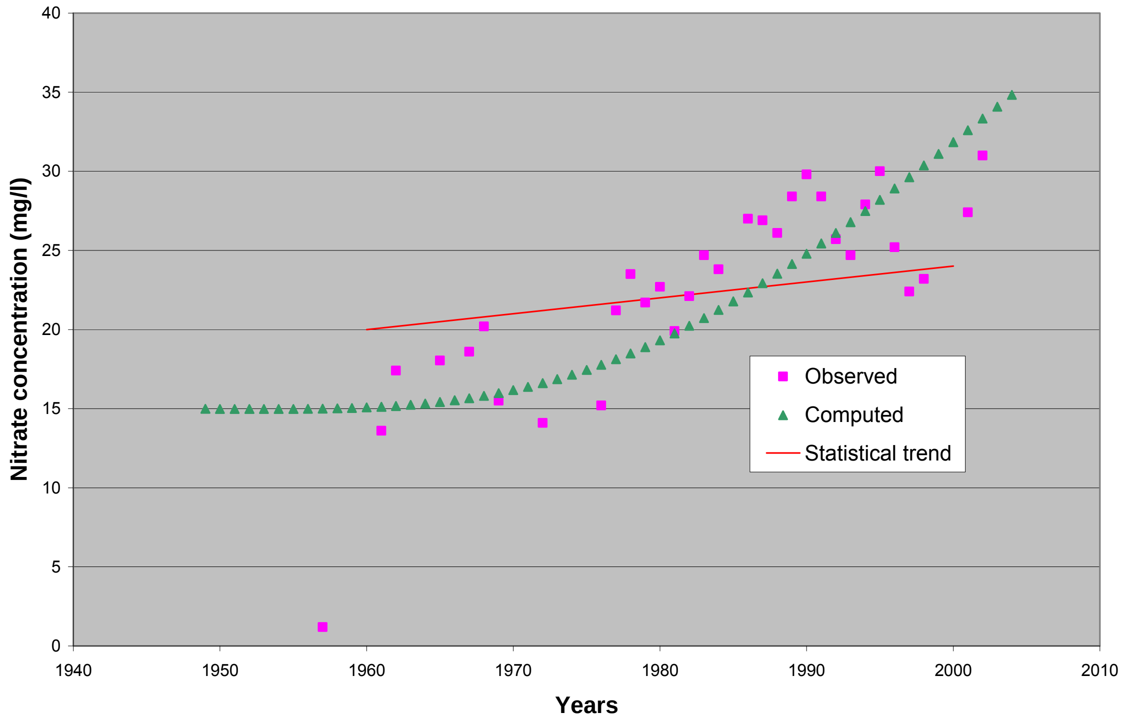

Figure 3.9. Comparison between the computed and observed time evolution of nitrate concentration in the well H9

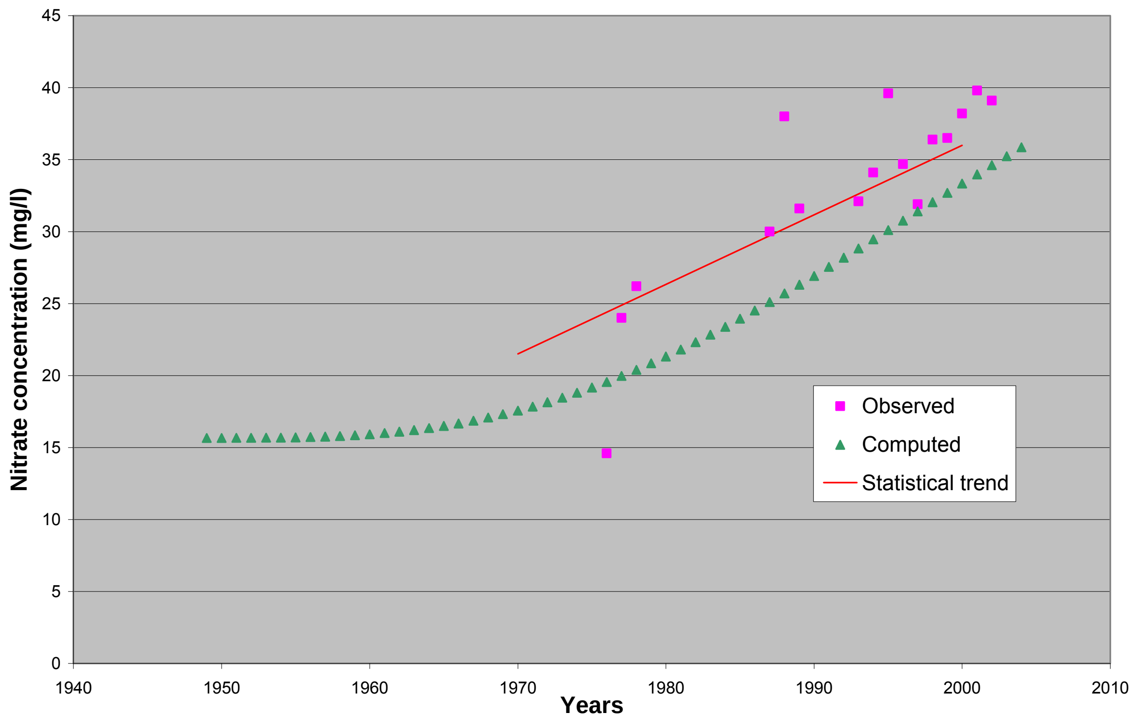

Figure 3.10. Comparison between the computed and observed time evolution of nitrate concentration in the well H18

Code of the well	Statistic trend (mg/y)	'Simplified slope' of the computed trend (mg/y)
H7	0.5883	0.497
H9	0.0928	0.374
H18	0.4803	0.405

Table 3.4. Comparison between the slope of the statistic trend and the computed trend

3.4 Extrapolation of nitrate trends

Two contrasting scenarios of nitrate input have been tested to assess the temporal evolution of nitrate concentrations in the groundwater. In Scenario 1, the nitrate concentration remains constant and equal to the concentration in the input at the beginning of the nineties. In Scenario 2, the nitrate concentration in the infiltration is set to zero after 2010 (Figure 3.11). Real input should a mix between these two extreme scenarios. Since the end of the nineties, efforts have been made to decrease the nitrogen load in agriculture. The model has been run in transient state for the period 1950-2050. The modelling period was divided in 101 time steps of 1 year.

The time evolution of nitrate concentration in groundwater is plotted for three nodes located along a North-South oriented transect (Figure 3.12). The depth to water table increases from point 1 to point 3 (Table 3.5). The concentrations are observed on the highest node in the saturated zone. The time evolution of the computed nitrate concentrations in groundwater for points 1 to 3 under Scenario 1 are presented in Figure 3.13. In 2050, the computed concentrations have not stabilised yet.

The time evolution of the computed nitrate concentrations in groundwater for points 1 to 3 under Scenario 2 are presented in Figure 3.14. Trend reversal occurs later from point 1 to point 3 (Table 3.5). As expected, the delay between changes in nitrate input and trend reversal in groundwater is mostly governed by the depth to water table.

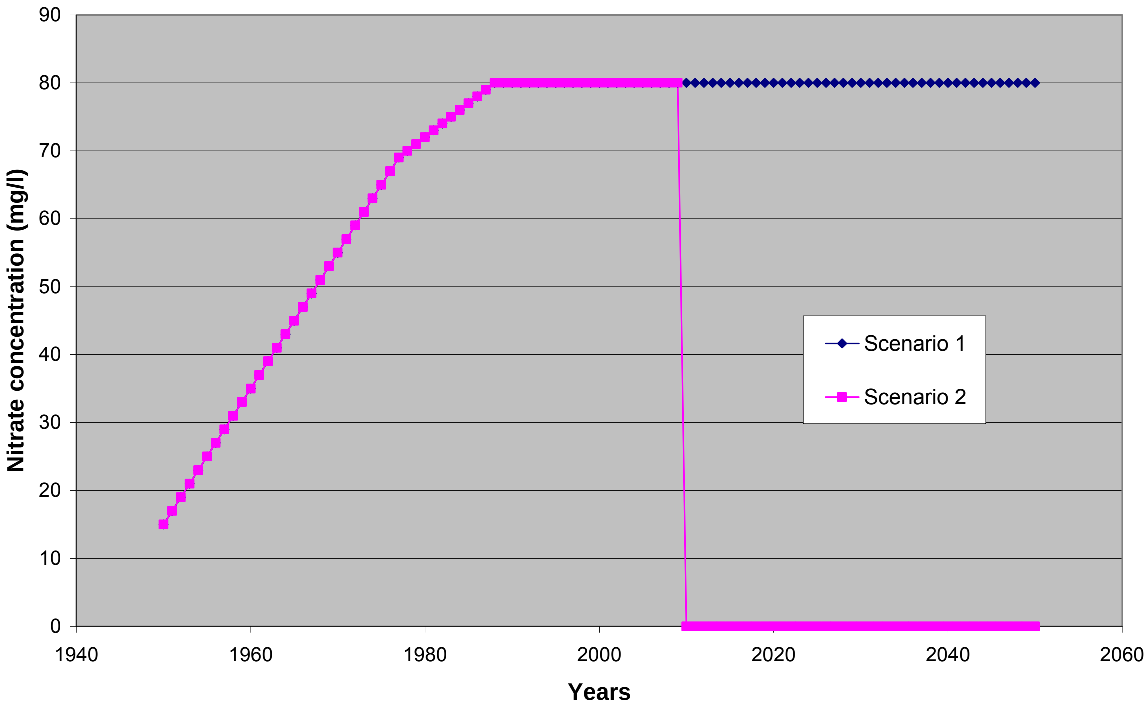

Figure 3.11. Simplified scenarios for nitrate concentration in the infiltration

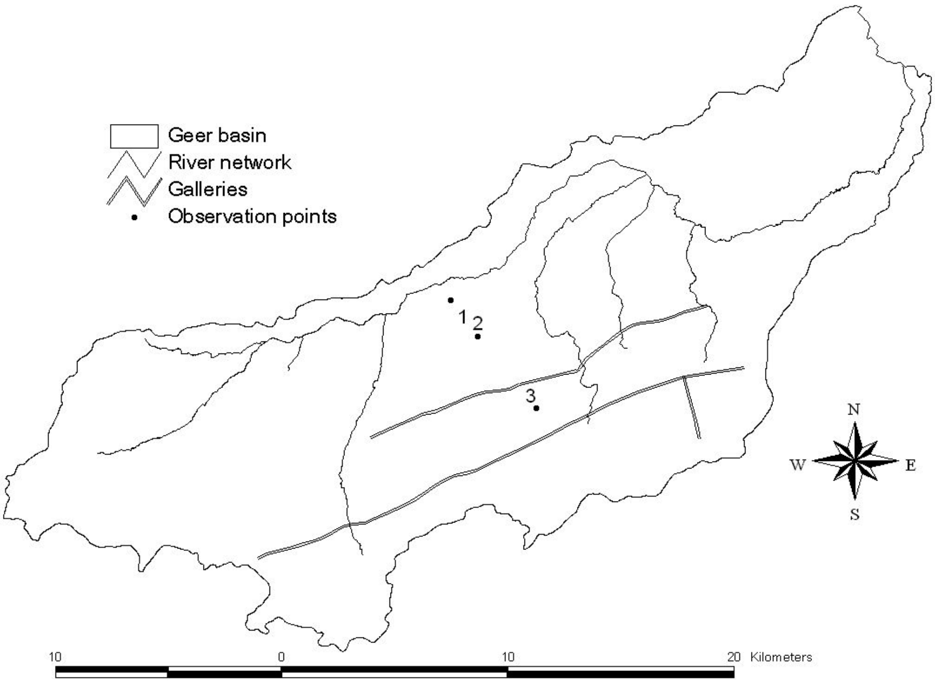

Figure 3.12. Location of the points where the evolution of nitrate concentrations as computed with the two scenarios are observed

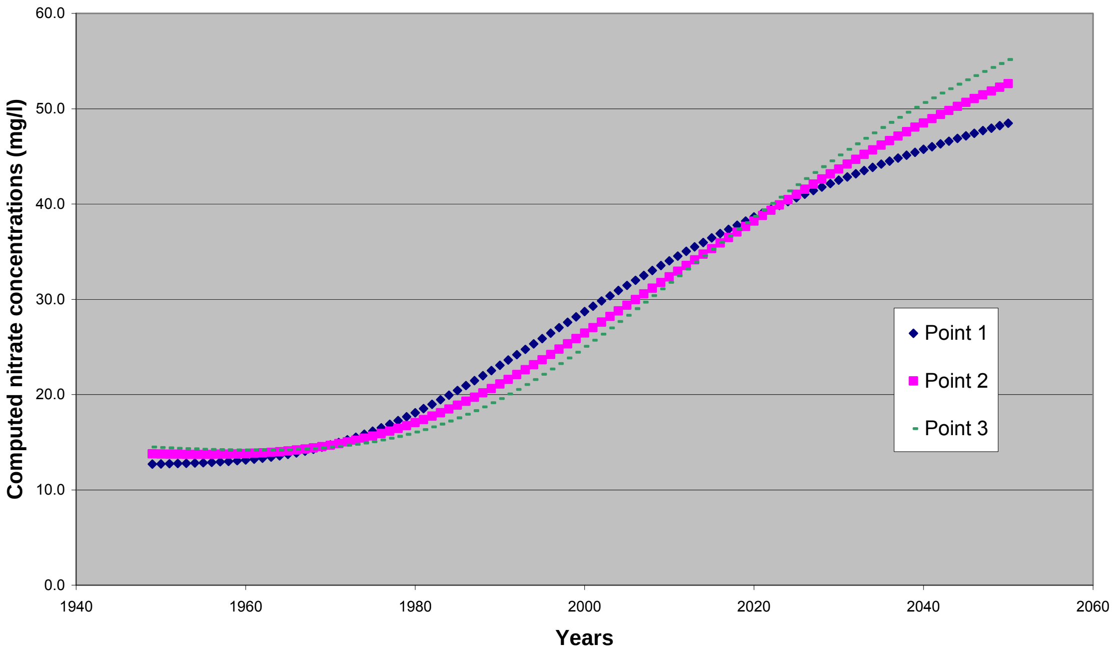

Figure 3.13. Time evolution of the nitrate concentrations computed with Scenario 1

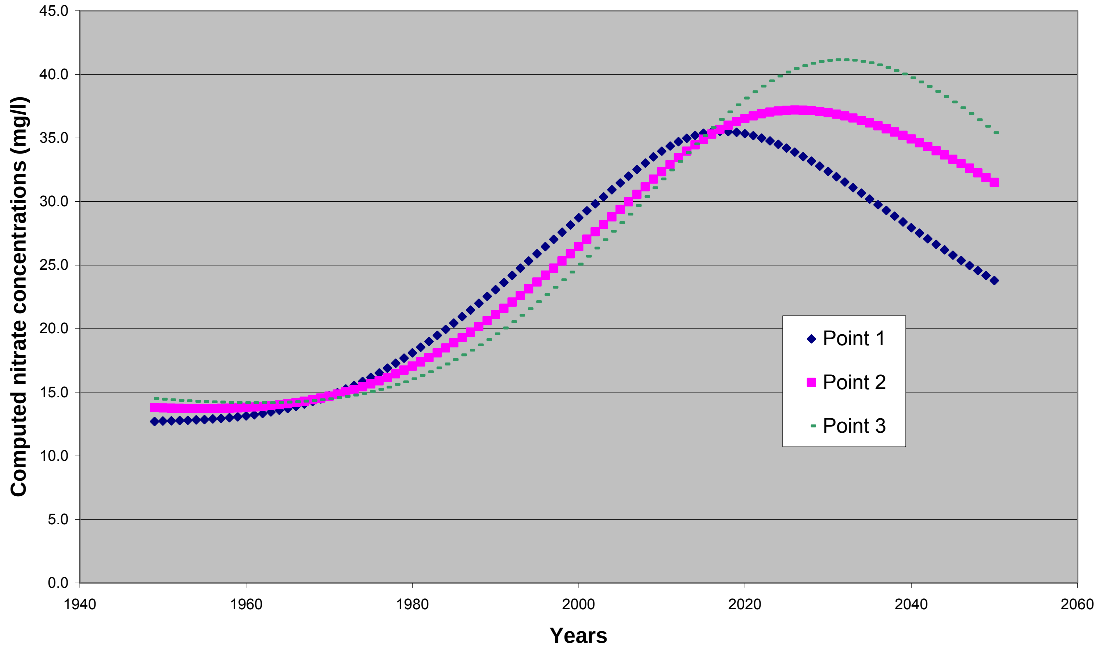

Figure 3.14. Time evolution of the nitrate concentrations computed with Scenario 2

Point Id	Depth to water table (m)	Year of trend reversal	Delay before trend reversal (year)
1	6.0	2017	7
2	15.6	2027	17
3	27	2034	24

Table 3.5. Relation between the depth to water table and the delay before the trend reversal

3.5 Conclusions and further works

In the framework of the AquaTerra project, HG-ULg has developed a spatially distributed physically based groundwater flow and solute transport model to reproduce and predict trends in groundwater quality in the Geer basin at the basin scale. The transport model has been calibrated using tritium and nitrate datasets. The model has proven its capability to represent nitrate concentrations on the order of measured ones and to reproduce upward trends. Due to the oversimplification of the nitrate input function, computed nitrate concentrations at specific locations in the basin can be affected by large uncertainty. However, this is only a matter of obtaining better estimates for the nitrate fluxes, which should be available from FUSAGx in the next months. The model has been also used to predict the time evolution until 2050 of nitrate concentrations in groundwater for two simplified scenario of nitrate input.

Further works will be devoted to the use of spatially distributed nitrate inputs as those computed by the EPIC-Grid model (UHAGx). A procedure has been identified, in collaboration with UHAGx, to transfer the data produced by the EPIC-Grid code as an input to the groundwater model.

4 Physically-deterministic determination and extrapolation of time trends at in the Brévilles' catchment (BRGM)

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4.1 Site description

The study site (Brévilles) is located 70 km west of Paris (49° 10' N, 1° 41' E). In this small (330 ha) catchment, silt loam and silty clay loam soils overlie unsaturated fractured (Lutetian) limestone, which in turn overlies the "Cuise sands" aquifer. A Parnassian clay layer constitutes the basement of the aquifer as shown in the schematic cross section through the study area (Figure 4.1). The groundwater system discharges via the Brévilles spring, which has been used locally for drinking water. Details on the geological and hydrogeological description of the Brévilles catchment are synthesized in AquaTerra Deliverable H2.4.

The water table quality and piezometric level have been monitored since 2001 in numerous piezometers and at the spring. The location of the various observation points is shown in Figure 4.2

The climate in the area is humid temperate with a mean annual rainfall of ca. 675 mm. The land use is predominantly arable, although forest covers ca. 25% of the catchment area, covering most of the steeper slopes with shallow water tables. Maize is currently cropped on ca. 20% of the arable land. Atrazine was used for the control of weeds in maize from the early 1960s and this has resulted in the contamination of the groundwater, with concentrations at the spring averaging around 0.2 µg/l in the period 1999–2003 (i.e. exceeding the EU legal threshold of 0.1 µg/l) with temporal variations between 0.07 and 0.43 µg/l. Atrazine use in the catchment was discontinued in April 1999 while the use of the spring for drinking water purposes ceased in 2002 due to atrazine contamination.

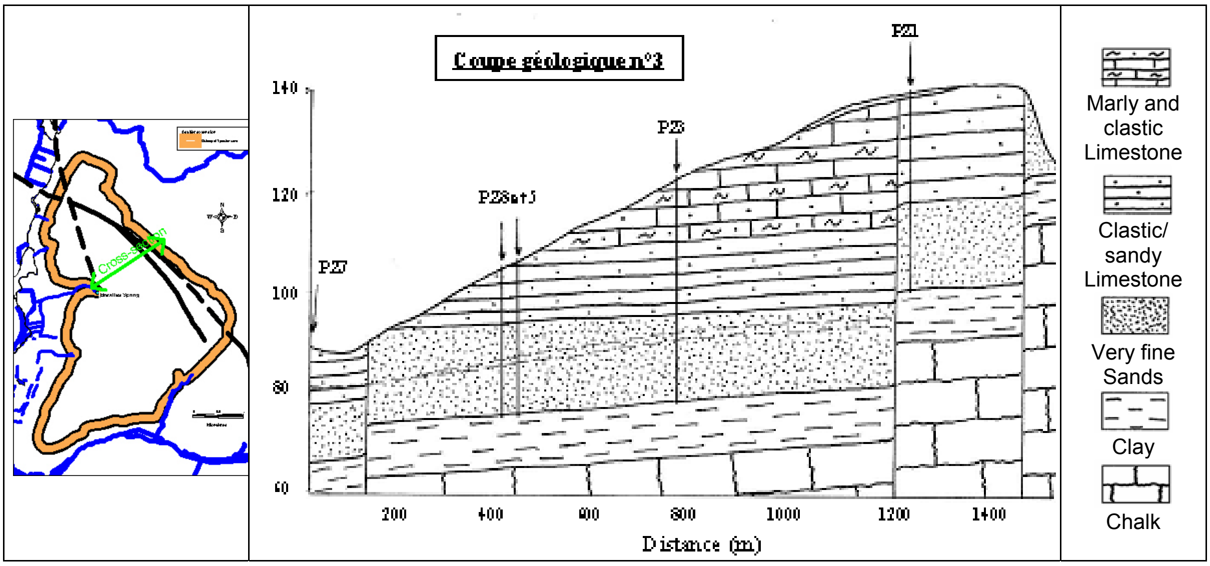

Figure 4.1: Schematic cross section through the study area

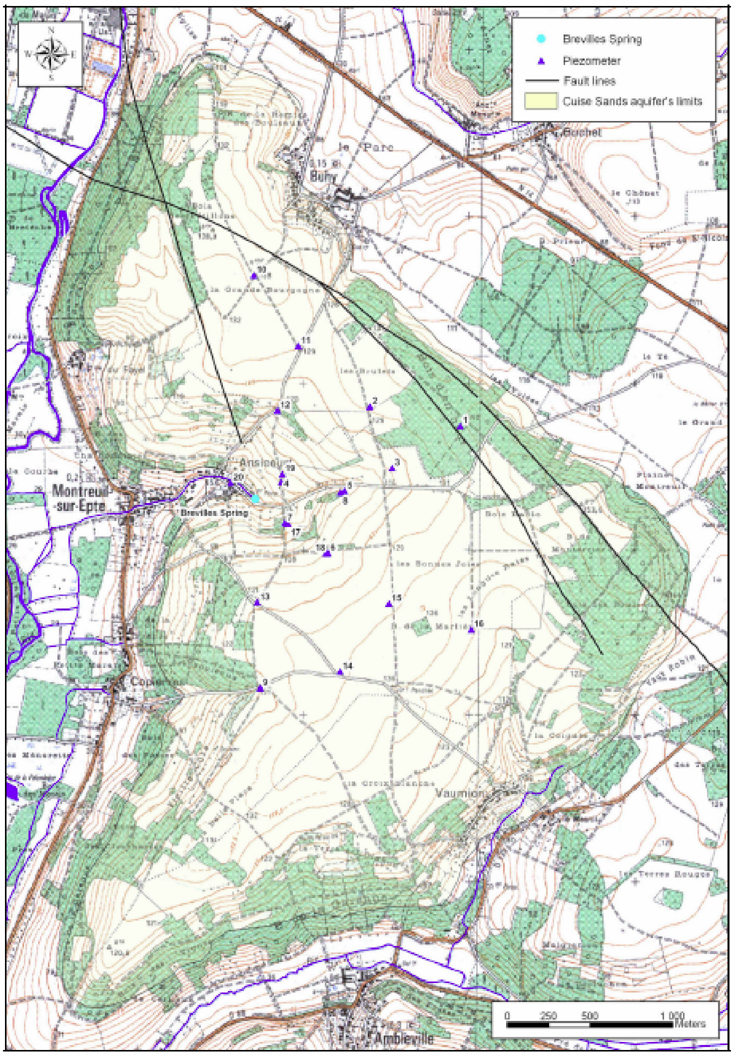

Figure 4.2: Map showing the extension of the "Cuise sands" aquifer and the location of the piezometers and the Brévilles spring.

4.2 Models and modelling strategy

4.2.1 Modelling strategy

The modelling study was aimed at simulating the gradual change in pesticide concentrations in the groundwater system and the spring for the past 10 years and the coming 30 years. A comprehensive modelling approach taking into account water flow and pesticide transfers in the soil, the unsaturated limestone and the “Cuise sands” aquifer was thus adopted. A coupled 1D-2D approach where results from an 1D soil leaching model (MACRO) were fed into a 2D groundwater model (MARTHE) was adopted. Percolation fluxes and pesticide concentrations in leachate calculated by the MACRO model at the bottom of the 1D leaching columns were used as input data in the 2D groundwater model to simulate the water flow and pesticide transfers in the aquifer system.
The MACRO running and the extraction of relevant data to be used as inputs in MARTHE were fully automated to allow a smoother sequential running of the two models. The combination of the two models is 'one-way' in that MARTHE does not have the possibility to feed information into MACRO. A conceptual view of the coupled transport model (1D MACRO/2D MARTHE) is presented in Figure 4.3.

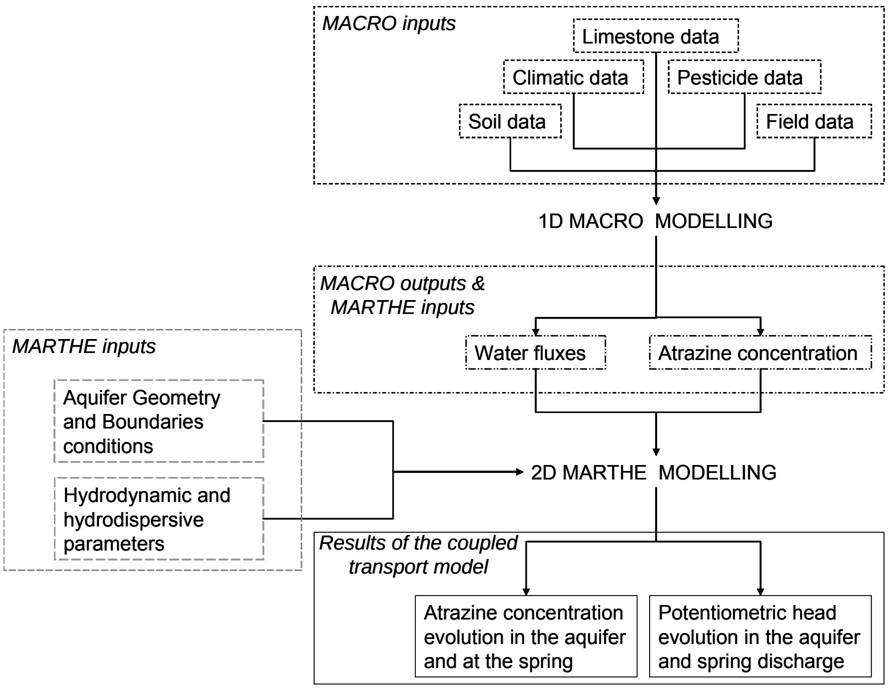

Figure 4.3: Conceptual view of the coupled transport model

4.2.2 Brief description of the two models used

<u>Description of MACRO</u>

MACRO is a physically based 1-D model which considers two flow domains (i.e. 'micropores' and 'macropores') to describe the transport of water and reactive solutes in soils. In the micropores, the water retention curve is described using the Van Genuchten function (Van Genuchten, 1980) while the hydraulic conductivity function is simulated using the Mualem's model (Mualem, 1976). In the macropores, flow is simulated using a non-capillary laminar flow process only driven by gravity. The

hydraulic conductivity in the macropores is expressed as a power function of the macropore water content. The division between micropores and macropores is made through the definition of a boundary water potential and the corresponding saturated micropore water content and hydraulic conductivity in the micropores. Water flow from macropores to micropores is described using a first-order approximation to the water diffusion equation while an instant transfer of excess water from micropores to macropores is assumed when the micropores become saturated.
Solute transport in the micropores is described using the convection-dispersion equation with a source-sink term representing mass exchange between flow domains. Solute transport in the macropore region is assumed to be a purely convective transport. The solute concentration in the water routed into the macropores at the soil surface is calculated assuming instantaneous equilibrium in a thin surface layer.

Description of MARTHE

MARTHE (Modelling Aquifers with an irregular Rectangular grid, Transport, Hydrodynamics and Exchanges) is a versatile 3D groundwater model designed to enable the hydrodynamic and hydro-dispersive modelling of groundwater flow in hydrosystems (Thiéry D., 1990, 1993, 1994, 1995; Thiéry D. & Golaz C., 2002). Hydrodynamic calculations are based on a fully implicit scheme with a 3D finite volumes discretisation. The system simulated may be represented with a classical confined/unconfined scheme or by an unsaturated zone – saturated zone continuum. In this latter case Richards' equation is solved with retention and relative hydraulic conductivity relations selected from a number of formulations (Van Genuchten, Brusaert, Brooks and Corey, Gardner, homographic, power law etc.). Advective, diffusive and dispersive transport can be simulated using three different techniques (donor cell, total variation diminishing (TVD) and method of characteristics using particles (MOC) depending on the situation considered (predominance of convection or dispersion). Calculations for energy temperature, mass and water fluxes are simultaneously fully coupled within the model.

4.3 Coupled transport model (1D MACRO and 2D MARTHE) for the Brévilles catchment & model calibration activities

The coupled transport model of the Brévilles basin carried out within the TREND2 module framework use i) a series of 1D MACRO models to simulate the water flow and the atrazine transport through the soil and the unsaturated zone and ii) a 2D groundwater model for flow and transport in the "Cuise sands" aquifer to simulate the pollution plume in the water table and its evolution according to several climatic scenarios.

4.3.1 Basin discretisation and MACRO modeling

MACRO being a 1D model, the Brévilles catchment was divided in a series of homogeneous zones to represent the catchment area ('homogeneous' meaning presenting similar behaviour with regard to pesticide transport). MACRO requires soil and agronomic data on a field-by-field basis over a significant number of years. The zones were therefore taken as every field x soil combinations over the 330 ha Brévilles catchment.

Data on land use were reconstructed by conducting interviews with the farmers and land-owners in the catchment. Information on crop rotation since 1994 was collected for 32 fields on the catchment.
Data on soils were collected by the excavation of soil pits on the site. Before the opening of the pits, soil variability was estimated using a spade and a hand auger with random sampling at along selected transects of the catchment. Soil types were reduced to 4 basic types with similar characteristics. One profile pit was excavated to describe each soil type and to collect soil samples. More information on soil and field data can be found in the AquaTerra Deliverable T2.7.
The number of soil x field combinations was determined using ARCMAP (ArcGIS 9, ESRI) through the crossing of the map for the 4 soils and the map with the 32 fields. Since the 4 soils were not represented in each of the 32 fields on the catchment, the total number of individual combinations was 83. The coding and method used for the crossing of the maps is explained in detail in the AquaTerra Deliverable T2.7.
83 MACRO parameterizations were undertaken to represent the 83 zones. Crop parameters were taken from FOCUS reports (FOCUS, 2001). Values for the soil parameters were estimated through a combination of pedotransfer functions (Hollis and Woods, 1989; Jarvis et al., 1997; Wösten et al., 1999) and by expert judgement. Atrazine sorption parameters were measured by batch experiments. Degradation was derived from half-life values extracted from the FOOTPRINT PPDB database (FOOTPRINT, 2007). Parameters for limestone were estimated by expert judgment based on results from an earlier modelling study (Roulier et al, 2006). Date and doses of atrazine applications are known since 1994, but atrazine applications started in the late 1960s. Hypothetical annual applications of 0.5kg/ha from 1973 to 1994 were therefore assumed in the modelling.
The outputs of interest from the 83 MACRO simulations included: i) daily predictions for percolation at the bottom of the leaching columns; and, ii) daily concentrations of atrazine in leachate. These outputs were fed into the MARTHE model.
The bottom of the leaching column was considered to be the head of the water table. To calculate the limestone layer thickness from the soil to the water table, the ground elevation map was subtracted to the elevation map of the average water table level with the ARCMAP software. The water table elevation map is the result of a MNR (Magnetic Nuclear Resonance) campaigns in 2005 combined with water heads measurements gathered in February 2006 (Deliverable C2.8).

In the zones where land use was not known precisely, the four land uses (one for each soil type) representing the largest surfaces in the known part of the catchment were associated to the soils.

4.3.2 2D MARTHE model and coupled transport model (1D MACRO with 2D MARTHE)

The 2D groundwater flow model of the “Cuise Sand” aquifer developed within the AquaTerra COMPUTE (Deliverable C2.8) was used as a starting point for the coupled transport model.
In the initial COMPUTE 2D flow model developed in MARTHE, the study area was discretised into 4675 meshes of 50 m side. The aquifer geometry was represented in the model by the topographic level and the bottom of the "Cuise Sands". The model was limited horizontally by the outcrops of the "Cuise sands" and the whole aquifer boundaries were simulated as no-flow boundaries. Since the water table was

unconfined, water table could overflow in each model cell whenever the water head arose above ground level. Spring discharges were computed as natural overflowing of the aquifer. The aquifer recharge was calculated from precipitation, evapotranspiration (PET) data and soil parameters using a GARDENIA scheme integrated in the MARTHE Software. This 2D flow model has been calibrated in transient flow condition over the period 1988-2006 with monthly time step (DL C2.10).

The COMPUTE model was improved and adapted in order to be coupled with the MACRO output results. Improvements were related to the following points:

- In the coupled transport model, the Northern and the Eastern part (Figure 4.4) of the catchment were not taken into account because the water table is often dried out in these parts of the basin. The reduction in the model extension in its northern part improved the model convergence greatly and reduced the computing time by a factor of 10. The boundary condition applied along the fault was of the imposed flow type except if the watershed overlapped the fault. In this case, a no flow boundary condition was applied. The imposed flow values were calculated from recharge calculated by MACRO in the northern part of the basin.

- The input files of the 2D MARTHE model were adapted to receive the output files of the 83 MACRO simulations. Daily water fluxes at the bottom of the leaching column and the daily concentrations of atrazine in leachate were fed into MARTHE. The delineation of the MARTHE zones in which these water fluxes and atrazine concentrations were applied is shown in Figure 4.5.

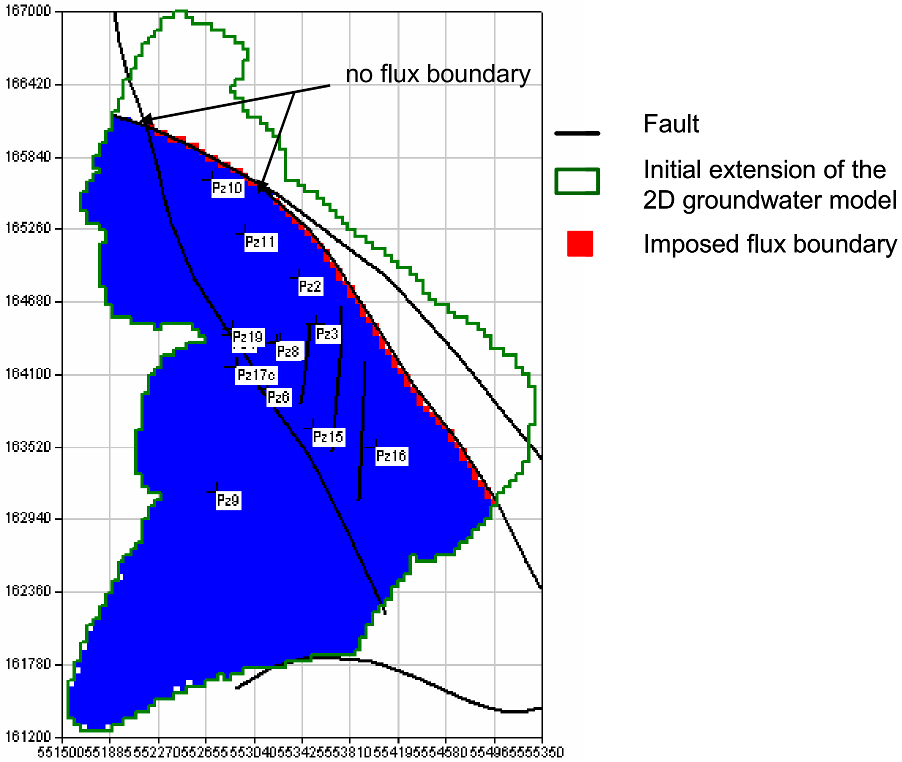

Figure 4.4: Reduction in the groundwater model extension used in coupled transport model and boundaries conditions defined along the fault.

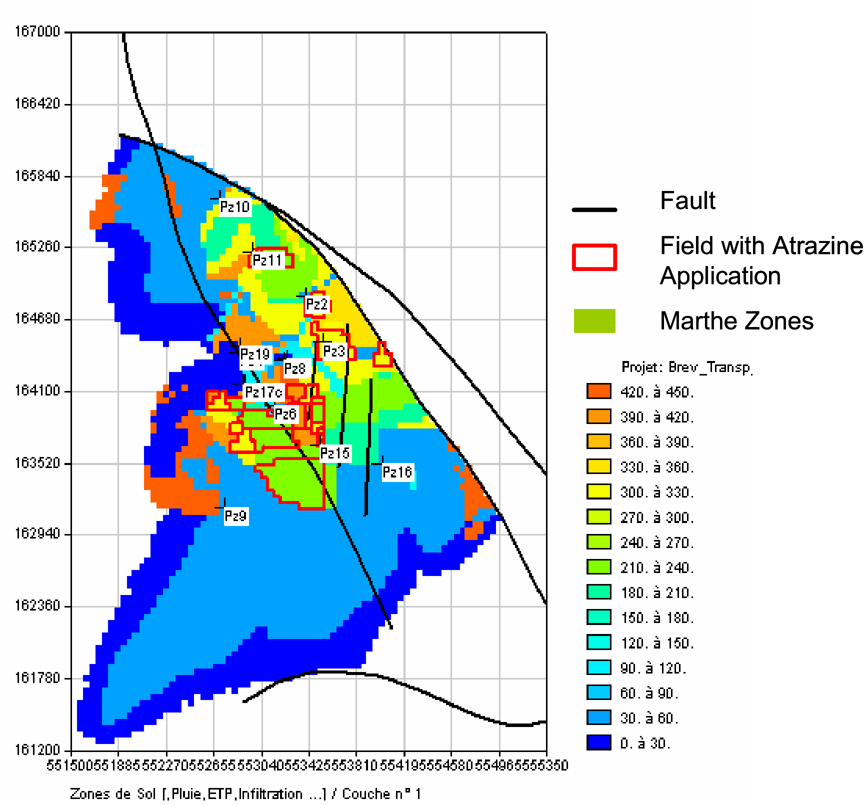

Figure 4.5: Recharge zones of the aquifer and location of fields in which an application of atrazine were taken into account in the coupled transport model. The colour interval corresponds to a coding related to soil and field combinations

Input data in 2D MARTHE transport model

The hydrodynamics parameters (permeability map and storage coefficient) introduced into the 2D transport model were those obtained within the COMPUTE module after the first model calibration over the period 1988-2006. Additional data were introduced to simulate the transport of the atrazine in the groundwater system.

A value of effective porosity equals to 0.14 was used for the Brévilles sand aquifer. This value was extracted from a tracer test interpretation (AquaTerra DL H2.3). The values of the longitudinal and the transversal dispersion were 50 m and 5 m, respectively. The diffusivity coefficient value used in the model was set to10^{-9} m^2/s.

Atrazine sorption experiments were carried out on saturated zone rock samples. These samples were collected at the time of the piezometers drillings on the site. Sorption coefficient (Kd) values for piezometers Pz1, Pz2, Pz7 & Pz8 ranged from 0.02 l/kg to 0.15 l/kg with an average value of 0.13 l/kg. In Pz4, the values reached 1.9 l/kg (PEGASE EU project, Report BRGM/RP-52897-FR). A single Kd value of 0.13 l/kg was used in the modelling to represent sorption in the aquifer system. Degradation experiments with aquifer sediments showed no significant degradation of atrazine in the system.

Simulations and model calibration

The coupled transport model of Brévilles basin was calibrated over the period 1988-2005 with a daily time step. A simulation over the period 1972-1987 was carried out for model initialisation. The atrazine concentration map and the piezometric map calculated in December 1987 were used to set the initial state of the aquifer for the calibration period.

The calibration of the 2D transport model was undertaken because the aquifer recharge calculated by GARDENIA in the previous modelling exercises was different from that coming out of MACRO.

4.3.3 Extrapolation

The climate was considered to be homogeneous over the study site since the catchment is limited in size. Rainfall data starting in 1988 were sourced from the Buhy station situated 500 m north of the study site. Potential evapotranspiration and temperature daily data starting from 2000 were taken from the Evreux station, which is located at 37 km west of Brévilles. For simulation purposes, the data collected were rearranged and a meteorological dataset from 1972 to 1987 was created. This extra period was used to allow the hydrology of the model to equilibrate and to apply atrazine before the studied period (1988-2006). To generate transient climate scenarios for the Brévilles catchment, a multi-model approach was adopted, using 13 Regional Climate Model (RCM) integrations (Figure 4.6) from the PRUDENCE project (Christensen et al., 2002). More detail on the RCMs can be found in Jacob et al. (2007). The Global Climatic Model (GCM) provided lateral boundary conditions for each RCM.

INSTITUTE	RCM	DRIVING GCM	PRUDENCE ACRONYM	AQUATERRA ACRONYM
DMI	HIRHAM	HadAM3H A2	HC1	HIRHAMH
DMI	HIRHAM	ECHAM4/OPYC A2	ecctrl	HIRHAME
HC	HadRM3P	HadAM3P A2	adeha	HADH
SMHI	RCAO	HadAM3H A2	HCCTL	RCAOH
SMHI	RCAO	ECHAM4/OPYC A2	MPICTL	RCAOE
Météo-France	Arpège	HadCM3 A2	DA9	ARPEGEH
ETH	CHRM	HadAM3H A2	HC_CTL	CHRMH
GKSS	CLM	HadAM3H A2	CTL	CLMH
MPI	REMO	HadAM3H A2	3003	REMOH
UCM	PROMES	HadAM3H A2	control	PROMESH
ICTP	RegCM	HadAM3H A2	ref	REGCMH
KNMI	RACMO	HadAM3H A2	HC1	RACMOH
METNO	HIRHAM	HadAM3H A2	HADCN	METNOH

Figure 4.6: The 13 RCMs generated by the PRUDENCE project. Alternative acronyms have been used within AquaTerra, the suffix of each denoting the driving GCM.

Simulations with the coupled model are only presented for the climatic dataset ARPEGEH in the present report. Several simulations with MACRO are available for some other climatic datasets.

4.4 Results & discussion

4.4.1 MACRO simulation results

Percolated water at the bottom of the unsaturated zone

Figure 4.7 displays a comparison of percolation predictions at 30-m depth for two soil types under an identical land use (Field #13) simulated with MACRO. The cumulated percolated water volume was 3300 mm over the period 1988-2005 (194 mm/year) for deep soils and 3800 mm (223 mm/year) for shallow soils. Daily maximum water volumes were found to be more important in shallow soils because of the influence of soil depth.

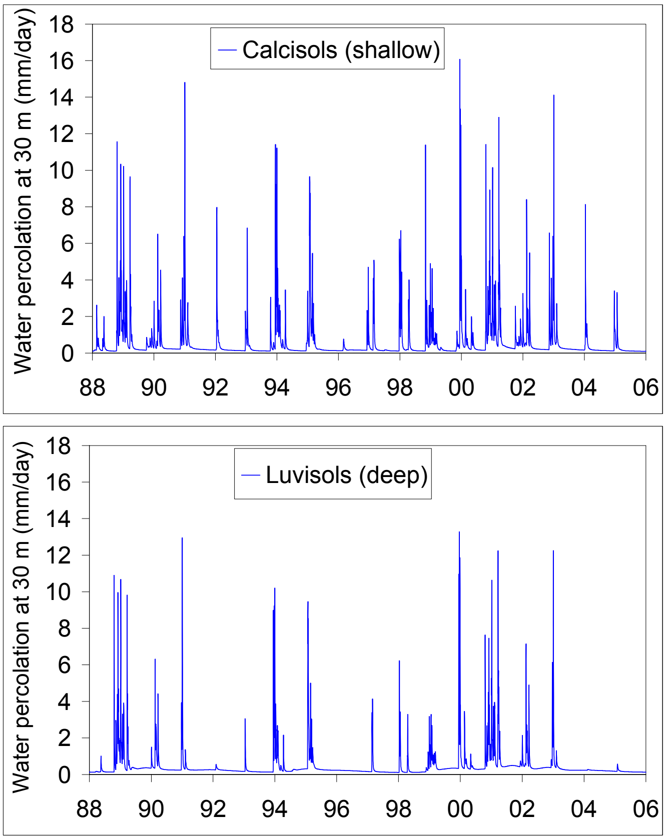

Figure 4.7: Comparison of predicted percolation volumes at 30-m depth in Calcisols and Luvisols for field #13 between 1988 and 2005

Atrazine concentrations in leachate at the bottom of the unsaturated zone

Figure 4.8 presents a comparison of predicted atrazine daily concentrations at 30 m below a calcisol and a luvisol. Despite the depth, concentration increases and decreases are predicted to be very fast, due to the importance of preferential flows in the limestone. The maximum concentrations are reached around 2000 below the two soil types but these are only 1.5 µg/l for calcisols and 4 µg/l for luvisols. Below some fields (#12 and #18) the largest concentrations were simulated around 1991 (no application after 1996). These two years (1991 and 2000) are the wettest in the study period. A second peak was simulated around the year 2000 below calcisols, but not below luvisols. Concentrations were larger on deeper soils because preferential flow has more importance on this type of soil. The importance of preferential flow also explains why there were some periods when atrazine percolation was zero below Luvisols. The transfer of atrazine was found to be slower, but more constant, in Calcisols.

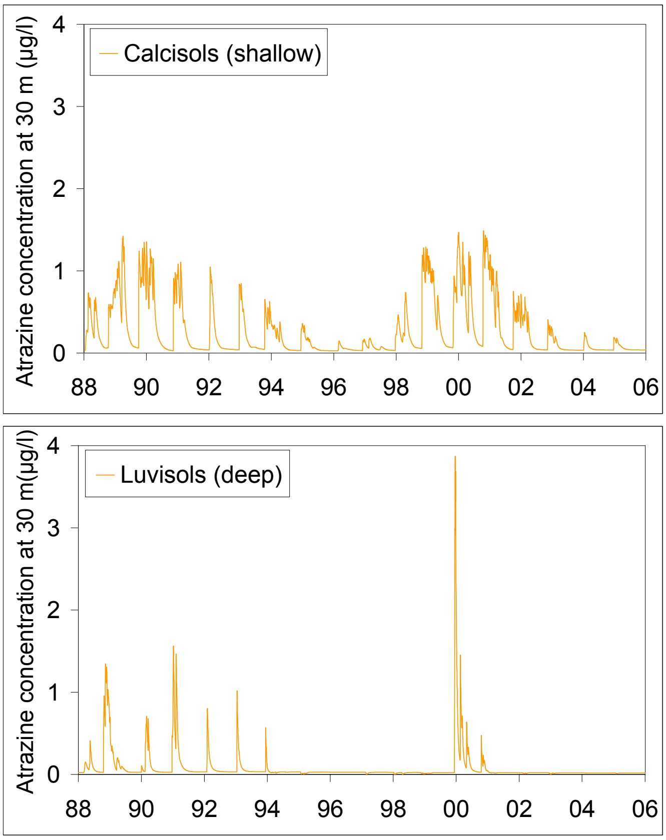

Figure 4.8: Comparison of predicted atrazine daily concentrations at 30-m depth below a calcisol and a luvisol for field #13 between 1988 and 2005

4.4.2 Coupled transport model results

<u>Changes in piezometric levels and spring discharge</u>

Figure 4.9 shows that piezometric levels calculated by the coupled transport model are in the same order of magnitude as those measured in piezometers Pz2, Pz4, Pz5, and Pz6. The variations of the water head in piezometers calculated by the model are comparable to the observed data except for the piezometer Pz2 where the model overestimates the water head. The model simulates faster recession of the water table. No seasonal fluctuations are observed on the site, but are simulated by the model. They are related to the fast flows simulated by the MACRO in fractured limestone, which generate fast variations of the water table. The very slow inertia measured on the water table shows that the preferential flows in limestone should be negligible and that the increase in the water table level is due to refilling through the limestone matrix and by lateral flow induced by the gradient of head.

Compared to the measured discharges at the Brévilles spring, the calculated discharges are rather well reproduced by the coupled transport model (Figure 4.10).

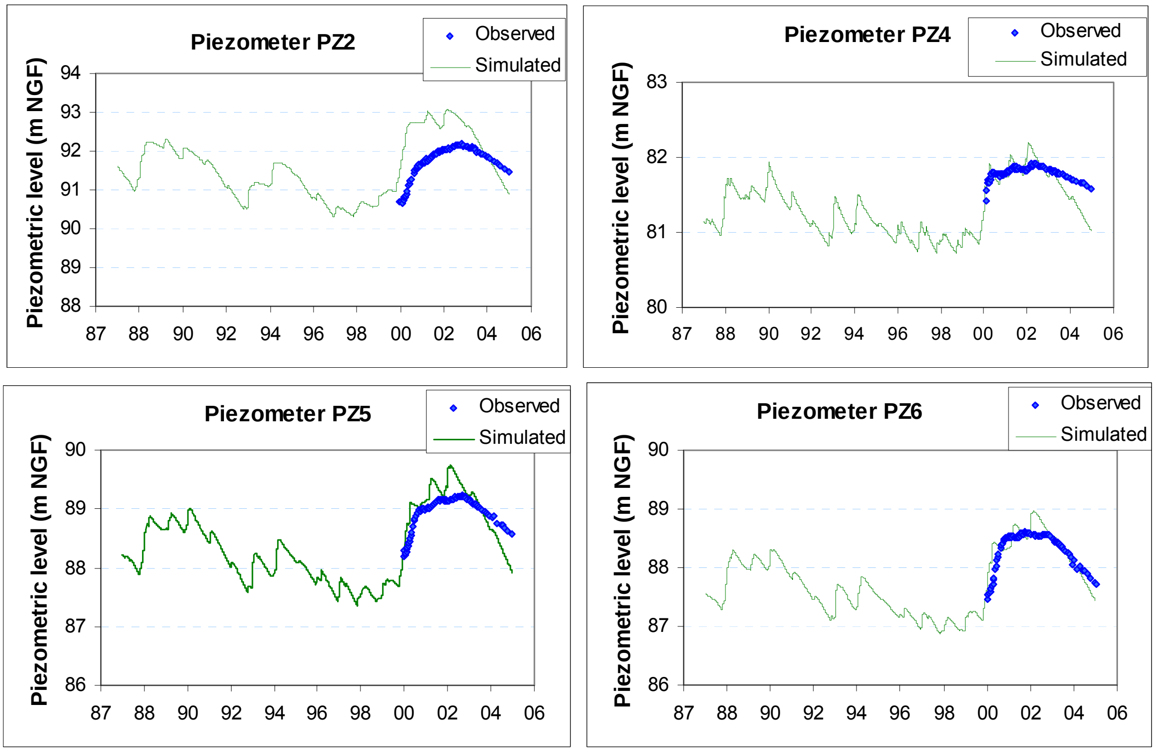

Figure 4.9: Measured and predicted piezometric levels by coupled transport model in piezometers (Pz2, Pz4, Pz5 and Pz6).

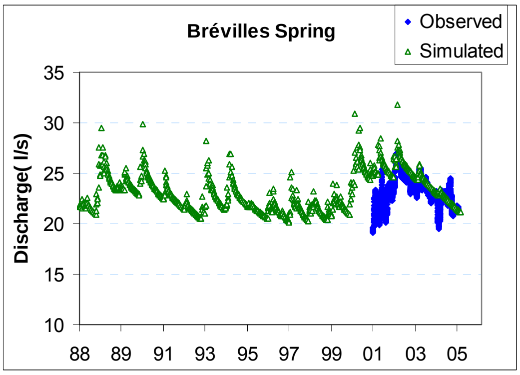

Figure 4.10: Measured and simulated discharge at Brévilles spring as predicted by the coupled transport model.

<u>Atrazine transport in the aquifer and concentrations calculated at the Brévilles spring</u>

Figure 4.11 shows atrazine concentrations calculated in December 2005 by the coupled transport model. Atrazine concentrations presented are an average for the wet part of the aquifer. In this map, the most significant predicted concentrations are in the south of Brévilles catchment and reach a value of 0.17 µg/l.

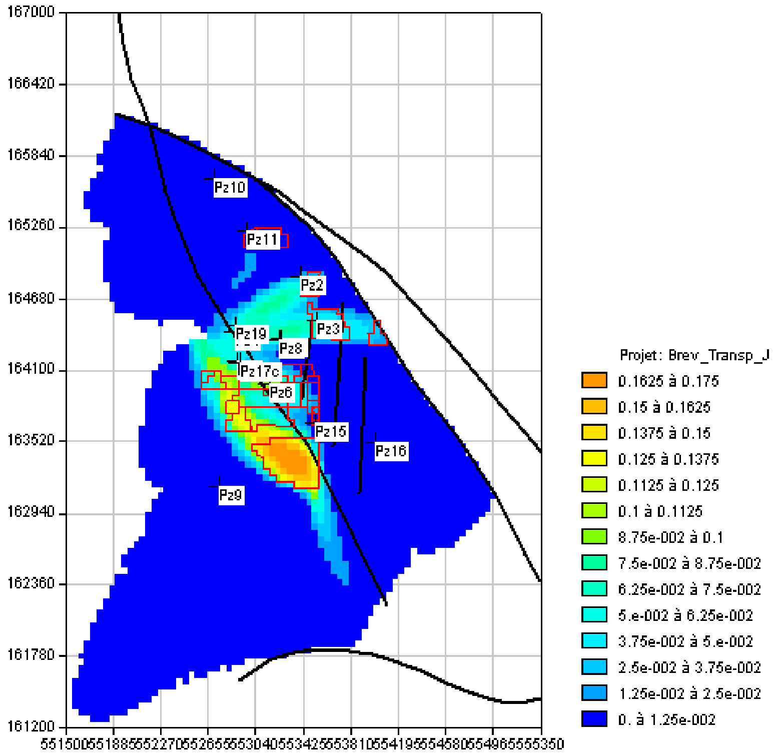

Figure 4.11: Predicted atrazine concentrations (µg/l) for December 2005 by the coupled transport model. Fields which have received applications of atrazine are shown with a red frame.

Figure 4.12 shows that the average concentrations are relatively well reproduced for the piezometers Pz2, Pz3 and Pz6. In contrast, predicted atrazine concentrations in Pz5 are very low compared with those observed. Pz5 is localised in a field in which maize was cultivated during several decades before the ban on atrazine in 2001 and the large concentrations measured in Pz5 (and in Pz8, located a few meters further) have probably been induced by a significant use of atrazine in this field. Since these applications were not integrated in the coupled transport model, the simulated concentrations are lower than the observed ones. Predicted concentrations in Pz5 only originate from fields located upstream in which applications of atrazine were taken into account in the coupled transport model.

Measured and simulated atrazine concentrations at the Brévilles spring are presented in Figure 4.13. The measured concentrations are slightly underestimated. The average predicted concentration is 0.1 µg/l whereas average measured concentration is 0.15 µg/l with a peak of 0.45 µg/l measured in 2001. This under-estimation is probably partly due to the under- estimation of concentrations in Pz5, which have a significant contribution to the spring pollution. An integration of atrazine applications in the Pz5 field in the coupled transport model would improve the average concentrations calculated in the piezometers Pz5, Pz8 and at the spring.

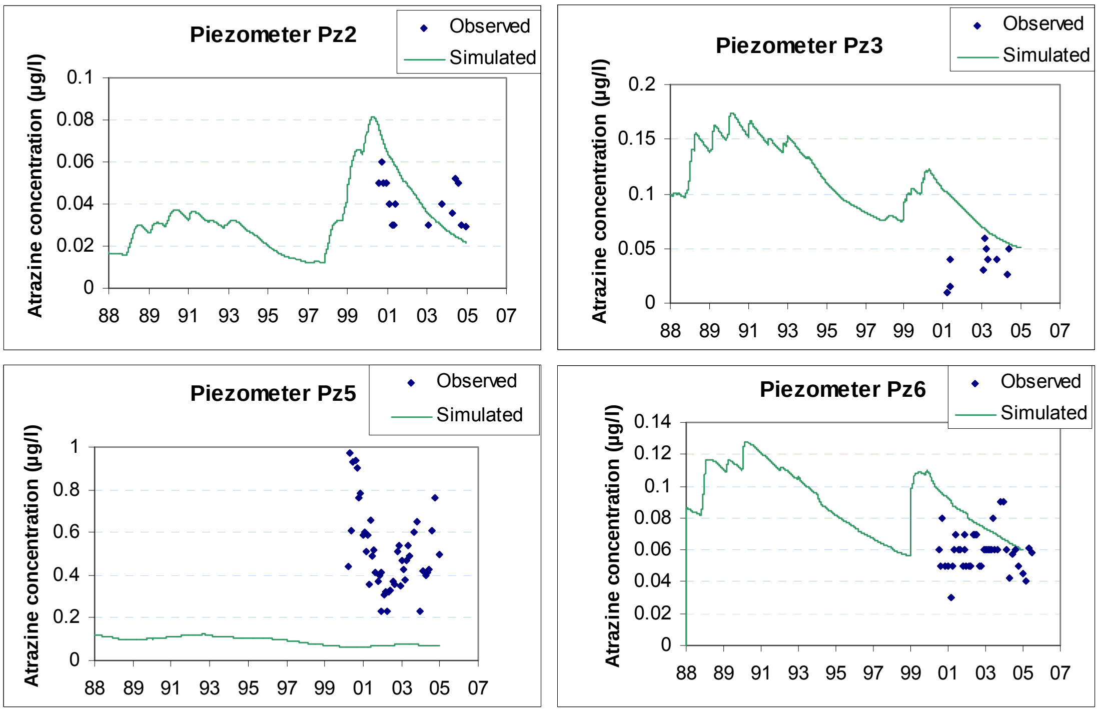

Figure 4.12: Measured and simulated values of atrazine concentrations in piezometers Pz2, Pz3, Pz5 & Pz6.

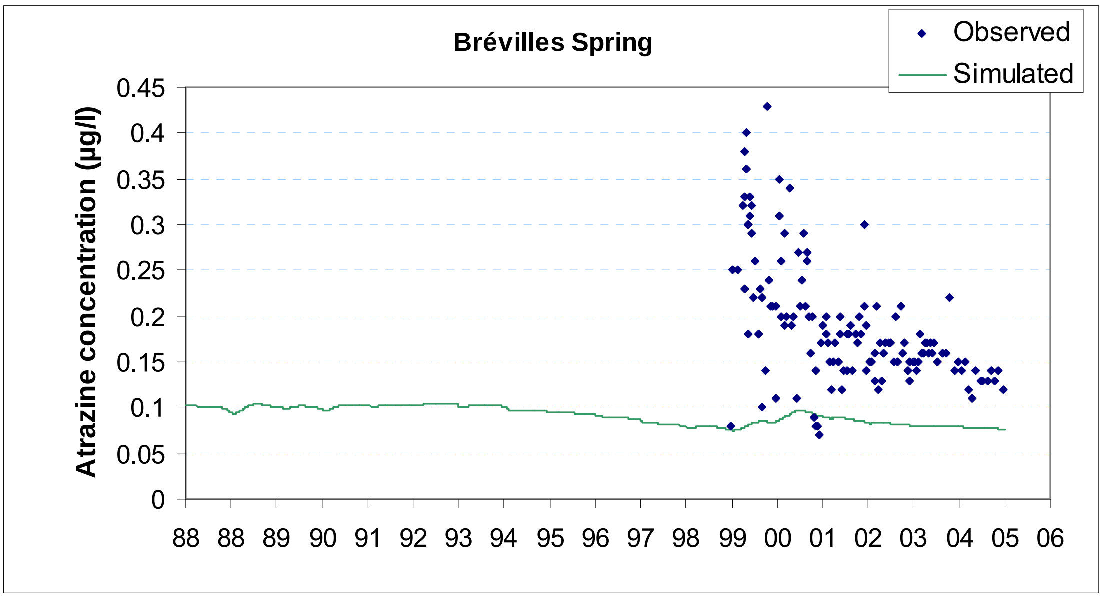

Figure 4.13: Measured and simulated atrazine concentration at the Brévilles Spring.

4.4.3 Extrapolation

<u>MACRO simulations</u>

Six climatic scenarios were used in MACRO to simulated water fluxes during the period 2005-2020. First results show that even if the amount of percolated water differs with the scenarios (Figure 4.14), changes in concentrations remain almost the same (Figure 4.15). In 2020, atrazine concentration is predicted to be around zero in all cases. The scenario with the highest annual volume of percolated water is ARPEGEH (240 mm/year) and the smallest is HIRHAME (204 mm/year) on the period 2005-2020. Over the period 1988-2005, the annual volume of percolated water was 194 mm/year.

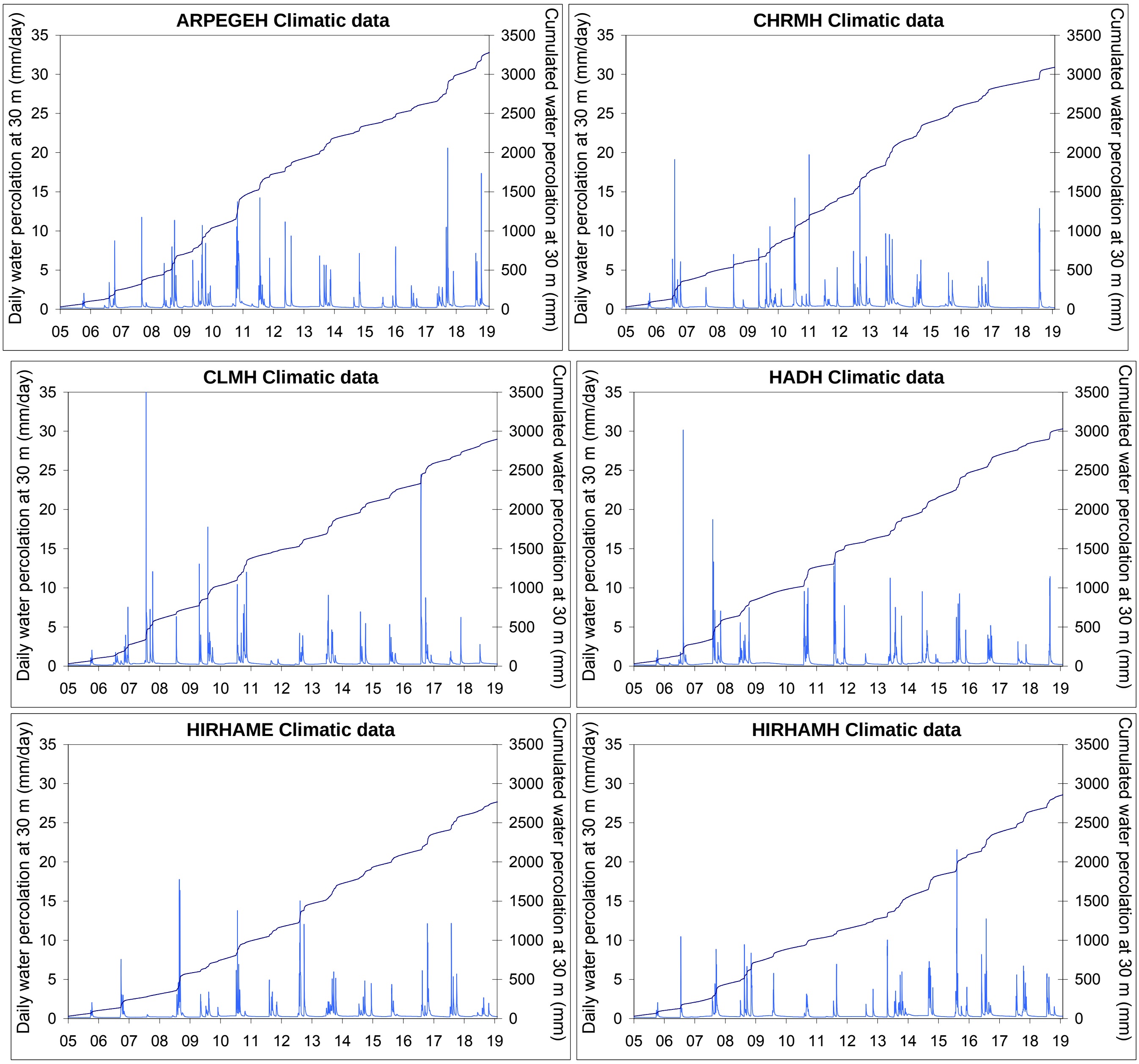

Figure 4.14: Percolated water volume at 30-m depth for field #13 between 2005 and 2020 for six climatic scenarios (ARPEGEH, CLMH, CHRMH, HADH, HIRHAME and HIRHAMH)

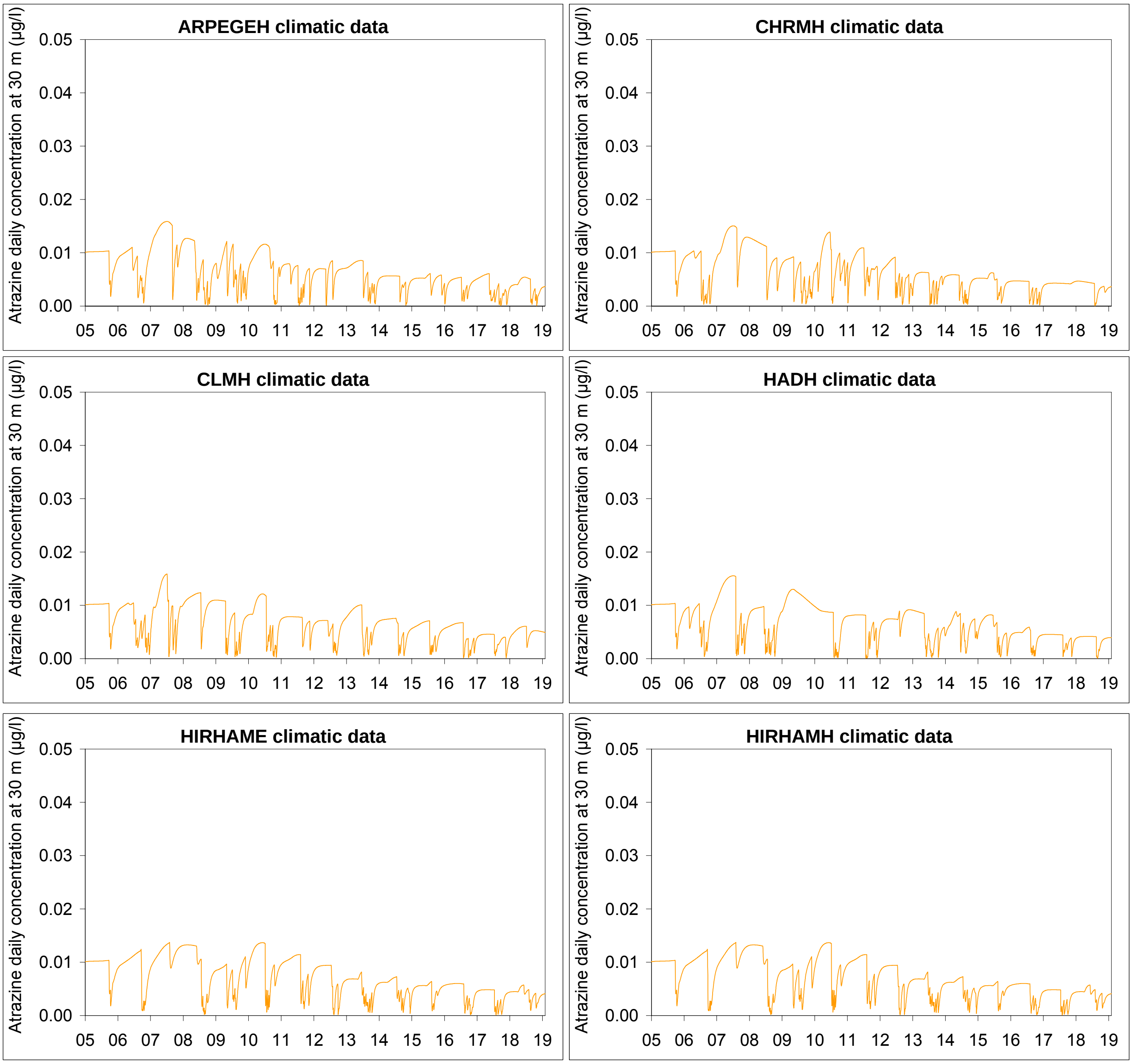

Figure 4.15: Predicted atrazine daily concentrations at 30-m depth for field #13 between 2005 and 2020 for six climatic scenarios (ARPEGEH, CLMH, CHRMH, HADH, HIRHAME and HIRHAMH).

Figure 4.16 presents a comparison of predictions for atrazine daily concentrations at 30-m depth for two soils (a calcisol and a luvisol) using the climatic scenario ARPEGEH. Concentrations were predicted to be larger after 2005 below calcisols because the profile depth for these soil types is much smaller than in luvisols. Also, water fluxes are more rapid in luvisols, which means that the large majority of the contamination will have been leached before 2001. In both types of soils, the trend is a gentle decrease in concentration with time. In both types of soils, the trend is a gentle decrease in concentration with time.

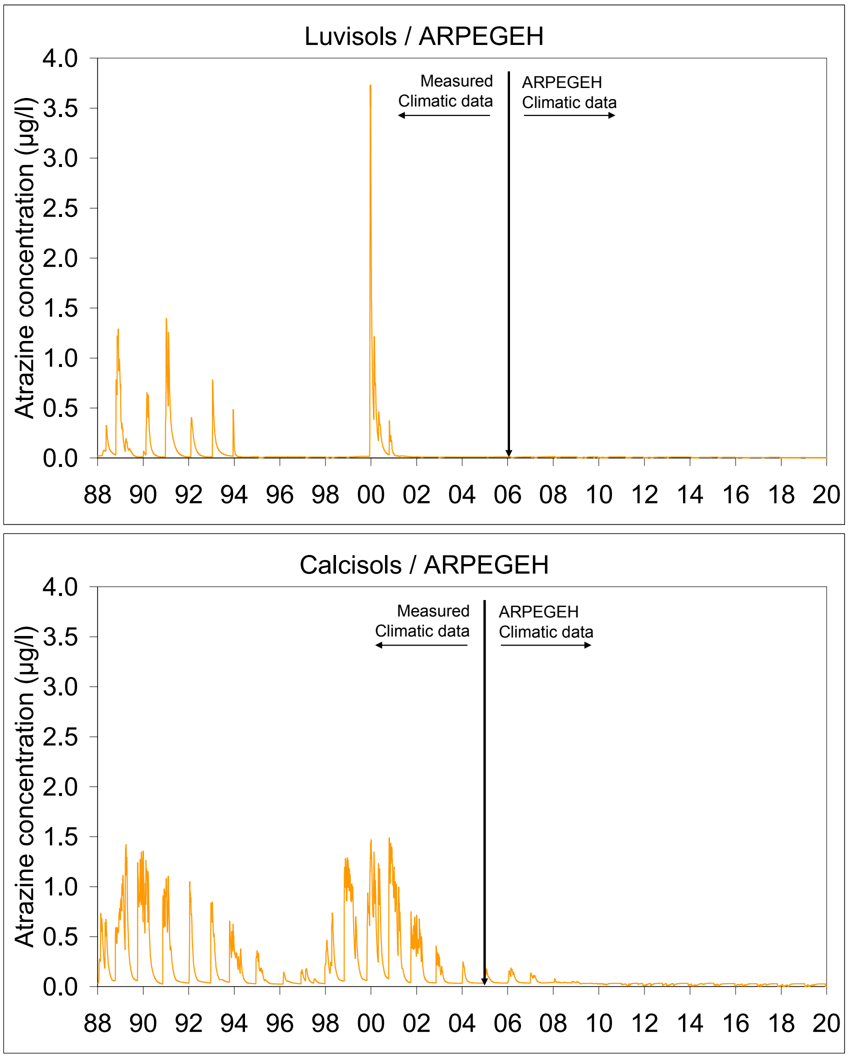

Figure 4.16: Predicted atrazine daily concentrations at 30-m depth for a calcisol and a luvisol for field #13 between 2000 and 2020

<u>Coupled transport model simulation</u>

Atrazine concentrations predicted in the aquifer and the spring over the period 2005-2020 were obtained based on the climatic scenario provided by ARPEGEH.

On Figure 4.17, the atrazine concentrations simulated over the period 2005-2020 are only drawn for the piezometers Pz2, Pz6 and Pz7 for which the average concentrations calculated by the model over the period of calibration are comparable with those measured. Since 2001 in the northern part of the Brévilles basin (Pz2), an exponential downward trend in concentrations is observed. For this piezometer, the calculated concentration in 2010 is 0.01µg/l. In the southern part of the Brévilles basin, the downward trend on Pz6 piezometer is faster until 2010 with an average concentration of 0.03 µg/l calculated at this date.

Atrazine concentration predicted at the Brévilles spring for the period 2005-2020 are displayed on Figure 4.18. Concentrations are predicted to decrease slowly from 2005 until 2010 to reach a value of 0.04 µg/l. In 2020, the forecasted value is 0.02 µg/l.

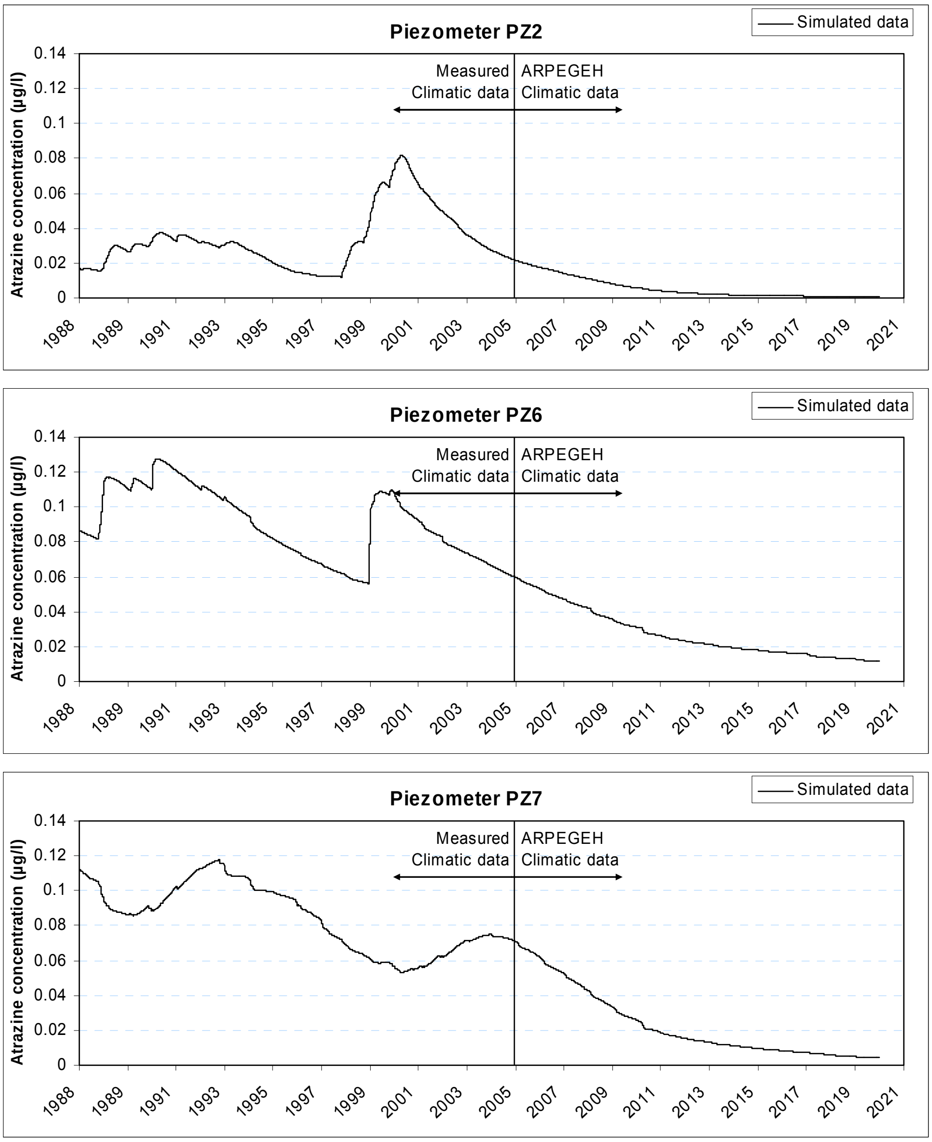

Figure 4.17: Atrazine daily concentrations predicted for the period 2005-2020 in the piezometers Pz2, Pz6 and Pz7

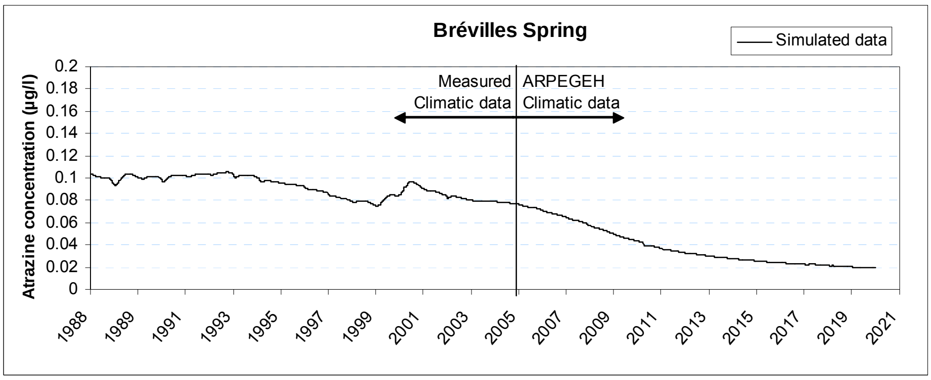

Figure 4.18: Atrazine daily concentrations predicted for the period 2005-2020 for the Brévilles spring

4.5 Conclusions & perspectives

A coupled transport modelling of the Brévilles' groundwater system has been achieved as part of the TREND2 to simulate changes over time in water fluxes and pesticide concentrations in piezometers and the Brévilles spring. In this coupled model, flow and pesticide transfer in soil and unsaturated limestone are simulated by 1D columns using the MACRO model. Percolation fluxes and concentrations of pesticides in leachate calculated by MACRO model at the bottom of the leaching column are used as input data in a 2D transport model in aquifer developed by the MARTHE software to simulate the water flow and pesticide transfer in the aquifer.
The coupled transport model has been initialized over the 1972-1987 period and then calibrated in unsteady state condition over the 1987-2005 period. Results obtained are promising and suggest that a good fit to the data would be obtained by further calibration activities. These could benefit from information on the sensitivity of the model to changes in selected input parameters such as parameters describing the presence of fractures in the limestone.
Predictions undertaken with climate change scenarios provided by the HYDRO module suggest that atrazine concentrations will decrease in the coming years to levels below regulatory concern. The modelling needs further adaptation to ascertain whether this would also be the case for its main metabolite DEA.

5 CONCLUSIONS AND IMPLEMENTATION OF RESULTS

The most complex tools to demonstrate and extrapolate trends in groundwater are the spatially distributed physically-deterministic models. These numerical models calculate the flow of groundwater through the system as well as the transport of contaminants by groundwater flow and are also referred to as “3D groundwater flow and transport models”. The TREND 2 partners TNO, HULg and BRGM each developed their own physically deterministic model, suitable for their respective study areas. The large diversity of study areas required different modelling approached from each team. The models were constructed, calibrated and tested against measurements and were proven capable of reproducing observed trends in groundwater quality.

Large 3D models are not well suited for predicting year-to-year variations because of the simplifications that are necessary to build the model. They are however capable of demonstrating trends in groundwater quality on time scales of decades, and also excellent tools to predict future trends in concentrations of contaminants, for example nitrate, zinc or atrazine, especially trends that will develop in the future but which are not yet present in the monitoring data. Furthermore, physically-deterministic models can also be used to predict trends in groundwater quality under changing pressures, such as land use or climate, using climate scenarios from different regional climate models provided by the HYDRO sub-project.

The following trends were predicted by the physically-deterministic models:

- in the Dutch part of the Meuse basin, nitrate and OXC concentrations will decrease in the near future under present day land use, but zinc concentrations will increase due to the leaching of zinc currently sorbed to the soil,
- in the Geer basin, nitrate concentrations will keep increasing at least up to 2050 under present day land use because of the long travel times in this system. Even when no new nitrate were applied, concentrations would increase over the next decade before decreasing,
- in the Brévilles catchment, atrazine concentrations will decrease in the coming years to levels below regulatory concern but the modelling needs further adaptation to ascertain whether this would also be the case for its main metabolite DEA.

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 for evaluating future scenarios and watershed management. 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. In specific areas of high ecological, sustainability or economical importance physically-deterministic models could and should be implemented as a tool to detect and predict trends in groundwater quality

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