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

Deliverable No.: **H2.7**
Title: **Interpretation of a tracer test at Brévilles after 2 years of monitoring.**
HGULg and BRGM

Due date of deliverable: March, 2008
Actual submission date: March, 2008

Start date of project: 01 June 2004 **Duration: 60 months**
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Revision: J.Barth ; H.J.Fowler

Project co-funded by the European Commission within the Sixth Framework Programme (2002-2006)		
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

To understand and predict the behaviour and transfer of diffuse contamination, a small catchment is intensively studied in the vicinity of Paris. A multi tracer test involving a new technique, the FVPDM method (Finite Volume Point Dilution Method - Brouyère et al., 2008) has been performed in natural flow conditions. Injections of four different tracers (uranine, sulforhodamine B, lithium chloride and potassium iodide) took place in four piezometers involving different areas of the aquifer and different depth. This tracer test follows a former test briefly described. A particularly long monitoring (Nov-2005 to Feb-2008) demonstrates the existence of several transport velocities within the sandy layer, which seems linked to the decrease of hydraulic conductivity with depth. Absence of recovery of two tracers confirms the results of the former test and identifies spatial heterogeneities probably due to the geological structure. The new insight and parameter quantification brought by interpretation of these tests contributes to a better characterization of the saturated zone and shall be exploited through modelling for transport predictions of pesticides.

MILESTONES REACHED (from DOW3 and 4)

DL H2.7 estimates the range of groundwater flow velocities in the saturated zone and quantifies parameters, which are needed in the prediction modeling carried out in the modules TRENDS 2 and COMPUTE 2.
This deliverable also give clues to the understanding of chemical spatial and temporal variations (including pesticide concentrations) in the aquifer and at its main outlet (the Brévilles Spring), to be used within HYDRO2 and FLUXES1.

Note that this deliverable is in the form of a manuscript to be submitted, and therefore does not adhere strictly to the usual format of deliverables.

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Tracer tests in the sandy aquifer of the Brévilles Spring (France) : making the tracer experiment more profitable.

Abstract

To understand and predict the behaviour and transfer of diffuse contamination, a small catchment is intensively studied in the vicinity of Paris. A multi tracer test involving a new technique, the FVPDM method (Brouyère et al., 2008) has been performed in natural flow conditions. Injections of four different tracers (uranine, sulforhodamine B, lithium chloride and potassium iodide) took place in four piezometers involving different lateral and depth distributions in the aquifer.. A particularly long monitoring (Nov-2005 to Feb-2008) demonstrates the existence of several transport velocities within the sandy layer, which seems linked to the decrease of hydraulic conductivity with depth. Absence of recovery of two tracers confirms the results of the former test and identifies spatial heterogeneities probably due to the geological structure. The new insight and parameter quantification brought by interpretation of these tests contributes to a better characterization of the saturated zone and shall be further exploited through modelling for transport predictions of pesticides.

Introduction

The sandy aquifer of the Brévilles spring is located in Montreuil-sur-Epte in the Val d'Oise, about 70 km North-West of Paris (France). The area is characterised by intensive agricultural activities, inducing nitrates and pesticides accumulation problems. At the Brévilles spring, the concentration of atrazine in water has exceeded the tolerance for drinking purpose (0.1 μg/L), and water withdrawals for public distribution have been interrupted since 2001 (Morvan, 2004). The Brévilles spring catchment has been chosen as a test site by BRGM and has been studied for 8 years in the scope of a EU FP5 project (PEGASE) and, more recently, in the FP6-IP AquaTerra project. The objective is a better understanding and prediction of the fate of pesticides in the subsurface. The Hydrogeology Group from University of Liège has been involved in this research with the task of performing tracer experiments between newly drilled piezometers and the Brévilles spring. The objectives of these experiments, performed in November 2005, are threefold:

1. to highlight vertical variations in groundwater fluxes, related to vertical variations in grain size distribution and hydraulic conductivity,
2. to estimate contaminant travel time from several locations in the catchment to the Brévilles spring,
3. to identify transport processes affecting the fate of solutes in the saturated part of the aquifer and to quantify associated parameters (effective porosity, dispersivity etc).

Besides being a remarkable case study with particularly long recovery breakthrough curves, these tracer experiments provide interesting data and new insights for the understanding of flow and transport functioning in a stratified sandy aquifer, the tracer experiments performed at the Brévilles spring test site also illustrate a good methodology of performing comprehensive tracer experiments campaigns. They took advantages of all information potentially available during and after the test, in order to make tracer experiments more profitable. Indeed, tracer tests are money and time consuming and most attention is usually focused exclusively on the recovery step. Moreover, due to the lack of knowledge and complexity of the underground environment, most tracers cannot completely be recovered at the selected monitoring points. It therefore often happens that tracer tests do not deliver the answers to the questions posed.

The methodology used for the multi tracer injections performed at Brévilles ensures a monitoring at two different levels. A 'traditional' recovery monitoring was performed at the aquifer's outlet or other chosen recovery point. Depending on the test site context, this traditional monitoring may be enhanced by choosing several sampling points along the assumed streamlines. The analysis of the breakthrough curves may provide information on flow and transport behaviour in different parts of the aquifer. We also applied a new technique, were the the tracer concentration was also monitored and measured in the injection well during the injection itself. This technique is known as the Finite Volume Point Dilution Method (Brouyère 2001, 2003, Brouyère *et al.* 2005, Brouyère *et al.* 2008) and enables better control of the entrance of tracer material in the aquifer. It also allows estimating accurately local groundwater fluxes near the injection point. The combination of the monitoring at these two levels (in the injections wells and at the outlet of the aquifer) provides more results from the same tracer test. This prevents dependency on unpredictable aspects of traditional tracer experiments and promises results even in cases of no recovery.

Geological and hydrogeological context of the Brévilles catchment

The aquifer associated to the Brévilles spring is mainly located in the Cuisian sandy formation limited at its base by impermeable clay and at the top by Lutetian limestone and marl. These sands are medium sands in the upper part of the formation to very fine sands in the lower part. Due to the relief and the structure of the geologic layers, the small aquifer can be individualized and identified as hydrogeologically independent (Figure 1). Along the western border of the aquifer, a spring line is observed along the outcropping limit between the sands and the clays. The aquifer extents over approximately 11.6 km^2, and the Brévilles spring constitutes its main outlet. In order to continuously measure water flow rate, a gauging station has been installed 200 m downstream the spring. All across the year, the water flow rate is significantly more important at this station ($\approx$ 21 to 28 l/s) than at the spring itself ($\approx$ 6 to 8 l/s). This indicates that the spring does not drain the total thickness of the aquifer. Based on topographical considerations, and assuming groundwater stratification, it is probable that the spring drains groundwater coming from the upper part of the sandy layer only. Groundwater from the lower part of the aquifer is assumed to flow out diffusively between the spring and the gauging station (SP12).

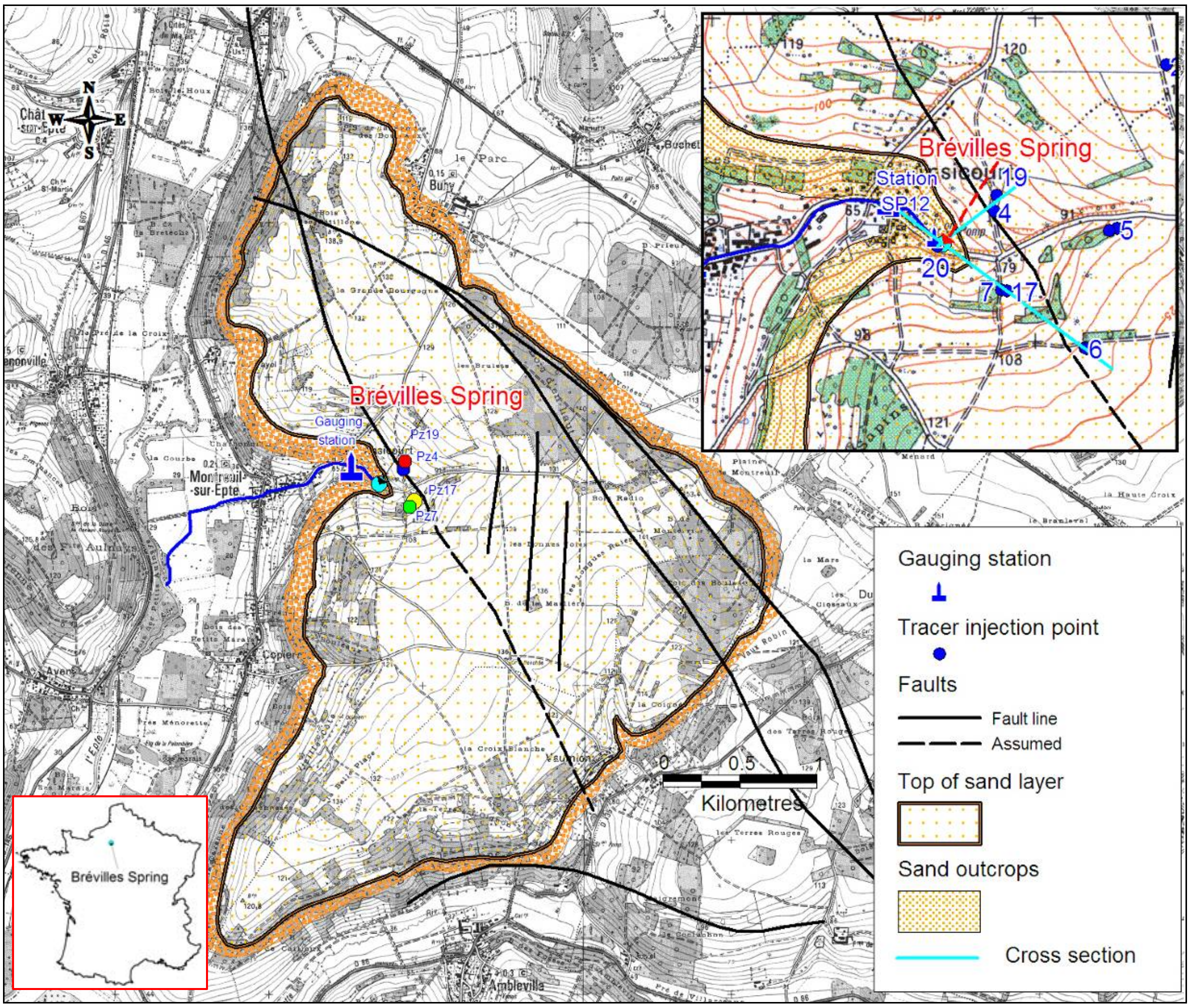


Figure 1 : The Brévilles test site (injection area)

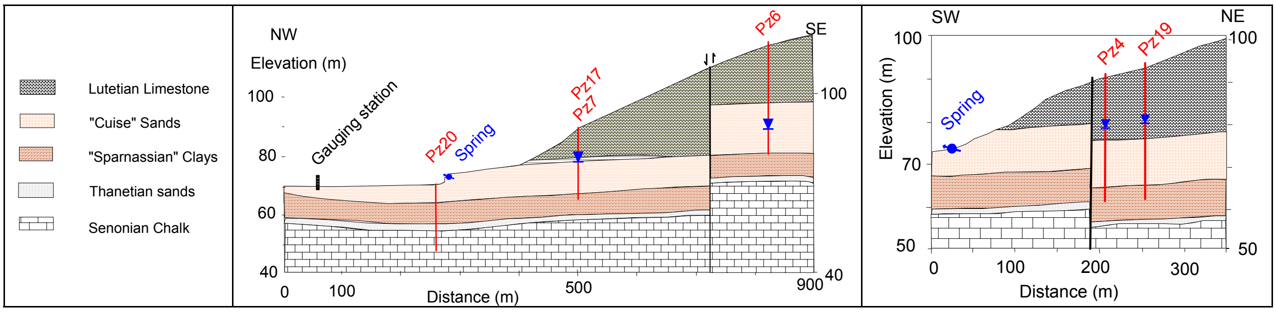


Figure 2 : Geological cross sections

Tracer tests campaigns

Previous campaign

Previously to the new tracer experiments of November 2005, two other tests were performed by BRGM in 2003. These more classical tracer experiments had given contrasting results and led to new interrogations and tests. Uranine was injected in PZ4 (0.465 kg, tracing distance : 187 m) and sulforhodamine G were injected in PZ7 (0.388 kg, tracing distance : 228 m). Samples were collected at the Brévilles spring.

Sulforhodamine G was first observed at the spring 7 days after the injection. The tracer concentration rose quite rapidly and then stabilized, with an almost constant concentration that was observed for about 1 year. Approximately 300 days after injection, 22.6 % of the tracer mass was recovered at the spring. This atypical concentration evolution was attributed to tracer capturing in the underground, probably close to the injection point. The complete breakthrough curve and the recovered mass evolution are presented in Figure 3. The main characteristics of these curves are summed up in Table 1.

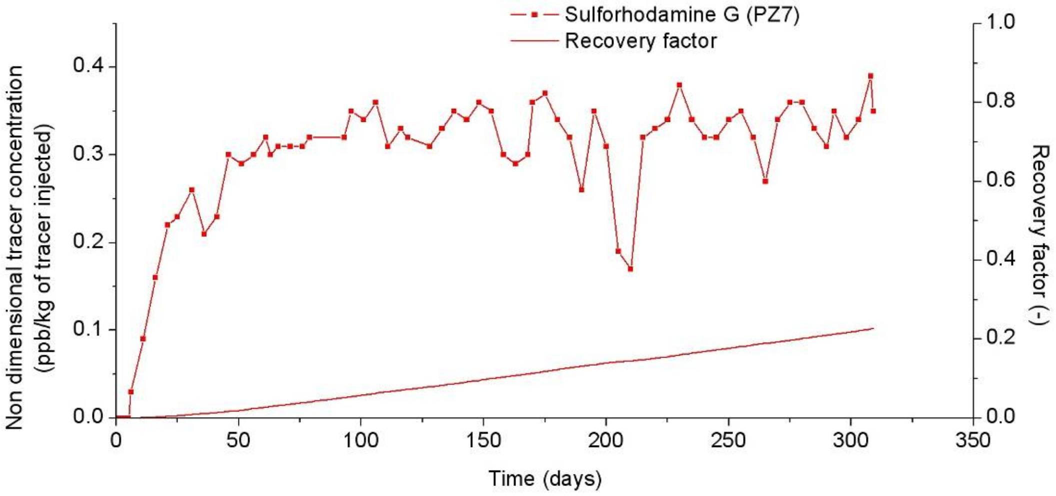


Figure 3 : Breakthrough curve and recovered mass evolution for the tracer test performed between PZ7 and Brévilles spring.

First detection time	7 days
Beginning of the stabilized stage	61 days
Mean concentration during the stabilized stage	0.124 ppb 0.32 ppb/kg of tracer injected
Calculated speed corresponding to the first detection	32.57 m/day 3.77×10^{-4} m/s
Calculated speed corresponding to the beginning of the stabilized stage	3.74 m/day 4.33×10^{-5} m/s
Recovery factor after 309 days	22.67%

Table 1: Main characteristics of the breakthrough curve (Sulforhodamine G – Tracer test 2003)

The uranine tracer injected in PZ4 was never detected at the spring (sampling duration: about 1 year). Several reasons may explain the absence of recovery:

- PZ4 is only screened at the bottom of the sandy aquifer, where the hydraulic conductivity is supposed to be lower and chemical parameters indicate a vertical stratification of groundwater. This may have caused the the tracer to migrate very slowly, as it may have been affected by dilution and dispersion so that concentrations were driven below the detection limit.

- Geophysical surveys showed that PZ4, located near a fault crossing the aquifer, could be in a relatively disturbed zone. This piezometer could thus be isolated in a collapsed zone, with little or no circulation. This is corroborated by the absence of tritium, nitrates and pesticides in this piezometer.
- Further sampling operations were performed at the Brévilles spring only. As mentioned above, it is assumed that the spring drains mostly water from the upper part of the sandy aquifer. It is thus possible that uranine was not detected at the spring because it arrived downstream of the sampling location. Simulations performed using a 2D model developed using MARTHE (BRGM modelling code, Thiéry, 1989; Thiéry et Gutierrez 2007) also showed that the tracer plume could have arrived between the spring and the gauging station and was therefore missed.

New campaign

In order to validate or invalidate the above hypotheses and to obtain more exploitable experimental data, new tracer experiments were performed in November 2005. Different tracers were injected in 4 piezometers (PZ17b, PZ17c, PZ4 and PZ19) located near the Brévilles spring (Figure 4). The four tracer experiments were dimensioned considering two main goals, at two different levels and times : injection monitoring and recovery monitoring at the Brévilles spring.

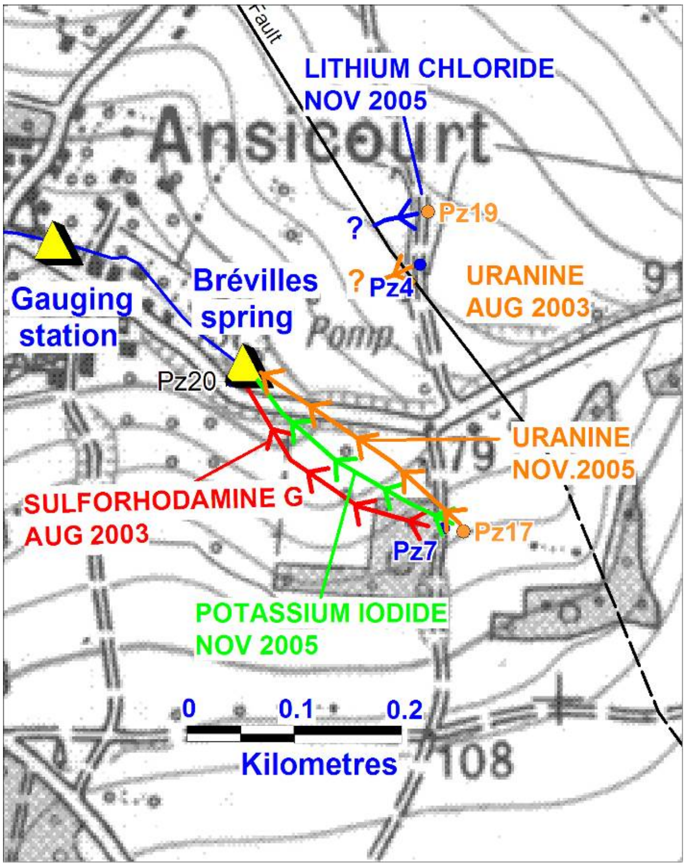


Figure 4 : Summary of tracer tests carried out at Brévilles

Each injection has been dimensioned and monitored according to the FVPDM method concepts (Brouyère *et al.*, 2008). This method, briefly described below, enables to better control the entrance of the tracer in the aquifer and to achieve accurate estimations of local Darcy's fluxes around the injection well.

Besides this first objective, the total quantity of injected tracer has been dimensioned with the goal of obtaining a detectable response at the aquifer outlet. The quantities and tracers used, as well as the distances from the injection wells to the spring, are summarized in Table 2. In order to best monitor the tracer concentration evolutions, automatic samplers were installed at two locations: at the Brévilles spring and at the level of the gauging station, 200 m downstream the spring. As mentioned above, the gauging station can be considered as a more global outlet of the aquifer, while the Brévilles spring drains only the upper part of the sandy aquifer. Sampling at the gauging station therefore increased the probability of recovering the injected tracers. Sampling simultaneously at both locations enables to detect and dissociate tracer arrivals from the different aquifer levels. Water from the spring would contain tracer flowing through the upper part of the aquifer (medium sands), and water collected at the gauging station would contain tracer flowing through both upper (coming from the spring) and the lower part of the aquifer (fine sands).

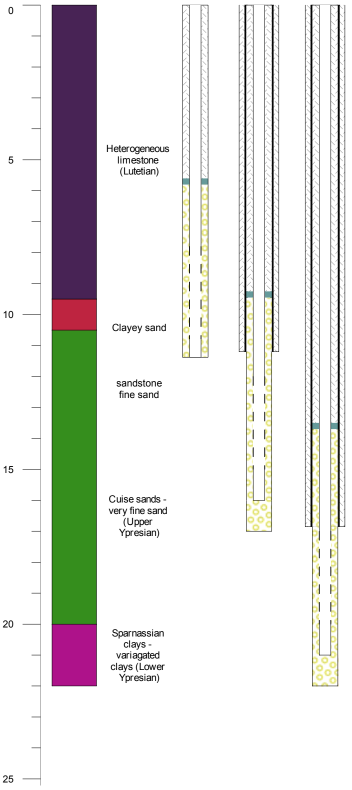
	PZ4	**PZ19**	**PZ17b**	**PZ17c**
Distance to the spring	187 m	223 m	245 m	245 m
Tracer	Lithium Li+	Sulforhodamine B	Iodide I-	Uranine
Quantity	6.6 kg Li^+	10 kg	19.2 kg	5 kg

Table 2 : Main characteristics of the tracer experiments (2nd campaign 2005)

The piezometers of group PZ17 are located close to the older piezometer PZ7. It is composed of three piezometric boreholes that have a distance of about 2 m from each other. The three boreholes are screened at 3 different levels of the aquifer (Figure 5), respectively in the Lutetian limestone (PZ17a) and in the upper (PZ17b) and lower part (PZ17c) of the Cuise Sands (Gutierrez *et al.*, 2005). Tracer tests were carried out in PZ17b (iodide I^-) and PZ17c (uranine) to highlight and quantify vertical variations in aquifer properties and groundwater fluxes in the sandy layer aquifer. Pz17a was not considered for these tests because its saturated thickness is shallow and permeability of the lutetian limestone is very low due to the presence of marls and clays. The use of two different tracers injected at two different depths targeted comparison of transport behaviour in the lower and upper parts of the formation and identification of possible vertical interactions.

PZ4 was used during a previous campaign for tracer experiments with uranine that was never detected at the spring. The borehole is screened in the lower part of the aquifer, between 17.85 m and 26.70 m, where the flow is assumed to be slower and where anoxic conditions prevail. For the new experiment, tracer quantities were increased in to ensure better chances of detecting potential tracer arrival at the spring and at the gauging station.

The tracer test performed in PZ19 with sulforhodamine B was dimensioned to give new information on the area located at the north-east of the spring. The borehole, screened between 18 and 30 m, interacts with almost the whole thickness of the aquifer.



National number: 0125-7X-0017
Designation : PZ17a-b-c (from left to right)

Figure 5 : Nested well Pz17

Injection operations and results

Methodology

For each experiment, injection operations were performed using the « Finite Volume Point Dilution Method" (Brouyère *et al.* 2008), that generalizes the "Single-well Point Dilution Method" to the case of finite volumes of tracer fluid and water flush. Thanks to an analytical solution, the FVPDM enables to model the tracer concentration evolution in a well during and after a tracer injection, and to obtain accurate values of Darcy fluxes near the injection well. The FVPDM is also used to simulate the real tracer input into the aquifer, according to the tracer liquid concentration, the tracer injection flow rate and the water transitional flow rate across the well screen in natural conditions. The analytical solution is based on water and tracer balances, and on equations of radial and tangential Darcy fluxes near a well in function of injection flow rates (Bidaux and Tsang, 1991). The analytical solution is expressed as follows :

$$C_w(t) = \frac{Q_{in}C_{in} - \left(Q_{in}C_{in} - Q_{out}C_{w,0}\right)\exp\left(-\frac{Q_{out}}{V_w}(t - t_0)\right)}{Q_{out}} \tag{1}$$

$$Q_{out} = Q_{in} + Q_t^{in} \tag{2}$$

$$Q_{out} = 2 r_w e_{scr} v_{ap} \sin\left(\arccos Q_{in}^{*}\right) + \frac{Q_{in}}{2\pi}\left(2\pi - 2\arccos Q_{in}^{*}\right) \tag{3}$$

C_w = variable accounting for the tracer concentration in the injection well [M L^{-3}]
$C_{w,0}$ = initial concentration of tracer in the injection well [M L^{-3}]
C_{in} = variable accounting for the concentration in the injected fluid [M L^{-3}]
e_{scr} = length of the well screens [L]
Q_{in} = variable accounting for the injection flow rate [L^3 T^{-1}]
Q_{out} = flow rate leaving the well through the screens [L^3 T^{-1}]
Q_t^{in} = variable accounting for the transit flow rate intercepted by the well screens as a function of the injection rate Q_{in} [L^3 T^{-1}]
Q_{cr} = critical injection flow rate [L^3 T^{-1}]
r_w = radius of the injection well [L]
v_D = Darcy flux in the aquifer [L T^{-1}]
V_w = volume of water in the injection well [L^3]

During the experiment, the tracer solution is injected in the well at a low flow rate (on the order of a few litres per hour). A recirculation system, with a much higher flow rate ensures the homogeneity of the tracer concentration in the well and enables consistent sampling operations. The schematic experimental device is shown on Figure 6. During the injection, the tracer concentration increases until reaching an equilibrium state, which depends on the concentration of the tracer liquid, the tracer injection flow rate and the water transit flow rate across the well screen in natural conditions (Eq. 1 to 3). Sampling operations enable to monitor this evolution and an analytical solution can be adjusted on experimental data, by varying natural Darcy flux values only. This can be achieved when all other parameter are known or dimensioned before the experiment. More details about the method can be found in Brouyère *et al.* (2008).

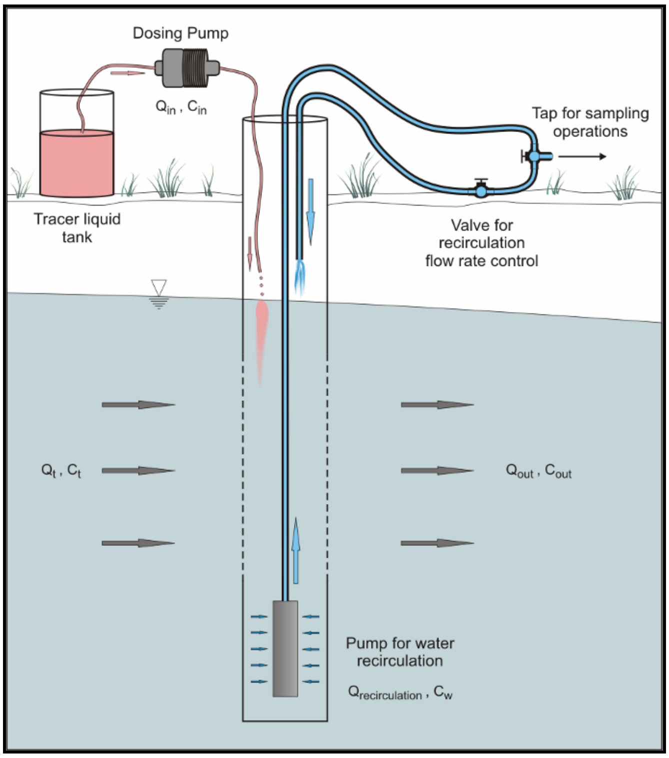


Figure 6 : Schematic experimental device to apply FVPDM

Description of the injections

For each experiment, consecutive steps of constant injection rates were performed. During each injection, samples were collected in the injection well at an approximate time intervall of 5 minutes. Generally, 2 to 4 injection steps were performed, after which, the remaining quantity of tracer was injected as a pulse injection to finalize the tracer injection in a reasonable time. Because of field conditions (electrical power supplied by a generator, no protection of the equipment against vandalism…), injections beyond a few hours were not carried out.

The information relative to the injections performed in the 4 wells is summarized in Table 7. PZ17b and PZ17c are respectively screened in the upper part (medium sands) and lower part (fine sands) of the aquifer. PZ4 is also screened in the fine sands. PZ19 intercepts the whole thickness of the aquifer. Figure 3 shows the injection steps together with the concentration evolution in each injection wells.

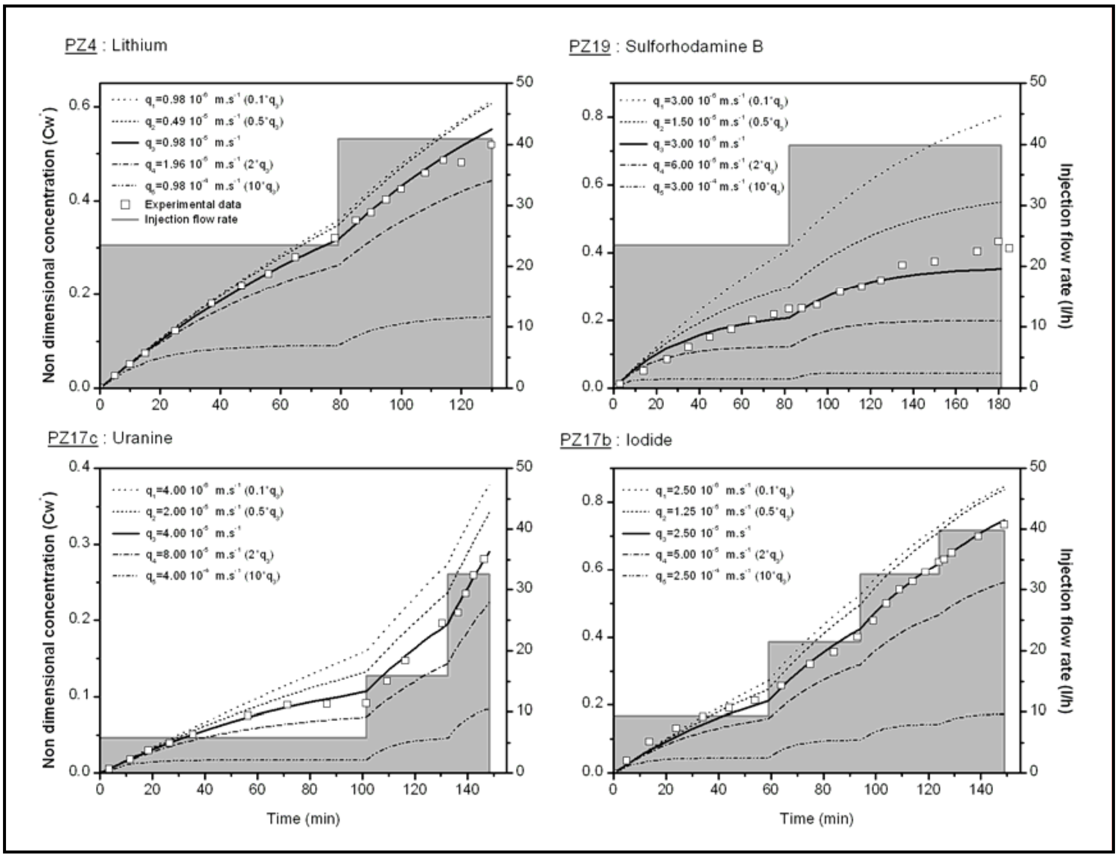


Figure 7 : Injection flow rates, monitored and modelled tracer concentrations in the injection wells

FVPDM method : Interpretation of the results

Using FVPDM concepts and equations, concentration curves were adjusted on experimental monitored concentrations by varying the magnitude of the Darcy flux only. The other terms appearing in Equation 1 and 2 are defined based on the experimental conditions (Q_{inj}, C_{inj}, V_w...).

Figure 4 allows comparisons of monitored concentrations in the injection wells and adjusted concentrations by trial-and-error. In each diagram, the thick line corresponds to the best adjustment of Darcy flux (Vd=q3). The other curves were calculated for Vd equal to 10*q3, 2*q3, 0.5*q3 and 0.1*q3, to test the sensitivity of the method. Figure 7 shows that the calculated curves almost perfectly match experimental data. Small deviations can however be observed for PZ19 and PZ17c. This is possibly due to slightly less controlled injection conditions. For these experiments, the tracer liquid concentration was very close to the solubility limit and tracer sedimentation was observed at the bottom of the mixing vessel. This suggests that, during the low flow rate injection steps, the tracer concentration in the injected fluid was lower than assumed. New adjustments were performed considering a lower concentration in the injected fluid and, as expected, this lead to lower values of Darcy flux.

All results are presented in Table 7. They are in good agreement with the *a priori* estimations of Darcy fluxes obtained using the results of pumping test and the application of Darcy's law between the injection point and the spring. This confirms that the FVPDM method is a valid technique for providing point quantification of Darcy fluxes in selected piezometers.

In PZ17c, the Darcy flux calculated based on the FVPDM interpretation was greater than the value obtained using Darcy's law. It was determined larger than the Darcy flux calculated for the upper part of the Cuise sands where the aquifer material is coarser. This might seem contradictory to the results of pumping tests performed in 2005, which indicated a diminution of hydraulic conductivity with depth. However, the deviation could be explained by local effects (flow distortion, local steeper gradients etc.). It should also be pointed out that the FVPDM provides a local (point) estimation of Darcy flux, while the application of Darcy's law provides a mean estimated Darcy flux that integrates a larger volume of aquifer (corresponding to the distance over which the piezometric gradient is calculated) and a mean hydraulic conductivity.

	Estimated flux through the screen (Darcy's law between PZ and the spring)	Estimated flux through the screen (FVPDM method)	Estimated critical injection rate (FVPDM method)
PZ4	1.13×10^{-5} m/s	9.80×10^{-6} m/s	2.18×10^{-5} m^3/s (78.43 l/h)
PZ19	1.53×10^{-5} m/s	3.00×10^{-5} m/s	9.04×10^{-5} m^3/s (325.56 l/h)
PZ17b	1.97×10^{-5} m/s	2.50×10^{-5} m/s	1.82×10^{-5} m^3/s (65.56 l/h)
PZ17c	0.62×10^{-5} m/s	4.00×10^{-5} m/s	2.91×10^{-5} m^3/s (104.90 l/h)

Table 3 : Results obtained using the FVPDM method

When applying the FVPDM method, reaching a stabilized concentration in the injection well allows *a priori* an easier and more reliable estimation of Darcy fluxes prevailing in the aquifer close to the injection well, since the ratio C_w/C_{inj} is directly proportional to the relative importance of Q_{inj} and Q_t (Brouyère, 2001). In the ascending part, the theoretical curves are closer to each other and it could be more difficult to evaluate the quality of two different configurations.

The experimental conditions prevailing during the injections did not allow sufficient time in the field for each injection to reach equilibrium. However, the results obtained in Brévilles indicate that the FVPDM method seems to be sufficiently sensible to obtain satisfying results, even in less controlled conditions (see sensitivity in Figure 7).

One of the essential conditions for being able to calculate Darcy fluxes with the FVPDM is to inject the tracers at a rate lower than the critical injection rate. For the injections performed in the Brévilles Catchment, the injection rates have never been larger than the critical injection rate. The methodology seems thus reliable if a priori estimates of Darcy fluxes are available.

Most often, tracer injection is performed considering a "classical" source term. As shown by Brouyère et al. (2005), this may lead to erroneous interpretation of the tracer test results if the injection has some influence on the shape of the breakthrough curve (because of temporary tracer capturing in the well bore). The FVPDM has the further advantage to reconstitute a "good estimate" of the tracer input function in the aquifer, in order to verify *a posteriori* that injection conditions did not influence the results, particularly when the volume of injected tracer fluid is comparable to the volume of water in the injection well.

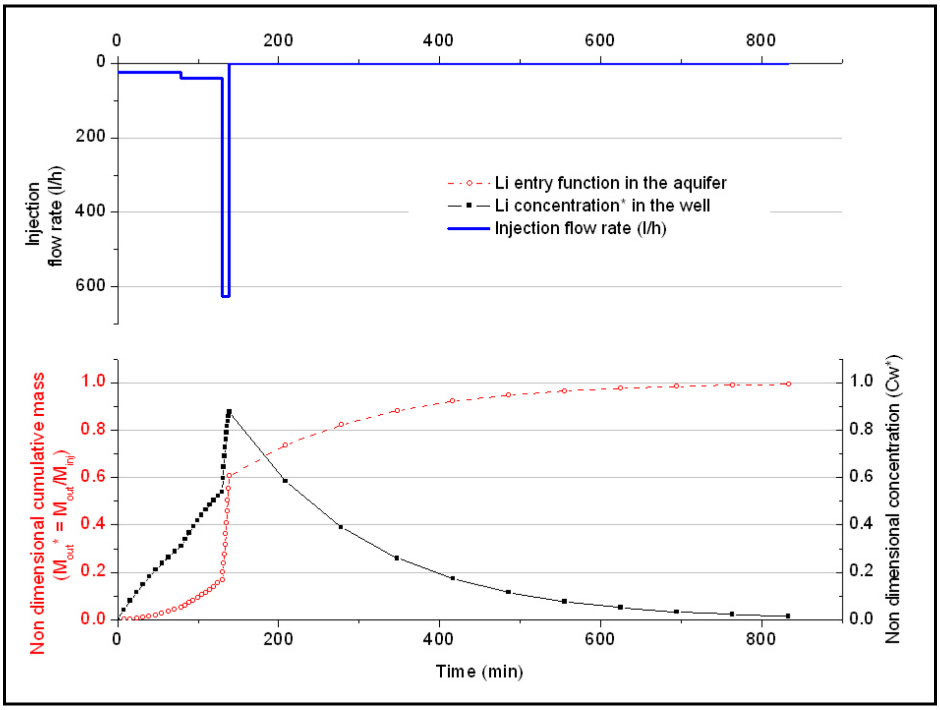


Figure 8 : Tracer input function in the aquifer (PZ4)

Figure 8 presents the theoretical input function of cumulative tracer mass in the aquifer (PZ4 injection), calculated using the well-aquifer interaction parameter (transit flow rate) obtained using the FVPDM calibration. The cumulative input function is calculated from the beginning of injection operations until more than 600 min (10 hours) after the end of these operations. The two low flow rate injection steps performed for the FVPDM analysis were followed by a Dirac type injection (more than 600 l/h) of the remaining quantity of tracer.

The curve of cumulative mass of tracer in the aquifer shows that 4 hours after the end of injection, 90 % of the tracer mass injected had entered the aquifer. The entry functions of the three other tracer tests are similar. Compared to the transit times (several days) of the tracer from the injection points to the Brévilles spring, this is very short and the injections can thus be considered as impulses. Seventeen months after injection, samples collected in the well Pz17c still showed presence of 1 g/l of Uranine, probably stuck in the silting tube, at the bottom of the piezometer. This small quantity confirmed that more than 99% of the tracer had left the piezometer. Therefore, tracer trapping in the well cannot be invoked here as a possible reason to explain the non-detection of tracers at the spring or atypical shapes of breakthrough curves.

Recovery operations and results

Besides the injection monitoring step, the objective of the tracer campaign was the recovery monitoring at the outlet of the aquifer. This monitoring gives new information, complementary to the results given by the application of FVPDM method during the injections. While FVPDM allows local scale interpretations, analysis of the breakthrough curves provides information at a larger scale about transport behaviour between the injection point and the outlet of the aquifer.

In order to increase the chances of recovery and to try to dissociate arrivals from lower and upper parts of the aquifer, samples were collected at the Brévilles spring and at the level of the gauging station (cf. figure 1). Sampling operations were conducted, using automatic samplers, for more than 2 years at both locations, with a time step gradually increased from 6 hours just after the injections to 10 days (after 2 years of sampling) until the end of the sampling campaign.

Description of the breakthrough curves

Lithium Li+ (PZ4) and sulforhodamine B (PZ19) have not been clearly detected neither at the spring nor at the gauging station. Iodide I- and uranine appeared at the spring respectively 4 and 16 days after injection. Their concentration breakthrough curves at both sampling places are presented in Figure 9 . In order to facilitate comparisons, concentrations are plotted after subtraction of the background concentrations and normalized according to the injected mass of tracer. The main characteristics of the breakthrough curves are summarized in Table 4.

		Uranine		iodide	
		spring	*gauging station*	*spring*	*gauging station*
First arrival	*Time*	16 days		4 days	
	Calculated speed	1.8×10^{-4} m/s (14.3 m/d)		7.1×10^{-4} m/s (57 m/d)	
Peak 1	*Modal time*	35 days		25 days	
	*Modal concentration**	9.16×10^{-2} ppb	1.78×10^{-2} ppb	4.3×10^{-3} ppm	1.2×10^{-3} ppm
	Calculated speed	7.5×10^{-5} m/s (6.5 m/d)		1.1×10^{-4} m/s (9.1 m/d)	
Peak 2	*Modal time*	164 days	131 days	271 days	
	*Modal concentration**	0.44 ppb	0.16 ppb	4.6×10^{-3} ppm	
	Calculated speed	1.4 m/d	3 m/d	0.8 m/d	
Peak 3	*Modal time*	≈ 749 days			
	*Modal concentration**	2.4 ppb	0.56 ppb		
Recovery factor after 800 days		32 %	42 %	55 %	
Background value		0.027 ppb	0.092 ppb	0.008 ppm	0.008 ppm

Table 4 : Main characteristics of uranine and iodide breakthrough curves (tracer tests Novembre 2005)

* : Modal concentration per kilogram of tracer injected

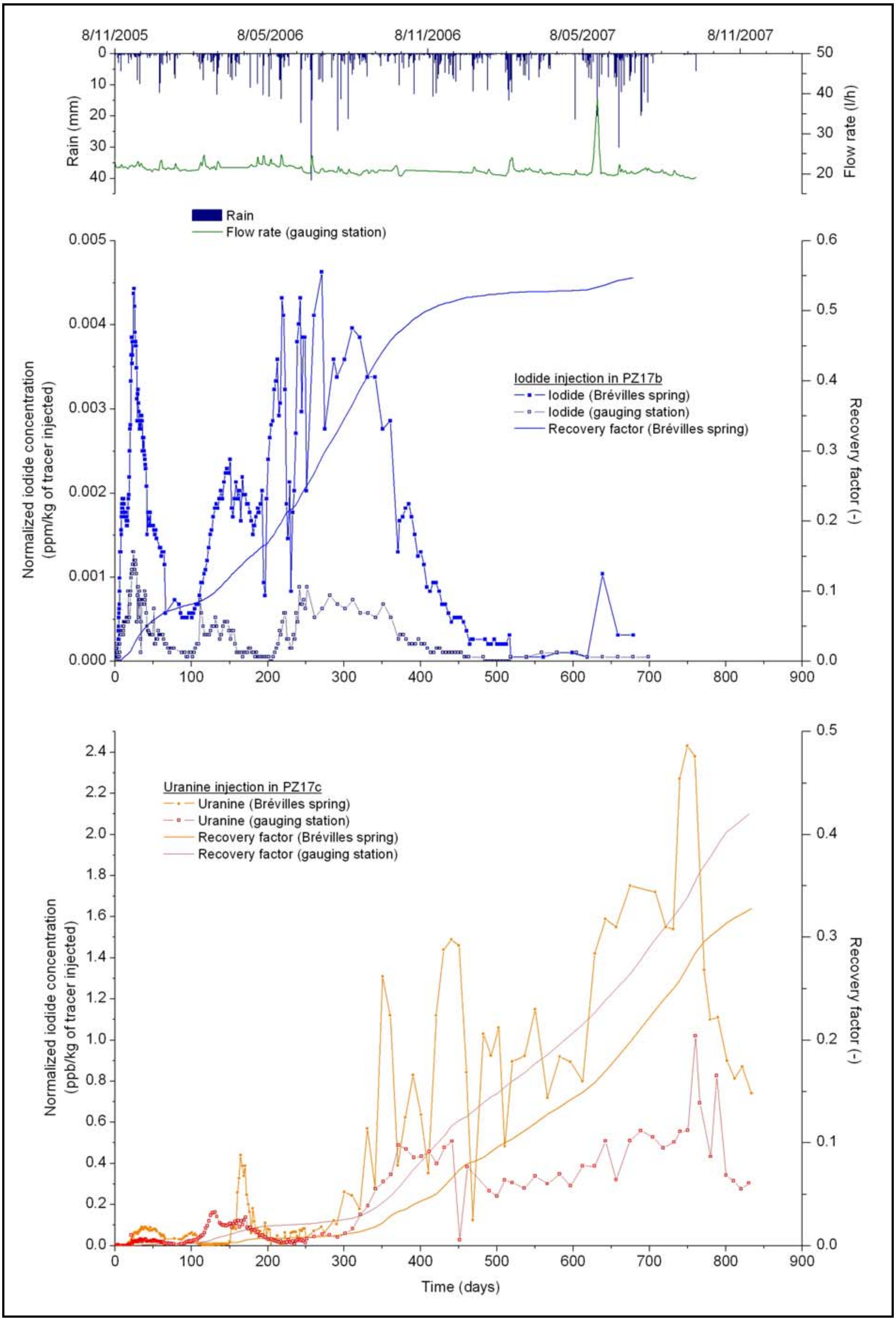


Figure 9 : Normalized concentration (by quantity of tracer injected) breakthrough curves of iodide I- and uranine at the spring and at gauging station.

Recovery operations : Interpretation of the results

The breakthrough curves presented in Figure 9 show very irregular shapes with successive peaks of concentration, far from "traditional" Gaussian curves. It is possible that precipitations had intermittent dilution impacts on concentration values. This could be particularly the case at the gauging station, which also receives run off water from slopes which would significantly dilute the sample. However, rainfall events and variations in flow rates cannot exclusively explain the general shape of the breakthrough curves.

Such irregularities in the tracer recoveries are probably essentially related to the complexity of groundwater flow and transport mechanisms in the aquifer. As this complexity is still difficult to capture, the following interpretation is conceptual, by formulating several hypotheses to explain flow mechanisms that may occur between the injection wells and Brévilles spring. Interpretations are completed by first attempts of adjustment of analytical solutions on each identified peak separately. The objective is not to obtain accurate values of transport parameters, what is not possible with a simplistic analytical approach in the complex Brévilles environment, but to help identifying the successive peaks and differentiate them with basic characteristics. In order to realistically estimate the transport parameters, a comprehensive 3D groundwater model should be set up and calibrated with the flow and transport data available for the Brévilles catchment. The new tracer tests performed at Brévilles provide precious and exceptional data for the development of such models. This will be concerned by future work.

PZ17b tracer test (Iodide)

For iodide I^-, the calculated recovery factor is equal to 55 % at the spring after 680 days. To do this calculation, the water flow rate at the spring (no continuous monitoring) has been reconstituted from the continuous monitoring at the gauging station, 200 m downstream. The ratio between the spring and the gauging station flow rates was assumed to be 3.5, according to instant discharge measurements performed by BRGM in 2001. After 309 days, the recovery factor was equal to 28 %, which is above the recovery factor of the 2003 tracer test (23%) from PZ7 (with sulforhodamine G), after the same time interval. At the gauging station, measured concentrations are very close to the background concentration. The iodide concentration peaks can still be detected at the gauging station, but the ratio between the measured concentration and background concentration is too low to calculate any recovery factor with these data. No significant peak, different from those observed at the spring, can be identified at the gauging station. Therefore, it can be assumed that iodide was released exclusively at the spring and its concentration was less important at the gauging station. This is confirmed by dilution related to the augmentation of water flow rate downstream from the spring. The percentage of tracer recovery associated with each identified peak is shown at Figure 10.

The complex tracer restitution observed cannot be explained by simple advective and dispersive mechanisms. This suggests that Darcy fluxes are non-homogeneous within the aquifer. The bimodal shape of Iodide restitution at the spring suggests two different tracer velocities, which can give way to two different simple advection/dispersion interpretations.

Using the software CATTI (Sauty *et al.* 1992), two theoretical curves have been adjusted on the concentration peaks of the iodide breakthrough curve obtained at the Brévilles spring (Figure 10). A one-dimensional solution has been used with the effective porosity n_e being considered as an adjustable parameter.

This model also requires an estimation of the mean Darcy flux. 'Calibration 1' uses fluxes calculated with Darcy's law applied using pumping test results and the hydraulic gradient estimated between PZ17b and the spring. Calibration attempts for the first peak provide estimates of the effective porosity n_e at the order of 24 %, value probably near the upper limit for this parameter considering the type of geological formation. The only way to adjust a curve for the second peak was to significantly decrease the Darcy flux value by a factor 10. As a matter of fact, the second peak, which induces a slower velocity of the tracer would correspond either to a much higher effective porosity (which is not consistent) or a lower Darcy flux. Therefore, 'Calibration 2' uses the same parameter value for effective porosity as 'Calibration 1' and has been adjusted by varying the value of Darcy flux. However effective porosity and Darcy fluxes are linked and different couples of values could be proposed leading to the same result. Nonetheless, the range of effective porosity is much more limited than the possible variation of Darcy fluxes. In such aquifers, effective porosity could be overestimated by a factor 2, whereas Darcy fluxes may vary by a factor 100. Keeping a constant effective porosity, simplifies the reasoning without impacting the conclusions. All results are presented in Table 5.

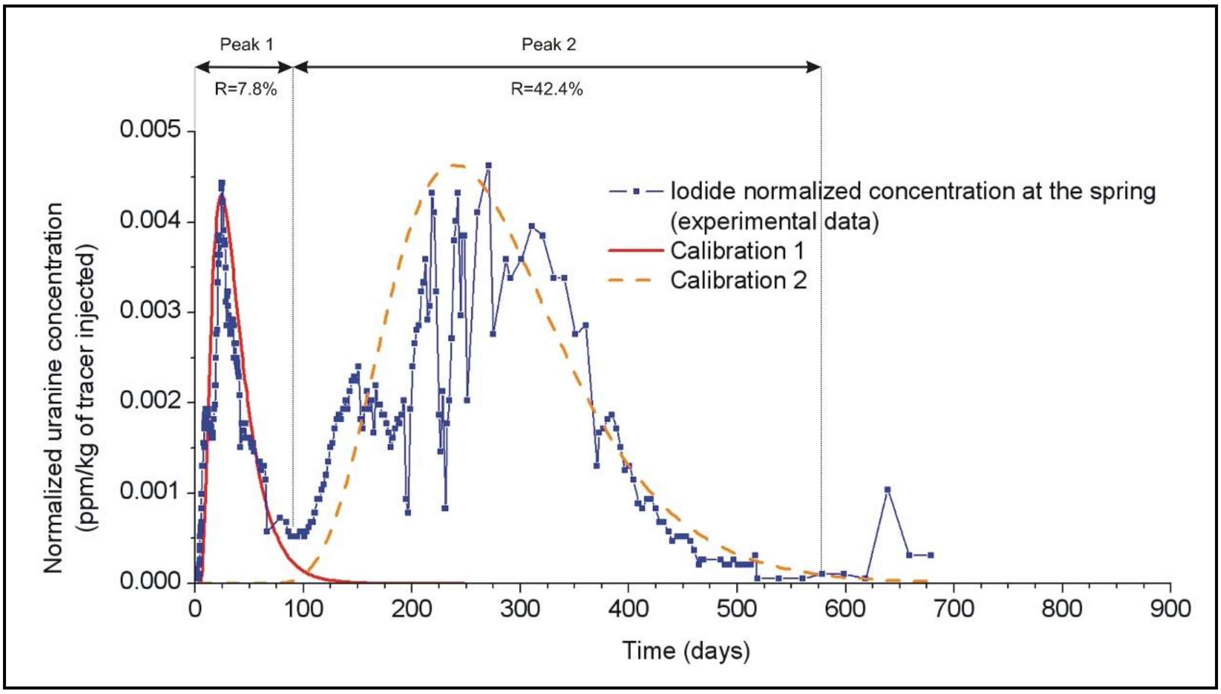


Figure 10: Adjustment of theoretical curves using "CATTI" (Iodide breakthrough curve at the spring)

(R= Recovery factor for each peak)

Breakthrough curve	'Calibration 1' (spring)	'Calibration 2 (spring)
Darcy flux	2.0×10^{-5} m/s (from pumping test and Darcy's law – Table 3)	2.7×10^{-6} m/s

Effective porosity	0.24	0.24
Dispersion	~ 20 m	~ 13 m

Table 5 : Adjustment of theoretical curves on the iodide breakthrough curve - Adjusted parameters

The bimodal peaks clearly indicate two different velocities. The second peak has the same magnitude as the first one but occurs 246 days later. The tracer being of anionic type, "classical" sorption processes are difficult to invoke for explaining such a strong retardation. It is more likely that a non-negligible part of the iodide tracer has travelled along less pervious pathways or that it has been retarded by "large-scale" dual-porosity effects through temporary capturing in less pervious horizons. The shape and nature of the geological layers might provide an important clue. At Pz17b, the Lutetian limestone is marly and probably of very low permeability. Core drilling and observation of drawdown at Pz17a have evidenced this fact and refuted the hypothesis of a fast transfer in a fractured Lutetian limestone. However, near the top of the sandy layer, beds of hard glauconitic sandstones, alternatively with glauconitic sand are observed. They are also present near the spring. The glauconitic sand is coarser than the fine sand constituting the main formation. The sandstone might be fractured and we assume that this thin layer (probably about 1 m in total) has a significantly higher permeability than the rest of the aquifer. This permeable layer, which can be estimated at a depth between 11-12 m is not directly in front of the well screen (located between 12 and 15 m) but is connected to it through the gravel pack. Therefore, it is conceivable that once injected in Pz17b the tracer took two different pathways: the upper pathway where permeability is high, and the middle pathway corresponding to the middle part of the aquifer with a permeability ten times lower.

PZ17c tracer test (uranine)

The recovery factor for uranine is equal to 42 % after 830 days, using the concentration and flow rate values measured at the gauging station. The recovery factor at the level of the spring is estimated to 32 % (Figure 9). Figure 11 shows the tracer recovery for each identified peak at the spring and the gauging station.

The 10 % difference in recovery factor between the two sampling locations suggests that a part of the tracer reached the gauging station without being observed at the spring. The graphic analysis of the uranine breakthrough curves (Figure 9 and Figure 11) confirms this hypothesis. From 0 to 100 days after injection, the two concentrations are strongly correlated, with an almost constant concentration ratio equal to 3.5, identical to the discharge ratio between the two points. This indicates that, during the first 100 days, the uranine tracer which is observed at the two sampling locations reaches the spring first (and only the spring) and follows the Brévilles brook to reach the gauging station. During that period, the uranine is observed at the downstream sampling location at a lower concentration because of dilution related to the increase of water flow rate between the spring and the gauging station.

After 100 days however, the concentration at the spring falls abruptly to almost zero while the uranine concentration starts to increase at the gauging station, the concentration ratio between the two curves consequently decreases. This indicates a single arrival between the spring and the gauging station. After 150 days, a second peak is observed at the spring. The ratio between concentrations then becomes much more variable and is probably a

consequence of simultaneous arrivals at the spring and along the brook between the spring and the gauging station. After 300 days, a third more important peak is detected at the spring and the gauging station. The difference between the calculated recovery factors at both locations continues to increase with time, still indicating diffuse arrivals between the spring and the gauging station. The third peak is characterized by a highly pronounced variability of the uranine concentration, especially at the spring.
Some more intensive sampling operations, not plotted on the graph of Figure 6, have shown that this variability was visible at least at the scale of daily time intervals. Therefore, the many irregularities of the third peak should not be compared with the second peak of the spring breakthrough curve, the shape of which is similar. The second peak corresponds to a distinct tracer recovery and is not due to the variability of the concentration.

Because they are dependent on the water flow rates, the breakthrough curves presented in Figure 11 are not very representative of the tracer quantities that really arrived at the spring and along the brook. In order to have a more explicit view and to make the analysis easier, the breakthrough curves were plotted using tracer mass flux units (kg of tracer s^{-1}). The quantity of uranine "already" detected at the spring has been subtracted from the quantity of uranine detected at the gauging station so as to obtain a breakthrough curve that only corresponds to tracer arrivals between the spring and the gauging station. The resulting breakthrough curves for the period from the injection to 250 days are presented in Figure 12, where the relative importance of each identified peak can be much more easily visualized and understood. For the period after 300 days, the high variability of the uranine concentrations makes this graphical analysis less meaningful and has not been performed.

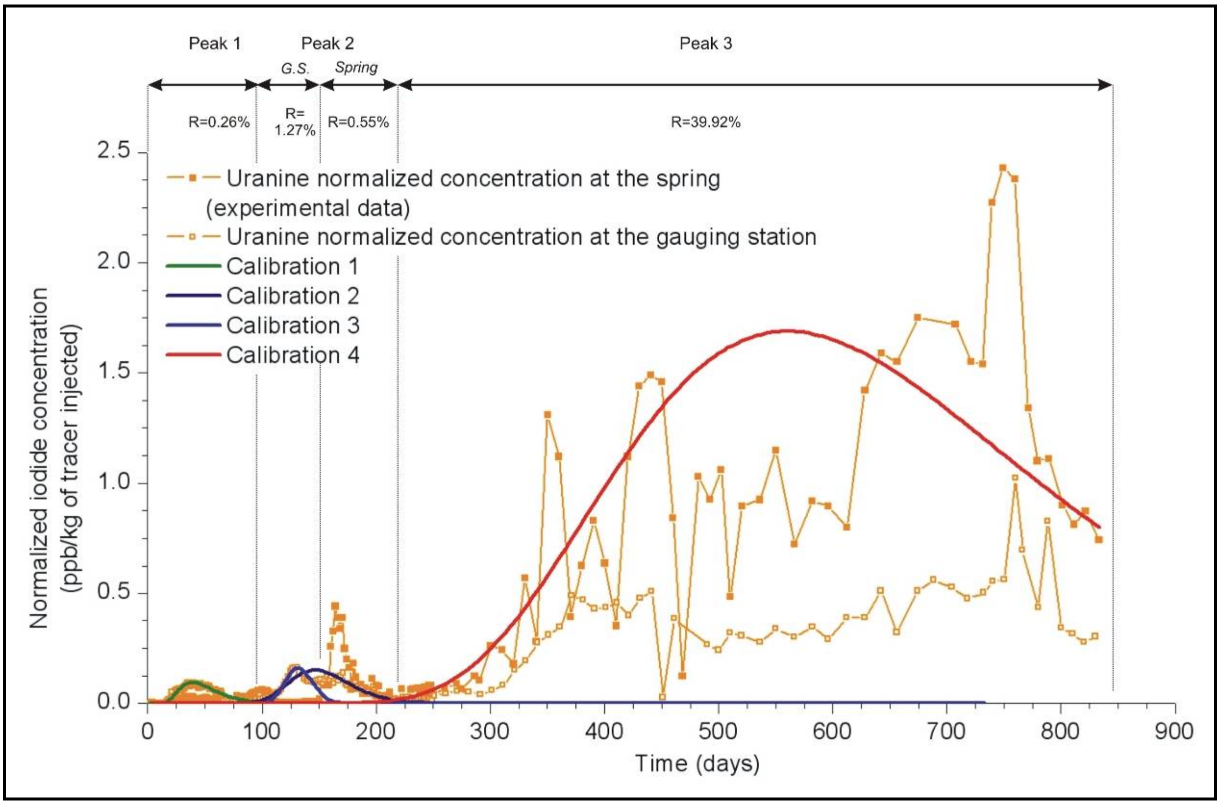


Figure 11 : Adjustment of theoretical curves on the uranine breakthrough curves at the spring and at the gauging station, using CATTI

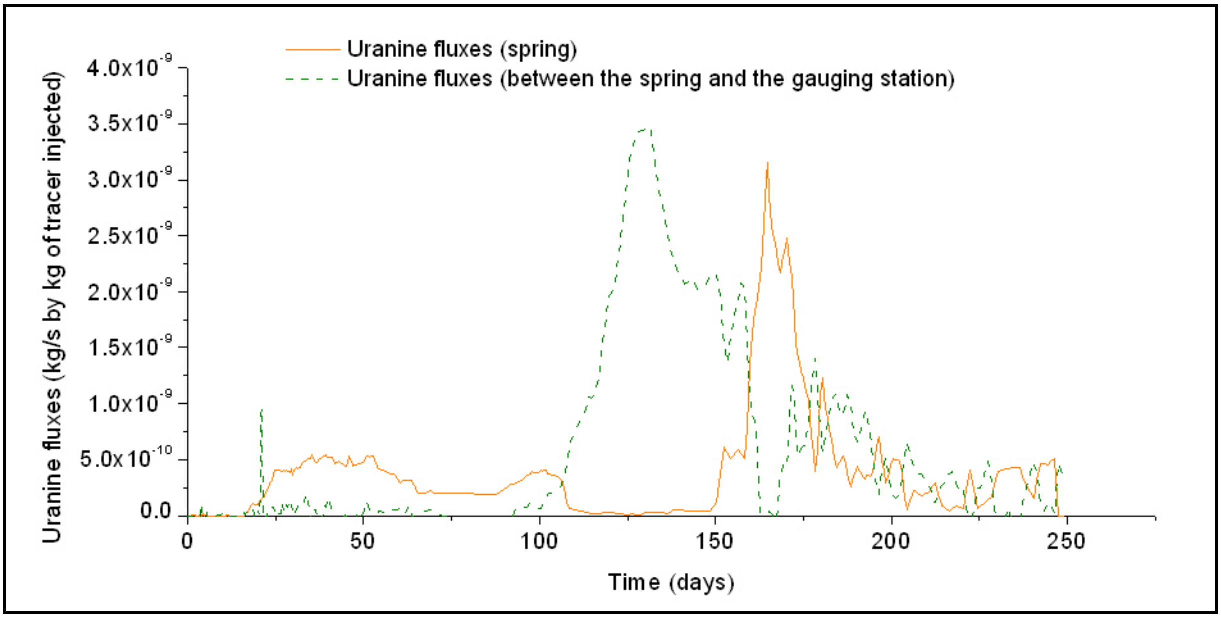


Figure 12 : Uranine mass flux (kg/s) at the spring and the gauging station

Breakthrough curve	'Calibration 1' (spring)	'Calibration 2' (gauging station)	'Calibration 3' (gauging station)	'Calibration 4' (spring)
Darcy flux	0.62×10^{-5} m/s (from pumping test and Darcy's law)	0.62×10^{-5} m/s	0.62×10^{-5} m/s	4.80×10^{-7} m/s
Effective porosity	0.10	0.15	0.18	0.10
Dispersion	20 m	2 m	8 m	13 m

Table 6 : Parameters adjusted on the uranine breakthrough curves

As for the tracer test performed in PZ17b, theoretical curves were adjusted on the concentration peaks at the spring and at the gauging station. This was carried out using a Darcy flux calculated with Darcy's law (0.62×10^{-5}m/s). Results are presented in Figure 11 and Table 6. The adjustment of a theoretical curve to the peak occurring at 175 days on the spring breakthrough curve only could not be performed with acceptable parameters. The theoretical curve 'Calibration 3' has been adjusted considering that the second peak at the spring and the second peak at the gauging station were constituents of a same tracer arrival. As for the Iodide breakthrough curves, the adjustment of a theoretical curves on the last (incomplete) peak, keeping the same value of Darcy fluxes, turned out impossible with realistic values of the effective porosity. The curve 'Calibration 4' has therefore been adjusted by varying the magnitude of Darcy's fluxes and dispersion. The resulting values of effective porosity range from 10% to 18%, which is less than for the tracer experiment performed in PZ17b. This is in accordance with the idea of a grain size distribution decreasing with depth.

The first peak observed at the spring is approximately five times less prominent compared to the peak observed at the gauging station, considering the actual quantities of tracer recovered (Table 4). The hypothesis of a tracer transfer through a hypothetic connection between PZ17c and PZ17b would involve a strong retardation factor for uranine; otherwise this tracer would have appeared simultaneously with iodide. Note that iodide reached the spring 4 times faster than uranine. Although uranine is known to be more sensitive to sorption than saline tracers (Smart and Laidlaw, 1977; Kasnavia et al, 1999), its properties under a pH value around 7.2 should not result in such a ratio. The most probable hypothesis is the occurrence of vertical interactions within the aquifer, with a small part of the tracer reaching more permeable levels. This would simultaneously explain the time transfer between the well and the spring (faster than the first uranine detection time at the gauging station but slower than the first iodide detection at the spring) and the more important dispersion that characterised the first uranine peak observed at the spring.

The second peak observed between 100 and 150 days at the gauging station and not at the spring is particularly enigmatic because it seems that a sudden change in the pathways occurred during within a matter of few weeks . Considering the short delay between the decrease of concentration at the spring and the increase at the gauging station, the tracer has most likely encountered the brook in the vicinity of the spring, with the rest of the transport being extremely fast in surface water. As this fast disappearance of the tracer would occur at almost any runoff scenario, no clear correlation with discharge nor with rainfall could be established.

The sharp concentration peak detected at the spring after more than 150 days could be compared to the second iodide peak. Its occurrence is probably due to a similar pathway, occurring in the middle part of the aquifer.

If we consider that most of the flow is horizontal flow, then most of the tracer mass injected in the lower part of the aquifer will remain at this level and will progress slowly to the spring. The spring is located in the upper middle part of the aquifer (Figure 2). In a strictly horizontal flow uranine should not have appeared at the spring. However, geochemical characteristics and isotopic measurements on spring water proved that the Brévilles spring is a mixture of water from different origins (Baran et al., 2007; Brenot et al., 2008) and uranine did appear significantly at the spring. A vertical component to the groundwater flow is therefore necessary to explain this mixing.

The cumulative mass curve, plotted for the two tracers at the spring (Figure 13) clearly evidences the velocity difference between the lower part, traced with uranine, and the middle and upper part traced with iodide. While iodide appears quickly with two distinct arrivals, cumulative mass curve of uranine indicates that there is no significant tracer restitution before 300 days. Therefore, the significance of the first two peaks observed at the spring and at the gauging station should be nuanced. The tracer concentration representative of groundwater flow in the lower part of the aquifer seems to decrease after 750 days, giving a mean convection velocity of 0.3 m/day and a corresponding Darcy flux of $3.2.10^{-7}$ m/s considering a 10% effective porosity (calibration 4 considers a higher Darcy flux corresponding to $4.8.10^{-7}$ m/s). These values are about 100 times lower than the Darcy Flux estimated by the FVPDM method, which gives a very local value, probably influenced by the local well conditions (higher permeability due to the well completion). However, the estimated Darcy flux from the tracer peak might be underestimated because of the difficulty to establish a peak among the perturbed signal and because of probable sorption of uranine which results in a loss of tracer (fixed in the aquifer) and a delay to reach the maximum of recovery (not yet reached).

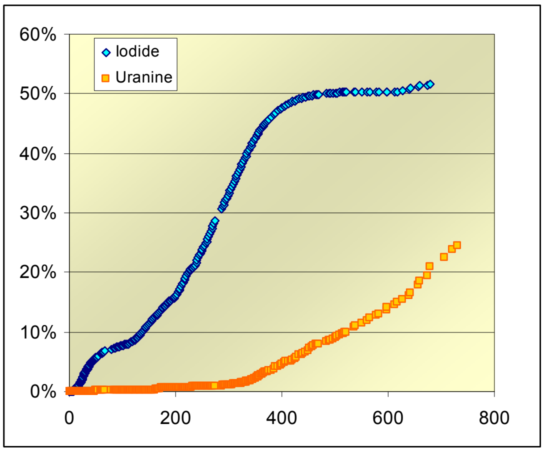


Figure 13 : Cumulative mass curves compared at the spring for both tracers Iodide and Uranine.

PZ4 (Lithium) and PZ19 (Sulforhodamine B) tracer tests

Tracers injected at PZ4 and PZ19, located at 50 m from one another and at 187 m and 223 m from the spring respectively, were never detected clearly neither at the spring nor at the gauging station. Several points should be considered to understand the reasons of this phenomenon:

1. Monitoring the concentration evolutions in the injection wells has confirmed that the tracer did not remain in the well. Other tests such as pumping tests also showed that these wells are not clogged.
2. PZ4 is screened in the deeper part of the aquifer but Pz19 is not. Therefore the explanation might not lie only in the vertical differentiation of the aquifer.
3. The question of detection limit could be considered. Would the mass of product used for this experiment (6.6 kg of Lithium and 10 kg of sulforhodamine B), be enough to appear at the spring above detection limit? A simple comparison can be made using the maximum concentration peaks observed for the tracer experiments performed in PZ17b and Pz17c. Considering the same ratio between the maximum concentration and the tracer quantity injected in PZ17c, the maximum concentration in Lithium (PZ4) would have been of 15.84 ppb, which is close to the 10 ppb detection limit. Although hydraulic conditions are probably different between PZ17c and PZ4, this simple comparison suggests that observed Lithium concentrations could be below the detection limit, which could explain the absence of recovery. On the other hand, this reasoning is not valid for uranine injected in 2003 (PZ4) and Sulforhodamine B injected in 2005 (PZ19). Even when considering the most restrictive ratio between tracer quantity and maximum concentration (associated to the peaks observed during the first year for PZ17c experiment), extrapolated maximum concentration would be 0.204 ppb for uranine (2003 – PZ4) and 4.4 ppb for Sulforhodamine B (2005 – PZ19), which is well above the 0.01 ppb detection limit for these fluorescent tracers. Therefore, for these two experiments, the quantity of tracer injected can probably not explain alone the absence of recovery.

Finally a few hypotheses remain:

1. Groundwater flow in the PZ4-PZ19 area is extremely slow and the tracers are still on their way. Sampling will go on to verify this hypothesis.

2. Adsorption of the tracers in the aquifer is possible. Finer particles of sand could also induce greater retardation effects (macroscale dual porosity effects). Batch or column experiments would verify this hypothesis.
3. The presence of a fault has been evidenced by geophysical survey in the close vicinity of the two wells. The fault could slow down groundwater flows, acting as a barrier; trap the tracers in a compartmented "dead end" zone, or divert them to a remote location.
4. The diversion of the flow towards an unmonitored outlet is less probable. If the area where Pz19 and Pz4 stand were not contributing to the spring discharge, then the spring catchment would be reduced and its shape would be awkward. Water balance and piezometry established during the AquaTerra project are not in accordance with this hypothesis (Gutierrez and Gigleux, 2005)

Isotope analysis showed that Pz4 water, which comes from the lower part of the aquifer because of the location of the well screen, had no tritium (nor nitrates or pesticides). PZ4 water is therefore older than about 40 years, which indeed is a sign of very slow progression. However Pz17c shows the same geochemical characteristics and the injected tracer did appear at the spring. The hypothesis of the trapping of the tracer is nonetheless plausible because of the presence of the fault structure, which blocks deep groundwater flow.

On the contrary, nitrates, pesticides and tritium were detected at PZ19 in the same range as Pz17b. Both show isotopic signatures similar to the spring water (Brenot et al., 2008). No conclusion can thus be made as to explain the absence of recovery of the tracer injected at PZ19. Nonetheless this absence clearly shows that this subsurface part of the catchment is far more complex than previously assumed.

General Conclusions and perspectives

Multi tracer tests using the FVPDM method has shown several advantages, and lead to formulate interesting hypothesis or conclusions.

First, the campaign has provided interesting data and new insights for the understanding of the hydro-geological functioning of the Brévilles aquifer. The results achieved are quite difficult to interpret and express the complexity of the aquifer. Based on the analysis and interpretation of the breakthrough curves obtained at the Brévilles spring and at the downstream gauging station, some general conclusions and conceptual model can be proposed for groundwater flow and transport in this kind of sandy layers.

Generally, the transfer times between the injection wells and the spring are larger for uranine, injected in the lower part of the sand layer. The quantity of uranine recovered after more than 2 years is also less important than the quantity of iodide recovered. This is in accordance with the idea of a higher hydraulic conductivity at the top of the formation and decreasing with depth. A third level, of even higher hydraulic conductivity is also suspected at the top of the sand formation where sandstone is frequent and the sand is coarser.

The hypothesis of a strict stratification of groundwater flow within the aquifer is confirmed. However, most of the uranine injected in the lower part of the aquifer is detected at the level of the Brévilles spring. This suggests the occurrence of vertical transfers between the lower and middle part of the sand aquifer, either locally favoured by heterogeneities or by transverse dispersion between these two layers.

Finally, the absence of recovery from the tracers injected in PZ4 and PZ19 is probably linked to the geological structure. The presence of a fault may induce a considerable delay in the tracer transfer or it could divert the flow direction rendering the pathway longer to the Brévilles spring.

. More detailed modelling application would enable further and more accurate interpretations.

Secondly, the campaign performed in the Brévilles test site has presented a very convenient way of making tracer tests more profitable. The long monitoring allowed to demonstrate the slow velocity of the system while a shorter one (stopped for instance after the first peak) would have led to a complete misunderstanding of this point. Monitoring both injection and recovery steps allows to be more exhaustive and to give more robust interpretations. Sampling at several locations, at the level of the aquifer outlet, enables to increase the probability of recovery and to dissociate arrivals from different parts of the aquifer. Better controlling injection allows ruling out that the tracer is not trapped in the well, and provides the actual tracer entrance function in the aquifer. Using the FVPDM method gives estimates of local Darcy fluxes around the injection well, which makes the tracer test success less dependant on an uncertain recovery at the aquifer outlet. This new method can be used and dimensioned in different ways, in combination with recovery objectives downstream the injection point, or alone without any other objective. In this last case, the needed quantity of tracer does not exceed a few grams, which makes the test reliable, cheap and easy to perform, and opens better application perspectives.

In the context of the AquaTerra project which focuses at Brévilles on the pesticide transfer, the multi-tracer test carried out shows physical evidence of the complexity of transport in the saturated zone. It shows horizontal groundwater flow velocities varying of two orders of magnitude, from more than 50 m/day to less than 0.5 m/day, within the same aquifer layer and confirms the groundwater stratification. It gives an estimation of hydrodynamic and transport parameters to be used in the modelling module ("Compute") of the project. Hopefully, the contribution of the test carried out to the understanding of pesticide transfer and their prediction will constitute a significant input.

Acknowledgment

This work was funded by the European Union (integrated project AquaTerra, 505428 (GOCE) and BRGM Research Division. Mr. B. Belot, from the Laboratory of water analysis of HGULg performed the analyses of Lithium and Iodide. Dr. P. Meus, from the SME "*European Water Tracing Services - EWTS*", performed the analyses of the fluorescent tracers of the 2005 tracing campaign; he is gratefully acknowledged for his suggestions and comments on the interpretation of the results. BRGM also thanks Michel

Lepiller (Polytech Orléans), dramatically disappeared in 2006 and Bruno Leclerc (Geo-Hyd) for their contributions to the tracer detection and their fruitful discussions.

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		Pz4			Pz19			Pz17b					Pz17c			
	Borehole depth (m)	28			28.4			16					21			
	Water column h_w (m)	14.31			9.93			5.81					11.07			
	Well radius r_w (m)	0.040			0.040			0.040					0.040			
	Well volume V_w (m^3)	0.071			0.078			0.030					0.051			
	Screen length e_{scr} (m)	8.9			11.9			2.9					2.9			
	K_{mean} (pumping test) ($m \cdot s^{-1}$)	2.75×10^{-4}			4.00×10^{-4}			8.67×10^{-4}					2.75×10^{-4}			
	Estimated v_D ($m \cdot s^{-1}$)	1.1×10^{-5}			1.5×10^{-5}			1.9×10^{-5}					0.6×10^{-5}			
	Estimated Q_{cr} ($m^3 \cdot s^{-1}$)	2.6×10^{-5} ($93.6\ l.h^{-1}$)			4.6×10^{-5} ($165.6\ l.h^{-1}$)			1.5×10^{-5} ($54.0\ l.h^{-1}$)					4.7×10^{-6} ($16.9\ l.h^{-1}$)			
	Tracer	Li^+			Sulforhodamine B			I^-					Uranine			
	Total M_{inj} (kg)	6.6			10			19.2					5			
	Total V_{inj} (m^3)	0.16			0.098			0.16					0.045			
	C_{inj} ($kg \cdot m^{-3}$)	41.3			102.0			120.0					111.1			
	Q_{rec} ($m^3 \cdot h^{-1}$)	≈ 1.0			≈ 1.0			≈ 1.0					≈ 1.0			
		1	*2*	***Total***	*1*	*2*	***Total***	*1*	*2*	*3*	*4*	***Total***	*1*	*2*	*3*	***Total***
Injection parameters	**Q_{inj} ($l \cdot h^{-1}$)**	23.5	40.9		23.5	35.3		9.4	21.4	32.6	39.9		5.8	15.9	32.6	
	Time (min)	79	51	**130**	82	99	**181**	59	35	30	25	**149**	101.5	31	16	**149**
	Volume (m^3)	0.031	0.035	**0.066**	0.032	0.066	**0.098**	0.009	0.013	0.016	0.017	**0.055**	0.010	0.008	0.009	**0.027**
	Tracer mass (kg)	1.28	1.45	**2.73**	3.26	6.73	**10.00**	1.08	1.56	1.92	2.04	**6.60**	1.11	0.88	0.99	**2.98**
Results	**Calculated v_D ($m.s^{-1}$)**	9.8×10^{-6}			1.0×10^{-5} - 3.0×10^{-5}			2.5×10^{-5}					4.0×10^{-5}			
	Calculated Q_{cr} ($m^3.s^{-1}$)	2.18×10^{-5} ($78.5\ l.h^{-1}$)			3.01×10^{-5} - 9.04×10^{-5} ($108.4 – 325.4\ l.h^{-1}$)			1.82×10^{-5} ($65.5\ l.h^{-1}$)					2.91×10^{-5} ($104.8\ l.h^{-1}$)			

Table 7 : Experimental setup data specific to injections performed on the Brévilles spring test site