



## Long-term transfer of diffuse pollution at catchment scale: Respective roles of soil, and the unsaturated and saturated zones (Brévilles, France)

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### SUMMARY

Environmental management such as recommended by the European Water Framework Directive requires actions regarding diffuse pollution. However, the efficiency of these actions depends on the reaction time of the hydrosystem. In order to predict future groundwater-quality trends, especially after implementation of environmental measures such as substitution of pesticides or reduction of nitrogen loads, the response time has to be determined. In the case of diffuse pollution, the response time between fertilizer or pesticide application and the subsequent contamination of a spring or well may depend on the distance between the area where the products are spread and the point where water quality is measured. However, some aquifer systems may appear particularly slow, independently of the size of the catchment. At Montreuil-sur-Epte (Val d'Oise, France), no significant change in groundwater quality can be related to the suppression of atrazine application over the small agricultural catchment (4 sq km) of the Brévilles spring, 8 years ago (4 years before the national ban of atrazine). Besides the 8 year monitoring of groundwater quality at the spring and in several observation wells in its catchment, a particular attention was paid to the characterization of water and solute-infiltration rates at different scales. The role of each compartment (soil, and unsaturated and saturated zones) in the slowing down of flow and the transfer of solutes (pesticides and nitrate) from top soil to aquifer, and then to the spring, was determined by a multidisciplinary approach involving mainly soil science, hydrogeology and geochemistry. Detailed investigations aimed at identifying and quantifying the parameters that contribute to the inertia of the system, resulting in a long-term transfer of diffuse pollution. Data covering the monitoring of pesticide transfer in soil, the physico-chemical characteristics of soils, profiles of water contents, tritium activities, and nitrate concentrations measured on rock samples collected by drilling, were compared in order to describe the transfer time in soil and within several meters of unsaturated zone. Tracer tests in groundwater completed the understanding of transport in the saturated zone near the spring. Geochemical measurements and pumping tests confirmed the stratification of the saturated sandy aquifer leading to heterogeneous groundwater contamination. Finally, the inertia of the system results from several factors linked to the hydraulic properties of each underground compartment combined to intrinsic properties of the contaminants, partially explaining the limited improvement of groundwater quality 8 years after a major change in agricultural practice. The observations and results obtained by applying various tools and methods in this catchment led to a reflexion on sampling frequency, methodology and metrology to be adapted to this particular, but not unique, hydrogeological context in order to assess the groundwater-quality evolution and to understand the fate of solutes. The results of this study also raise questions on how efficient and how fast will the positive impact of product substitution or environmental regulations be.

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### Introduction

When studying diffuse pollution, hydrogeology is a key factor in any attempt to predict the evolution of contamination. Pesticides or nitrate applied at soil surface are conveyed by meteoric water through the soil and vadose zone to reach groundwater and its

observation points, which are generally drinking-water supply points. With enhanced monitoring programmes developed during the past 10 years (EEA, 1999; IFEN, 2004), contaminations by pesticides and nitrate have been noted throughout Europe. In metropolitan France, 55% of groundwater observation points are affected by pesticides (IFEN, 2007). The Public Health Code fixes the regulatory measures for drinking water, pursuant to European directive 98/83/CE. For pesticides in drinking water, the quality limits are fixed at  $0.10 \mu\text{g L}^{-1}$  for each pesticide (except for aldrin,

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dieldrin, heptachlor and heptachloroepoxyde) and  $0.5 \mu\text{g L}^{-1}$  for the total of measured substances.

The European Water Framework Directive (WFD – 2000/60/EC, OJEC, 2000) has set objectives aiming to achieve a good quantitative and chemical status of groundwater bodies by 2015. In order to estimate whether the observed contaminations will diminish and pass below the European threshold, or to determine if non-contaminated aquifers will remain safe, one should be able to understand the evolutionary trends of pollutant concentrations in underground systems.

Most of the research into the environmental fate of pesticides conducted during the past 30 years, focused on the soil compartment only (FOCUS Soil Modelling Work Group, 1997; Reus et al., 1999), which generally represents less than 10% of the vertical distance and less or much less than 0.1% of the horizontal transfer distance. This specific interest in the soil compartment was mainly due to the fact that pesticide concentrations will rapidly evolve in contact with the water and organic matter present in soil, with organic matter providing sites for physical and chemical adsorption. Soil, hosting a rich microbial population, is the place where the degradation of pesticides is optimum. This is also the place where most nitrate is consumed by plants or transformed in other nitrogen forms.

For pesticides, it is commonly considered that less than 1% of application doses pass beyond the soil compartment (Flury, 1996). However, in many cases, preferential flow in soil results in increased flow and transfer velocities, limiting the processes of sorption and degradation of the soil (Flury, 1996) and underlining the role of hydrodynamics on the leached quantity. The question that arises then concerns degradation: will the product(s) degrade in the other compartments i.e. unsaturated and saturated zones? Studies covering the unsaturated zone deeper than the soil layer and the saturated zone are rare. Regarding atrazine, degradation could theoretically take place in the vadose zone (Issa and Wood, 1999). However, Johnson et al. (2000) showed that atrazine is not degraded in a saturated zone of a chalk aquifer. Kristensen et al. (2001) demonstrated that the mineralization of aged atrazine in aquifer chalk with inoculated bacteria known to mineralize atrazine, was reduced with increasing residence time of the atrazine. Degradation kinetics in the unsaturated and saturated zones are clearly much slower than those observed in the soil compartment. However, as transfer times can take years or decades, even low degradation rates can result in a substantial reduction of concentration during the underground course of the contaminant.

Consequently, studies of diffuse pollution and particularly pesticide contamination should not be restricted to the soil compartment. The time needed to reach groundwater and the water observation points is paramount when evaluating contamination trends. This time is governed by hydrogeological parameters and physico-chemical interactions with the rock matrix.

Regarding pesticides, a particularly significant change in European agricultural practice was the ban of using atrazine in various countries (1991 in Germany, 2003 in France), but, to our knowledge, very few publications are available on the evolution of groundwater quality after the stop of using this pesticide (Tappe et al., 2002; Morvan et al., 2006; Baran et al., 2007, 2008). These publications, limited in number, demonstrated the presence of atrazine in groundwater several years after the last application. Several hypotheses can be put forward to explain this persistence.

Besides the probable existence of a pesticide stock in the soil and in the unsaturated or saturated zones that can be remobilized, the time needed to renew the water of the saturated zone, which includes the transfer time in the unsaturated zone, could be a determinant factor for explaining the persistence of contamination.

The present study aims at demonstrating the role of hydrogeological processes in pesticide transfer. It took place in a site chosen for its small size (an aquifer of less than  $12 \text{ km}^2$  within which the Brévilles spring catchment, recently revised thanks to additional drilling and geophysical surveys, accounts for about  $4 \text{ km}^2$ ), its agricultural context, a known contamination with pesticides and nitrate, and a simple hydrogeological configuration characterized by the absence of withdrawals and streams. In this unconfined aquifer, pesticide monitoring was simultaneously started in the soil, the unsaturated and saturated zones when the use of atrazine was stopped and substituted by acetochlor, an herbicide recently registered in France. This study provided a unique opportunity to study the fate and transfer of a pesticide in a context of substitution.

The objectives of this study were thus: (i) to characterize the impact of hydrodynamic processes on the transfer of contaminants by distinguishing each compartment from the soil-unsaturated zone-saturated zone continuum; (ii) to evaluate the relevance and representativeness of punctual data gathered by “classical” hydrogeological techniques; and (iii) to assess the validity of interpretation results—when starting from punctual data—on water resources management, from a quality point of view.

## Materials and methods

### *The Brévilles spring catchment*

The Brévilles spring is the main spring of a small aquifer of  $12 \text{ km}^2$  in extension located 70 km west of Paris (Fig. 1). The water table is mainly located in a sandy layer (“Cuisse sands” – Upper Ypresian) overlying an impervious clay layer of 10 m thickness, and overlain by heterogeneous marine clastic limestone of Lutetian age, becoming marly at the top of the formation. Bartonian sand and silicified limestone (millstones) occur in the highest places. Superimposed on the solid rocks are more recent loamy drift deposits of colluvium in shallow valleys, table-land loam covering part of the high ground, and stoneless drift soil covering the sand and clay around the site.

The layering is tabular, although slightly tilted westward and affected by faults, the most important of these forming the eastern limit of the aquifer. As a result of this structure, the water table does not emerge on the eastern brim of the aquifer. Most of the springs are located along the western border, contributing to the Epte river valley (tributary of the Seine River).

The 350 inhabitants of the village of Montreuil-sur-Epte used the Brévilles spring for their drinking-water supply until atrazine was found in concentrations exceeding legal thresholds, and use of the spring was discontinued in August 2001. In an effort to reduce contamination of the spring by atrazine, farmers in the catchment used acetochlor, an alternative for weed control in maize. Acetochlor was first applied in May 2000, at the same time as pesticide monitoring was started in the soil, the unsaturated zone and groundwater, which provided a unique opportunity for studying pesticide detection and fate in a context of product substitution.

### *Hydrodynamic and hydrogeological characterization at different scales*

From top to bottom, each compartment has been studied with the objective of identifying and quantifying the key parameters that rule water and mass transfer. Their spatial variability was also tackled, interpolating when possible the punctual data.

### *The soil compartment*

Soil characteristics and variability were investigated through field work carried out in collaboration with the French national

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