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title: Modeling nitrate transfer in an alluvial aquifer for estimating tendencies and short and medium term evolution of groundwater quality

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INTRODUCTION AND CONTEXT

The objective of the project was the modelling of nitrate transfer from soil to the outflow of an alluvial aquifer. The model will permit to estimate tendencies and evolution of NO₃ concentration in groundwater under various socio-economical scenarios. The multi-tools study included the use of environmental tracers to calculate water and pollutant transit time within the alluvial aquifer, geochemical data for the evaluation of the groundwater-surface water interaction, 1D modelling of nitrate transfer from soil to groundwater through the unsaturated zone, 2D modelling for the estimation of the nitrate transport within the aquifer and socio-economical assessment of agro-environmental measures that would permit a change in groundwater nitrate concentrations.

The studied site of about 260 km² is located in the eastern part of France, in a region dominated by maize crop (Fig. 1).



Figure 1. Localisation of the studied area - the Ain River alluvial plain, France.

The studied area covered the southern part of the groundwater body FRGW3676 untitled Alluvial Ain River plain. The plain is crossed by the Ain River and bordered by the Rhône River. The Ain River is considered to be isolated from the alluvial plain. Water and solutes exchanges may occur only in a narrow flooding area close to the river bed. The Rhône River is the natural aquifer exsurgence. The aquifer is mainly recharged by precipitation but also received water from the Dombes plain (North-West and West), intensively cultivated and from the Bugey and Jura karstic Mountains (North-East and Eastern part).

The alluvial aquifer is mainly used for drinking water of small size towns and for irrigation.

RESULTS AND DISCUSSION

Geochemical data

Two field sampling campaigns carried out in July 2008 and July 2009 permitted the collect of ground and surface water for the analyses of major and trace dissolved ions, δ^{2} H, δ^{18} O, CFC-11, CFC-12, CFC-113, SF₆, ³H (Fig. 1).

Groundwater is of HCO₃-Ca type. Nitrate concentrations of groundwater vary from 5.2 mg·l⁻¹ to 84.6 mg·l⁻¹. The northern part of the alluvial aquifer presents lower nitrates concentration. The highest NO₃ concentrations are measured in the south-east part of the aquifer in Loyettes sector.

Stable isotope data (δ^{18} O, δ^{2} H) was analysed at 23 groundwater wells and the two major Rivers crossing the area, the Ain and the Rhône Rivers (Fig. 2a). The local meteoric line (LML) is represented by Thonon-les-Bains precipitation (AIEA/WMO GNIP station). This meteorological station is located 150 km s upstream the alluvial plain in a mountainous region. Topographic and geographic data explained relatively depleted δ^{18} O and δ^{2} H values of "local" precipitation. The Worldwide Meteoric Water line (WML) is δ^{2} H = $8 \cdot \delta^{18}$ O +10 (Craig, 1961). The Rhône River is influenced by the Alps snow melting and high altitude precipitation and surface water is therefore showing low stable isotope values. Water of the Ain River is from the Jura Mountain and aquifers discharged. Isotope data of this water are closed to the alluvial groundwater values.



Figure 2. δ 180 and δ 2H of samples collected in 2008 in the alluvial aquifer, Ain and Rhône Rivers and weighted mean of regional rainfall collected at Thonon-les-Bains.

In the Figure 2b the two points representing the Ain River upstream and downstream are indistinguishable. Groundwater is plotted along the local meteorological line except for 4 points. At Loyettes south east part of the plain maize cropping is covering large areas and irrigation is needed from May to September and intensively from June to August. In July, sampling period, it is possible that the two wells Loyettes/corne and Loyettes/trompette were influenced by irrigation return. The two groundwater samples with highest deuterium excess are not drilled in the morainal and Miocene formations. Stable isotope data is confirming that the Ain River is quite independent to groundwater. The aquifer presents a high hydrodynamic heterogeneity due to glacio-fluvial sediment deposits of varied granulometry and lithology.

Age-dating using CFC-11, CFC-12, CFC-113, SF $_6$ and 3H gave a mean recharge date of 5 to 10 years.

Modelling nitrate tranfer and hydrodynamics

The transfer of nitrate from soil to groundwater thought the unsaturated zone is estimated using a global model called BICHE developed by the BRGM (Thiéry, 1990). The data on N cycle (amount of N used, plant needs, soil nitrogen mineralization, and N mineralization from vegetal crop residues) is coupled with a global hydrological model using ETP and precipitation (Fig. 3).





The collect of data from as much as possible years was necessary for nitrate transfer modelling.

Data of nitrate concentration in groundwater is usually available from 1991 up to now. Frequency, duration and nitrate variability and concentrations vary within the numerous monitoring stations available in the studied area (Fig. 4).



Figure 4. Nitrate concentrations in groundwater at two monitoring stations.

Thirteen more representative chronicles were selected for modelling purposes.

The hydrological input parameters (P and ETP) are the same than the ones used for the hydrodynamical modelling. The N cycle information is reconstituted from information given by the literature (Yara, 2004), agricultural enquiries, farmers' registry and soil and agricultural institutes.

Calibration and validation of the model (Fig. 5) was done using a great amount of data of various origin and type. This is a necessary step in order to use site specific modelling.



Figure 5. Nitrate transfer simulation using BICHE and observed concentration of nitrate in groundwater at site.

The hydrodynamic and hydrodispersive modelling of the entire alluvial plain was performed using MARTHE calculation code (Thiéry, 2004). It was calibrated on 8 years period at a 10 days time step, from 1999 to 2007. Results of the transitory regime calibration are satisfactory and allowed the use of this model for nitrate transfer estimation. For that purpose the 1D BICHE model outputs (flux of nitrate to the aquifer through time) was used as an entry of the 2D ni-

trate transfer modelling within the alluvial aquifer. In order to obtain spatial information, 12 homogeneous sectors were defined. Each sector has similar agricultural practices and hydrody-namic behaviour. For each sector it exists, at least, one chronicle of nitrate concentration in groundwater.

The coupled models is being used to assess the efficiency of various scenarios proposing changes (or not) in agricultural practices (crop type and fertilizer use).

The scenarios were prepared based on socio-economic studies considering today's practices and near future (< 2027) changes in land use and land management practices. Various scenarios were prepared after discussions with local stake holders and only four contrasted ones were finally selected. Such experiments have been conducted in other sites and using various calculation codes site specific (e.a. Krause et al., 2008) or simplified methods (e.a. Jackson et al, 2008).

Dating and modelling

Dating groundwater at a sampling point location means: determine the elapsed time (or residence time) from the time the water goes underground and became isolated from the atmosphere (i.e.: during its infiltration into the soil from precipitation or any other type of surface water) until it is removed/collected. The information obtained on the age of the water, and therefore its turnover time, is useful for management of aquifers.

There are two methods to estimate the age of groundwater. The first is based on the hydrodynamics of the system studied (Metcalfe et al., 1998). But the difficulty lies in the spatial distribution of permeability field that is generally known that in some points. On the other hand, it requires a set of sufficient data in order to achieve a satisfactory calibration model. This implies an uncertainty on the velocities and ages calculated. A second approach called "direct" is based on the use of tracers, geochemical or isotopic. In shallow aquifers, tracers most commonly used are events tracers that are related to a sudden or continuous contribution in the underground environment (³H, ⁸⁵Kr, CFC, SF₆, ...). Their introduction into the natural environment dates from the twentieth century. For these compounds few studies seek to compare independently the hydrodynamic and tracers approaches. The use of both methods of dating can make a "feedback" to improve understanding of the conceptual model. This comparison could provide valuable information on the hydrogeological understanding of the Ain alluvial plain.

However, the question arises of the physical meaning of "ages" obtained by the models of geochemical dating. Knowledge of an apparent age does not necessarily imply knowledge of a residence time of water. In this study we evaluated the limits of different models for estimating residence time obtained by the use of event-tracers. Comparisons will be made using the following hydrodynamic models; discretized type, global or developed black box.

Different approaches are possible for the comparison of tracer and hydrodynamical models. The first approach is to reproduce the contents of CFCs and SF₆ by modelling as tested for the study of a shallow aquifer in Germany (Duke et al., 1993). A multi-tracer approach improved modelling results in reducing uncertainty and enhanced the importance of CFC-113 adsorption. A second approach compared the ages obtained by CFC age-dating and residence time obtained by modelling. This method was used in a shallow sandy aquifer in Maryland (Reuilly et al., 1994). A first level calibration using various tracers ³H, CFC-11, CFC-12 highlighted the pheno-

menon of water mixing. A second level of calibration taking into account the results obtained using the CFC-dating led to the improvement of the age and the conceptual model.

In addition to these two methods, we will test a third approach consisting in entering CFC data in the hydrodynamical models in the aim of simulate its comportment in the system.

CONCLUSIONS

The system of the alluvial Ain river plain is an ideal study site for the use of tracers CFCs given the fast transfer velocities and known hydrodynamical characteristics. Using data ³H, CFCs and SF₆ and the hydrodynamic model validated on observed hydrodynamic perspective of this study are:

- Determine the properties of conservation and solubilization of tracers in the unsaturated and saturated zones,
- Judge the relevance of the geochemical approach and try to improve it by using the hydrodynamic model coupled to solute transport,
- Comparing the information obtained using the hydrogeological and geochemical models to enable a better definition of "age" of water and its meanings,
- Validating hypotheses and results obtained in the nitrate transfer modeling.

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