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Contaminant transfer and hydrodispersive parameters in basaltic lava flows: artificial tracer test and implications for long-term management

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Abstract: The aim of this paper is to evaluate the vulnerability after point source contamination and characterize water circulations in volcanic flows located in the Argnat basin volcanic system (Chaîne des Puys, French Massif Central) using a tracer test performed by injecting a iodide solution. The analysis of breakthrough curves allowed the hydrodispersive characteristics of the massive lava flows to be determined. Large Peclet numbers indicated a dominant advective transport. The multimodal feature of breakthrough curves combined with high values of mean velocity and low longitudinal dispersion coefficients indicated that water flows in an environment analogous to a fissure system, and only slightly interacts with a low porosity matrix (n_e < 1%). Combining this information with lava flow stratigraphy provided by several drillings allowed a conceptual scheme of potential contaminant behaviour to be designed. Although lava flows are vulnerable to point source pollution due to the rapid transfer of water within fractures, the saturated scoriaceous layers located between massive rocks should suffice to strongly buffer the transit of pollution through dilution and longer transit times. This was consistent with the low recovery rate of the presented tracer test.

Keywords: Groundwater; Tracer test; Lava flow; Volcanic aquifer; Vulnerability; France

1 Introduction

Exploitation of volcanic groundwater is vital in many locations around the world [1–12] therefore management and international policy (e.g., [13]) implementation of resource and related ecosystem protection [14–17] require the assessment of volcanic aquifer vulnerability to pollution in the context of local and global anthropogenic pressures.

Such evaluation however remains challenging as volcanic aquifers may harbor a complex geometry leading to highly variable hydrodispersive characteristics. Lava flow may be formed by porous ash flow tuffs or by massive rocks. Many lava flows during their settlement develop superficial and basal brecciated (scoriaceous) layers as the lava flow advances, and tend to create permeable zones close to the edge of lava flow, whereas the central part is more massive and shows columnar jointing formed during the cooling [18, 19]. Values of primary porosity and permeability depend on the cooling rate, degassing during cooling, and on the magma viscosity. Total porosity in basalts ranges from zero to more than 75%, but efficient porosity is usually very lower [20]. Laboratory experiments showed that hydraulic conductivity K may vary in a range com-

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prised between 10^{-1} and 10^{-15} m/s with some basalts presenting 10 ranges of magnitudes in the same formation [21] with strong variability on the vertical axis due to interflow spaces and horizontal fractures [22].

Consequently, groundwater pathways, storage and discharge may be very variable, as was pointed out in numerous systems such as the Golan heights in Israel [5], India [3], Reunion Island [2, 23, 24], Mayotte Island in the Comoros archipelago [1, 12], Madeira Island [25] Galapagos Islands [8]. These systems may behave as fine grained systems with poorly variable discharge [9, 26, 27] associated with slow groundwater transit. Conversely, the presence of fissured massive lava flows which potentially favours rapid transfer of chemicals, may make these aquifers unpredictable after a pollution event, especially when occurring from point sources [28]. As a consequence, resource protection and management may be difficult and may require special measures, such as perimeter protection or resilience evaluation after a pollution event.

The use of artificial tracer tests may be an alternate method to examine heterogeneous groundwater systems. This approach is especially suited to fractured systems where groundwater may have strongly preferential pathways which can lead to vulnerability being very delineated over a watershed [20, 29–32]. Furthermore, in contrast to pumping tests or rock texture evaluations, the analysis of the resulting breakthrough curves may provide an estimation of the above-mentioned hydrodispersive parameters along specific pathways and at the extended scale of several thousands of meters [33]. It could also allow the evaluation of double porosity in fractured systems-where water and chemicals are affected byboth fracture flow and matrix diffusion-and what is required when addressing contaminant problems within heterogeneous rocks [29, 32]. The majority of the literature has consistently focused on fractured crystalline and karstic aquifers by accounting for this dual porosity model [e.g. 34–40]. Volcanic aquifers are however still poorly understood in regards to the combined application of tracer testing and breakthrough curve analyses. To our knowledge, the few papers addressing this topic [32–35, 41] (with a great part focusing only on the Yucca mountain tuffs in Nevada with the perspective of nuclear waste storage) refer mainly to studies done in plutonic or metamorphic systems which possess the double porosity or double permeability mentioned above. However, the specifics of volcanic settlements remain to be thoroughly investigated, especially when taking into account that the common double porosity model for fractured material might be confounded by the role of extensive porous scoriaceous layers that may alter the fate of any contaminants.

The approach of using tracer tests and the resulting breakthrough curves appears well suited to examining volcanic aquifer sustainability because it can illustrate both the internal geometry of the systems but also the contaminant pathways according to this geometry. This paper presents the application of this method for the Argnat basin which is used for public water supply in the Chaîne des Puys area (Massif Central, France).Since the end of the 19th century, the volcanic structures of the Chaîne des Puys have been known to form aquifers characterized by good biological quality and flow regularity [27, 42] and hence they have been used as a source of fresh water (domestic water, bottled mineral water) since the first half of the 20th century [27, 43]. Currently, more than 20 natural springs or artificial water catchments (e.g., wells, galleries) are exploited. In order to better understand the groundwater recharge and flowpaths in the Argnat basin, a large number of geological, hydrochemical and geophysical properties have been studied [9, 28, 44–53]. These studies highlighted the aquifer's structural heterogeneity, which is characterized by the association of massive fissured lava flows and packed clinker layers of basalt, trachybasalt, and andesit. Coupled with the hydrological, geological and geochemical information from these previous works, this study seeks to identify the type and degree of heterogeneity of the system and to characterize pollutants' pathways along groundwater flowpaths. The aim is to develop a conceptual scheme with a special focus on the point source contamination fates to be proposed, this will allow management priorities to be highlighted for volcanic aquifers regarding pollution.

2 Geological and hydrogeological contexts

The Argnat basin is located in the Northern part of the French Massif Central, called the Chaîne des Puys. The Chaîne des Puys consists of Upper Pleistocene and Holocene volcanoes that run from North to South over the French Massif Central. The Argnat basin has a surface area of 26,1 km^2 with an altitude ranging from 380 m.a.s.l. to 1159 m.a.s.l. (811 m.a.s.l. average) (Figure 1).

From a geological point of view, the watershed is composed of five lava flows overlying the fractured plutonic basement called "Plateau des Dômes" upstream and the "Limagne" sedimentary basin downstream. The Plateau des Dômes and the Limagne are separated by the Limagne fault [51]. Geophysical and geological investigations [28, 45, 49, 50] indicated that the lava flows origi-

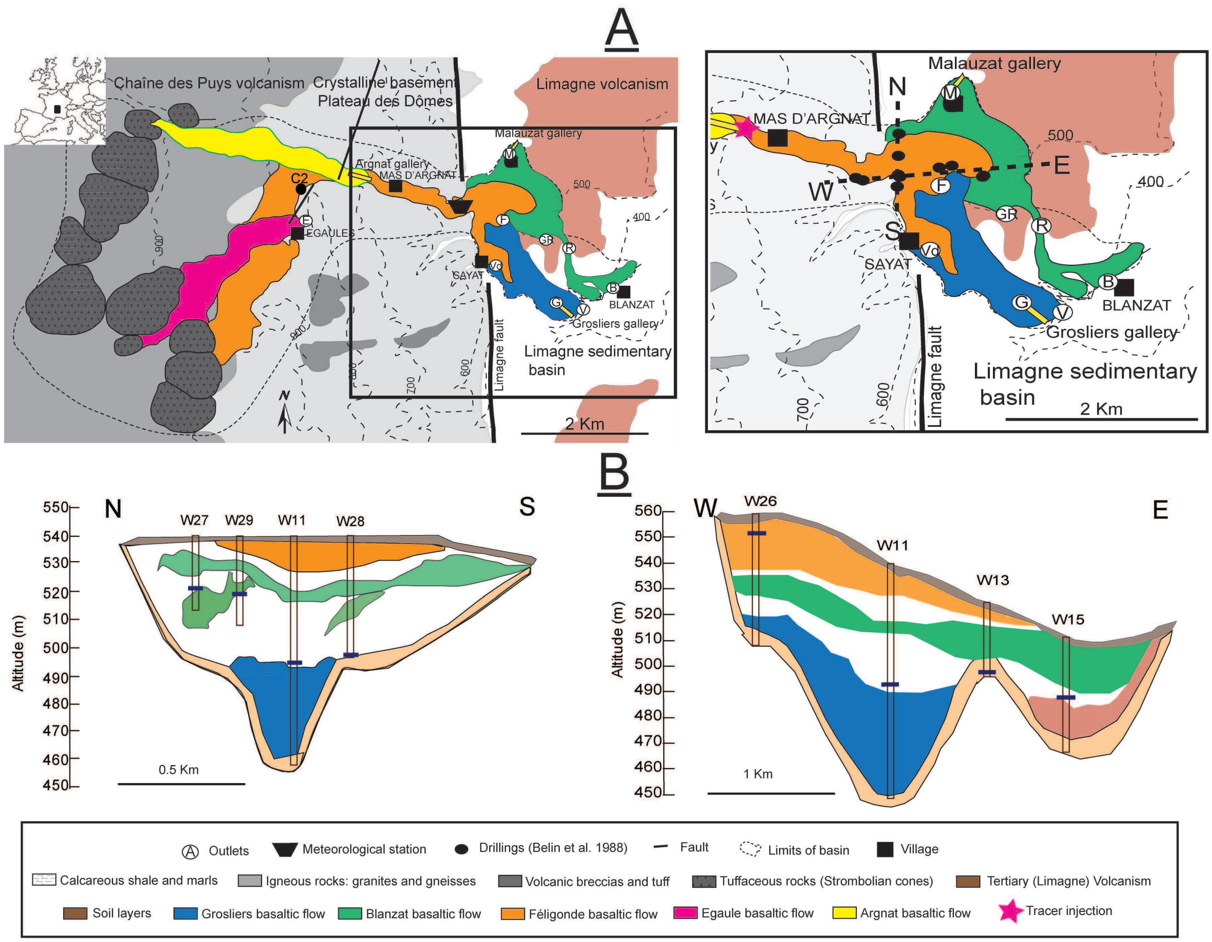

Figure 1: Location and geological settings of the Argnat basin; V: Vergnes, B: Blanzat, A: Argnat, E: Egaule, R: Reilhat catchment, Vd: Vernède, F: Féligonde, M: Malauzat, GR: Grande source de Reilhat, G: Grosliers.

nated from different strombolian volcanoes located in the central part of the Plateau des Dômes, and superimposed in a deep and narrow thalweg cutting the plutonic basement. The Grosliers volcanic flow settled first in a deep thalweg down to the locality of Saint-Vincent. The subsequent Blanzat lava flow took place in the same paleothalweg before the Limagne fault, then diverged to the North over the Limagne volcanics (of Tertiary age) locally present in the plain and stopped next to the present day village of Blanzat. Afterwards, it was overlain by the Féligonde lava flow extending to the village of Féligonde. Two subsequent lava flows, the Argnat and the Egaule flows, occurred upstream and stopped before the Limagne fault. Further erosion processes led to a relief inversion: sedimentary terrains overlaid by lava flows were conserved whereas surrounding areas were eroded (Figure 1A). The lava flows of the Argnat basin harbor two types of morphology. The Grosliers lava flow is of pahoehoe type, that is massive and poorly fractured [18, 19], harbouring a thin brecciated layer at the base. In contrast, the other lava flows are of a´a morphology, i.e., constituted by a mix of scoria and fractured basaltic blocks, with more extended brecciated basal and superficial zones [9, 19].

From a hydrogeological point of view, the drillings performed by Belin et al. [48] highlighted the existence of different piezometric surfaces (Figure 1B), suggesting strong heterogeneity of the hydrogeological properties and a hydraulic discontinuity between various groundwater systems. A first groundwater level (drillings W27 and W29, W28 and W13) has been associated with a scoriaceous layer located between the a´a Blanzat and the pahoehoe Grosliers lava flows. A second deeper groundwater system was defined associating the piezometric marks of drillings W26 and W11, which corresponds to a deeper circulation between the Grosliers flow and the basement. Due to its pahoehoe morphology, the Grosliers volcanic flow may act as an aquitard and captive or semi-captive groundwater flows might occur within some massive parts of the

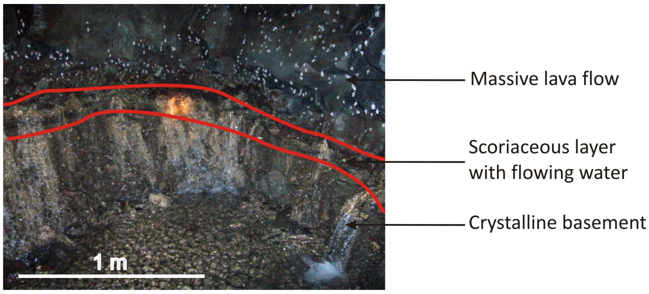

Figure 2: Grosliers gallery outlet.

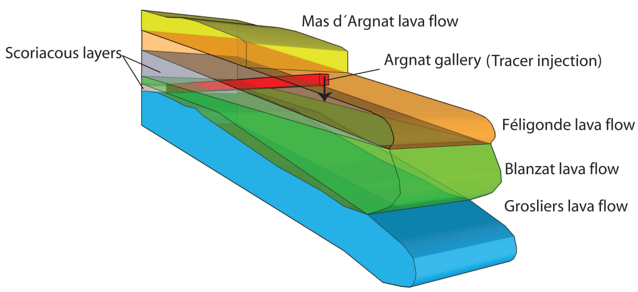

Figure 3: Scheme of Van der Min's field observations (1945, modified). Not to scale.

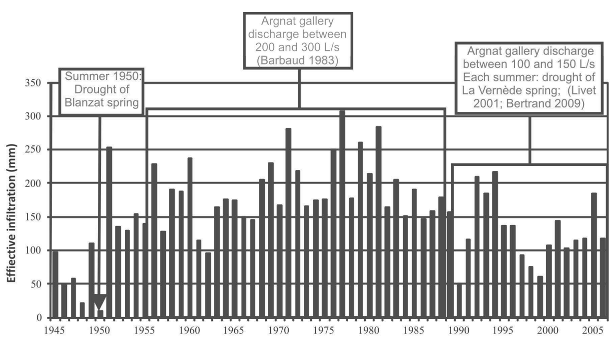

Figure 4: Annual effective infiltration on the Argnat basin from 1945 to 2006 and historically known hydrological evolutions (after Barbaud 1983; Livet 2001; Bertrand 2009).

structure [50, 54]. Consistently, field observations into the Grosliers gallery revealed that the water flow is limited to the basal scoriaceous zone (Figure 2). In addition, according to Boivin et al.'s investigations [51], the Féligonde spring, located between the Féligonde and the Blanzat lava flows, corresponds to a more superficial groundwater flowpath. Upstream, Van der Min [44] showed that the Argnat gallery catches water flowing within a scoriaceous layer located below several massive and porous basaltic layers (Figure 3).

In the Argnat gallery, the water is caught, drained to a chlorination reservoir and then used for domestic supply. The contractual catchment authorized to the exploitation company is 140 l/s. The excess is released into a fault located at the gallery exit.

Figure 4 shows the evolution of the calculated infiltration (or effective rain) rates for the 1945–2006 period at the basin scale according to Bertrand [55]. The mean rate is 157.5 mm/year, the maximum 307 mm occurred in 1977, and the minimum was 8.8 mm in 1950. Three main periods are defined by these data: (1) the 1945–1950 period during which the effective infiltration was the lowest, (2) the 1951–1988 period during which the effective infiltration remained below the average value of the two consecutive years, (3) the 1989–2006 period during which the effective infiltration reached the average value only four times. The discharge history of the Chaîne des Puys is unknown from these periods and few measurements were made until the beginning of the 21st century. However, some long-term trends for spring discharges across Europe have been published and show a similar temporal evolution as the Argnat basin effective infiltration. For example, Fiorillo et al. [56] reported that Serino's (Italy) spring discharge was lower than usual during the 1948–1959 and 1991–2004 periods. Lorenzo-Lacruz [57] found similar trends by averaging discharges of 45 rivers across the Iberic Peninsula. Known spring droughts or discharge decrease periods reported in the above-mentioned literature correspond closely to the deficit of infiltration periods, as shown in Figure 4. From the end of the 80s, La Vernède spring (Figure 1A) dried frequently between June and September [26, 55] and even dried completely from November 2005 to February 2006 and from June 2006 to January 2007 [55]. The Argnat gallery average discharge was around 200 l/s from 1983 to 1986 [26], but this value ranged only from 105 to 125 l/s between 2005 and 2007 (Figure 5). In addition the local population often refers to a total drying of the Blanzat spring during the summer 1950, although no historical document was found to support this. These facts suggest a limited storage capacity of the system and for a limited transit time of groundwater, which is around 1.5 years in the Argnat basin, according to Barbaud [28]. This could be of concern for the pollutant dilution, potentially impacted by infiltration rate modifications in case of climatic changes.

During the tracer test, in April 2007, the measured discharges (with a propeller type current meter or through gauged capacity) were about 13.5±2 l/s at La Vernède spring; 0.3±0.1 l/s at the Féligonde spring; 0.7±0.2 l/s at the Blanzat spring, 10.3±0.1 l/s in Les Grosliers gallery; and 120 ±5 l/s in the Argnat gallery (average over April; Figure 5).

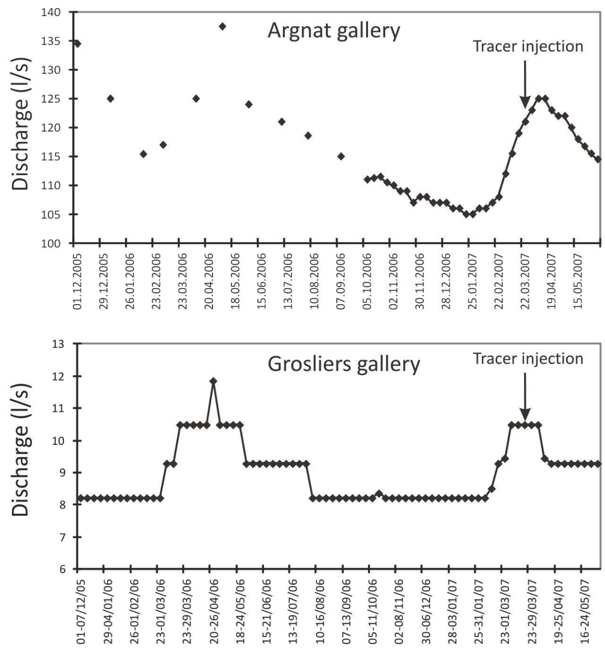

Figure 5: Discharge monitoring of the Argnat (punctual observations) and the Grosliers (weekly averaged) galleries (Dec. 2005–June 2007).

3 Methodology

3.1 Field methodology

The tracer test was carried out using potassium iodide (KI). Iodide (I^-) has been recognized to have a good chemical stability, a low tendency to adsorb, and to be harmless for people, fauna and vegetation [58]. Moreover, its quantification can be done in the field and with a low detection limit [30, 58, 59]. The probability of ionic adsorption by clays or organic matter [60] was considered low as these elements were not detected in significant amounts in the system [55]. In addition the tracer was injected directly into the subsurface, avoiding any influence of soil material. Some studies addressing matrix diffusion have shown that this tracer may be used accurately in silicate rocks [61].

An injection of 4.5 kg of potassium iodide was performed on 27/03/07 at 10:20 a.m. at the Argnat gallery exit where the excess volume of water(i.e. that not used to supply the public network) is released (Figure 1A; Figure 2). The discharge flow was set at 10 l/s during and after the time of injection (8 minutes in total). Automatic samplers were set up at La Vernède, Féligonde, Blanzat springs and Les Grosliers gallery (Figure 1A). The tracer test may therefore be considered as representative of the whole downstream part of the watershed, i.e. the Féligonde, the Grosliers and the Blanzat flows. The other outlets were not monitored for accessibility and/or security reasons. The automatic samplers were programmed to sample water every 4 hours.

The iodide concentrations were measured directly in the field by using a ISE 25 I probe (Radiometer Analytical) linked to a portable multimeter Sension 2 (Hach). Each analysis was performed in a sampling beaker after the addition of KNO_3 (0.1 M) which ensured a sufficient ionic force level for measurement accuracy. The accuracy is 0,5% of the detected concentration [62].

3.2 Estimation of hydrodispersives parameters

In order to characterize water pathways and hydrodispersive parameters of aquifers from tracer tests, we mathematically evaluated the breakthrough curves (BCs) using specific modelling procedures. The three-dimensional transport of an ideal tracer (such as iodide) can be described with the advection-dispersion equation (Eq. 1; [63]):

$$\frac{\partial C}{\partial t} = D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} + D_z \frac{\partial^2 C}{\partial z^2} - \upsilon \frac{\partial C}{\partial x} \quad (1)$$

where C is the concentration of the tracer, D_x isthe longitudinal dispersion in x-direction, D_y, D_z are the transversal and vertical dispersions, υ is the mean flow velocity, x, y, z are the spatial coordinate variables, t the time variable.

Several 1-D analytical solutions were defined for porous [63, 64], fissured [29, 30] or karstic aquifers [65]. Taking into account the diversity of the possible circulation media occurring in the Argnat basin (porous, fissured, mixing of these latters), and the fact that the tracer test was initiated in a massive fissured lava flow we employed the Single Fracture Dispersion Model (SFDM) [29, 30]. This method can be used for instantaneous injection, i.e. Dirac input function, which can be assumed from the comparison between injection and breakthrough durations. Furthermore, examples given by Maloszewski and Zuber [29, 30] suggested that experiments with a mean flow time up to 1 month are usually easily interpretable by the single-fissure approximation. This solution takes into account the behaviour of both the mobile water located in one main flow path and of the immobile water distributed in the matrix porosity or stagnant zones (Eq. 2 and Eq. 3).

$$\frac{\partial C}{\partial t} + \upsilon \frac{\partial C}{\partial x} - D \frac{\partial^2 C}{\partial x^2} - \frac{n_p D_p}{b} \frac{\partial C_{im}}{\partial y} \Big|_{y=b} = 0 \text{ for } 0 \le y \le b, \quad (2)$$

$$\frac{\partial C_{im}}{\partial t} - \frac{D_p}{R_{ap}} \frac{\partial^2 C}{\partial y^2} = 0 \text{ for } b \le y \le \infty, \quad (3)$$

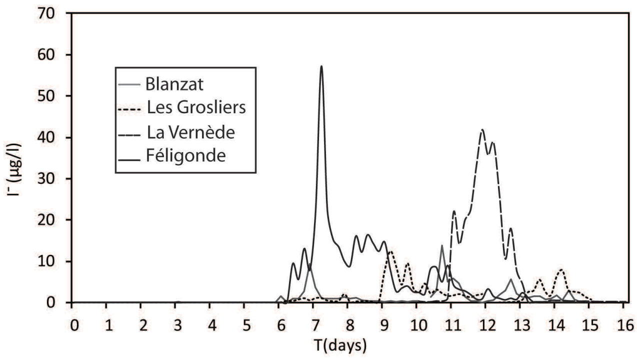

Figure 6: Breakthrough curves of the tracer test.

where C and C_{im} are tracer concentrations in mobile and immobile water respectively; D is the dispersion coefficient, y is the distance perpendicular to the flow axis x, while b and v are the half width and the mean water velocity respectively of the zone with mobile water. D_p is the effective diffusion coefficient of tracer in the stagnant water, equal to D_m/τ_p(D_m is the molecular diffusion coefficient in free water, and τ_p is the tortuosity factor for the matrix, usually fixed at 1,5 [30]), R_{ap} is the retardation factor for the porous matrix governed by instantaneous equilibrium adsorption (R_{ap} = 1 for an ideal tracer).

For Eq. 2 and 3, boundary and initial conditions are reported in Eq. 4a–4f:

$$C(x, 0) = 0 \quad (4a)$$

$$C(0, t) = \left(\frac{M}{Q}\right)\delta(t) \quad (4b)$$

$$C(\infty, t) \quad (4c)$$

$$C_{im}(y, x, 0) = 0 \quad (4d)$$

$$C_{im}(b, x, t) = C(x, t) \quad (4e)$$

$$C_{im}(\infty, x, t) = 0 \quad (4f)$$

where M is the injected mass of tracer, Q is the volumetric flow rate through the system, and $\delta(t)$ is the Dirac delta function. Eq. 4b defines the flux mode of tracer concentration in the fracture and Eq. 4f defines the single-fracture approximation which has been shown to be valid for short-term tracer tests (for tests lasting no longer than a few weeks [29, 30]) as it is assumed that the tracer has insufficient time to penetrate the matrix deep enough to be influenced by adjacent fractures.

By taking into account Eq. 4a–4f and performing mathematical transformations detailed by Maloszewski and Zuber [30], C_{im} and its derivative in y may be expressed as a function of C which is the tracer concentration in the fissure expressed along the x-axis(supposed homogeneous along the fissure width; Eq. 4e). These conditions and mathematical transformationsallow a 1D solution to be obtained (Eq. 5 [30]) for the initial 2D transport problem described in Eq. 2 and 3:

$$C(t) = \frac{aM\sqrt{Pet_0}}{2\pi Q}\int_0^t exp\left[-\frac{(t_0-\xi)^2}{4\xi t_0} - \frac{a^2\xi^2}{t-\xi}\right]\frac{d\xi}{\sqrt{\xi(t-\xi)^3}} \quad (5)$$

where ξ is the integration variable.

The fitting parameters are t_0 corresponding tothe mean transit time of water (Eq. 6), Pe, the Peclet's number (Eq. 7) and a called "diffusion parameter" (Eq. 8):

$$t_0 = x/v \quad (6)$$

$$Pe = vx/D = x/\alpha_L \quad (7)$$

$$a = n_p\left(D_p R_{ap}\right)^{1/2}/(2b) \quad (8)$$

For the known flow distance x, the mean water velocity and the longitudinal dispersivity α_L are obtained from t_0 and Pe. The fracture aperture $2b$ can then be calculated from Eq. 7, provided that the matrix porosity n_p and diffusion coefficient D_p are known.

4 Results and Discussion

4.1 Tracer recovery

The tracer was detected in all of the controlled outlets (Figure 6), indicating firstly a hydraulic link with the Argnat gallery water excess, and secondly the existence of vertical relationships between the different circulations determined by Belin et al. [48] (Figure 1B).

The tracer appeared approximately 6 days after the injection at Féligonde, La Vernède, Blanzat, and les Grosliers. Knowing that the distances between the injection points and these monitored points vary from 2400 to 4500 m (Figure 1), this suggests substantial variability of the hydrodynamic properties in the different volcanic formations. Using the time of the tracer's first appearance, it was calculated that La Vernède and Féligonde possess maximum velocities (227 and 400 m/day respectively) lower than values evaluated for springs located at

Table 1: Calculated tracer first apparition velocities for each spring. The values of first appearance velocity correspond to appearance with with [I⁻] > 1 µg/l. Latitude and longitude are indicated according to the WGS 84 coordinate system

Spring/gallery	Z (m a.s.l.)	X	Y	Range of first appearance velocity (m/day)
Argnat gallery leaving (injection point)	679	45°50'39"N	03°03'57"E	
Vernède	481	45°49'50"N	03°02'54"E	227
Féligonde	506	45°50'12"N	03°03'06"E	400
Grosliers	420	45°49'20"N	03°03'57"E	675
Blanzat	385	45°49'35"N	03°04'29"E	643

the end of volcanic flows such as Blanzat (643 m/day) and Les Grosliers (675 m/day) (Table 1). These values are consistent with the water circulation velocities of approximately 600 m/day calculated by Belin et al. [48] for the same two outlets. Similarly, Bouchet [47] calculated values for the modal velocity ranging from 1056 to 1680 m/day in the Aydat volcanic flow located southward of the Chaîne des Puys. These values confirm that volcanic flows may harbour extremely rapid water transfer such as those in karstic systems. In the present study, the fissured feature of lava flows coupled with high gradient slopes, may explain this similarity. This is of concern regarding their vulnerability to surface pollution.

The breakthrough curves generally showed multimodal features. However because no important rain event or discharge variations were observed during the test, this can be attributed to the existence of distinct groundwater pathways supplying the springs. A more detailed analysis of BC´s indicates that the tracer release is multimodal and weaker for the more distant outlets (Blanzat and Les Grosliers) than for the closest springs, which in contrast present uni- or bi-modal BC´s and higher concentrations (La Vernède and Féligonde).

The total recovery rate, estimated by integrating BCs and taking into account the discharge of each outlet, is 2%. This suggests the existence of other important connections between the Argnat gallery outlet and other non-monitored outlets. However, Belin et al. [48] similarly obtained a very poor recovery rate during tracer tests performed with 2.5 tons of salt, from the drilling C2 to the Argnat gallery and Reilhat (Figure 1A). This was attributed to the existence of slow groundwater flows that would have been identified with longer monitoring. This hypothesis was proposed by Witthüser et al. [66] in a fractured chalk aquifer to explain a recovery rate of about 4% between two wells located 10 m apart.Similar explanations have been proposed in works focusing on alpine karst systemswhere some springs showed a recovery rate lower than 1% [40]. Slower flows should likely occur in a fine-grained system such as the scoriaceous layers of the aquifer (Figure 1B). A part of the tracer may also be stored in low hydraulic conductivity brecciated zones (scoriaceous layers) or in perched brecciated areas.

These first observations suggest two kinds of hydraulic heterogeneities: (1) large scale heterogeneity related to the alternation of two types of rocks, massive fissured lava flows and porous (scoriaceous) layers, which could explain the low recovery rate and (2) intrinsic heterogeneity within the massive fissured rocks only, which could explain the multimodal feature of BC´s.

According to these preliminary observations, the functioning of the fissured massive lava flow will be evaluated and discussed to determine what the parameters are that control flow and transport within massive lava flows, and what their consequences are in terms of system vulnerability to pollution events.

4.2 Hydrodynamic properties of massive lava flows

Assuming a dual porosity (fissured and matrix) within the massive parts of lava flows, the evaluation of dispersivity should determine both the width of the fracture and the matrix porosity of the massive volcanic flows. This was done through the SFDM as reported by Eq. 3. Results of the SFDM can be seen on Figure 7 for La Vernède and Féligonde. Because of the weak iodide concentrations found in Blanzat and Les Grosliers, modelling was not performed for these two points. The fitting and resulting parameters for each modelled curve of La Vernède and Féligonde BC's are compiled in Table 2. The mean longitu-

Table 2: Hydrodynamic parameters for each BC's of La Vernède and Féligonde. The fitting parameters are typed in bold. t_0: Mean transit time; υ: Mean velocity; α_L: Longitudinal dispersivity; Pe: Péclet's Number; a: Diffusion parameter.

	Single Fracture Dispersion Model parameters
La Vernède	**t_0 = 11.83 d**
	υ = 211.26 m/d
	α_L = 1.78 m
	Pe = 1400
	a = 0.0148 $h^{-1/2}$
Féligonde BC 1	**t_0 = 7.06 d**
	υ = 342.85 m/d
	α_L = 0.49 m
	Pe = 4900
	a = 0.0365 $h^{-1/2}$
Féligonde BC 2	**t_0 = 8.96 d**
	υ = 267 m/d
	α_L = 18.46 m
	Pe = 130
	a = 0.0315 $h^{-1/2}$

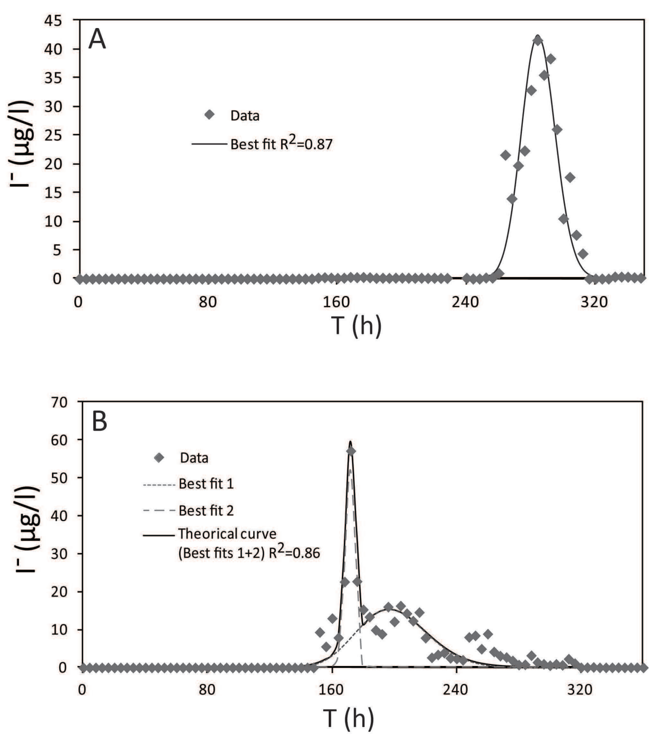

Figure 7: Single Fracture Dispersion Model (SFDM) applied for La Vernède (A) and Féligonde (B)'s tracer breakthrough curves.

dinal dispersivity for La Vernède is 1.6 m for 2500 m travelled. The Féligonde´s BC was considered the result of two circulations (BC 1 and BC 2, Figure 6). BC 1 presents a low dispersivity of approximately 0.65 m whereas the one of BC 2 is approximately 28.9 m. These values are in the lower range of data reviewed by Gelhar et al. [33] and references therein] for the scale of only a few kilometres, and not only show the hydrodispersive particularity of the massive volcanic structures but also indicate that the tracer found in the springs was mostly carried by advection. This was also illustrated by the high Peclet numbers used to fit the 3 BC´s, ranging from 130 to 4900.

In order to characterize the flowpath medium from a geometrical point of view, the fitting "a" parameter in the SFDM (Eq. 5) may be used to calculate the mean aperture $2b$ of the conductive fissures (Eq. 8). For this purpose, the matrix porosity n_p and diffusion coefficient D_p of the tracer in the porous matrix have to be independently known. The estimation of the matrix porosity n_p was performed in two steps according to Novakowski et al. [67]. Firstly Poiseuille's law and the difference in piezometric heads between the injection point (Argnat gallery surplus) and the monitored outlets (198 m a.s.l. and 173 m a.s.l., respectively for La Vernède and Féligonde; cf. Table 1 for piezometric heads) is used. From these data, the fissure aperture $2b$ can be estimated as:

$$2b = \sqrt{\frac{12\mu\tau_f x^2}{g t_0 \Delta H}} \quad (9)$$

where g is the gravity acceleration, ΔH is the difference between the hydraulic heads, μ is the kinematic viscosity of water (at 10°C, mean groundwater temperature [53]), τ_f is

Table 3: Calculated mean fracture aperture and porosities of the matrix.

	2b (μm)	n_p (%)
Vernède	272	0.20
Féligonde BC 1	363	0.60
Féligonde BC 2	321	0.46

the tortuosity factor of the fractures (assumed to be equal to 1.5; [30, 68]).

Secondly, once the calculated mean fracture aperture $2b$ is known, it is then possible to use this value in the Eq. 8 in order to estimate the matrix porosity. The value of D_p is estimated as:

$$D_p = \frac{D_m}{\tau_f} \tag{10}$$

D_m ,the iodide molecular coefficient diffusion in free water is 2,04.10^{-9} m^2/s [69].

The calculated fissure apertures and associated matrix porosities are reported in Table 3. The fissure apertures $2b$ range from 272 to 363 μm. A similar calculation performed by Reimus et al. [33] yielded values from 850 to 12800 μm in volcanic tuff of the Yucca Mountain. Himmelsbach et al. [70] calculated values between 100 and 300 μm in a low permeability section ($K = 5{\cdot}10^{-10}$ to 10^{-8} m/s) and between 400 and 600 μm in the most permeable zone ($K = 5{\cdot}10^{-6}$ to 10^{-4} m/s) of the Lindau site (Germany). According to the above examples our results suggest that the studied flowpaths occurred in moderately permeable fissures.

The calculated matrix porosities n_p ranging from 0.2 to 0.6% (Table 3), appear to be low. As a comparison, the geophysical approaches performed within the Chaîne des Puys [46] estimated effective porosities of around 8% for volcanic flows. The Canary Island Quaternary basalts show effective porosities between 2 and 5% [22], and between 1 and 2% [71]. Chandrashekhar et al. [72] indicated that the n_p of fissured basalts in the Karanjo basin (India) ranged from 0.9 to 1.7%, whereas Deolankar [73] found n_p to be equal to 1% at the Deccan plateau (India). The slightly lower values found in the present study could be explained by the fact that only the matrix effective porosity (and not fissure with matrix) has been evaluated. Consequently, if an average effective porosity of 8% is assumed for the Chaîne des Puys massive lava flows [46], it can be argued that the greater part of this porosity is related to fracture porosity rather than to the matrix porosity. This strengthens the general assumption that the massive part of lava behaves as a good conduit system but as a poor water reservoir in contrast to scoriaceous layers [22, 74].

4.3 Supply of Argnat basin springs and implication for the long-term management of volcanic aquifers

The combination of the recovery rate, the hydrodispersive parameters, and the information provided by drillings allows the design of a conceptual scheme of groundwater and chemical transit in the downstream part of Argnat basin (Figure 8). This scheme necessarily implies that there is a dual flowpath system supplying volcanic springs. Dual hydrological function is a well-known concept in karstic or fractured environments [29, 32, 34–40, 75–77] involving a fissure passing through a matrix porosity. However in the context of volcanic systems, this duality should be understood in a different way in regards to the above results. The low-concentrated and multimodal tracer releases observed are consistent with a fissured system (massive lava flows that are locally fractured or fissured)which is poorly dispersive (cf. Table 2) due to low matrix effective porosity (cf. Table 3) and is superimposed on to a (main) discharge from scoriaceous layers located between the lava flows. This is suggested by both the poorly variable spring discharge (Figure 5) similar to those observed in outlets of porous aquifers [14] and the low recovery rate of the tracer after relatively few weeks.

Accordingly, if chemical pollution occurred it would probably be highly diluted by the large volume of groundwater flowing within the saturated scoriaceous parts. Consequently, aquifer vulnerability should depend greatly on the sustainability of the quantity of groundwater. This quantity depends on the recharge rate and on the mean transit time of the water (between recharge area and outlets) related to the water flow velocity and storage capacity of the system [75, 78]. The current lower discharge in the main outlets of the basin – in comparison to the 1951–1988 period as well as the drought of the summer 1950 – suggests a rapid decrease of the storage that could be linked to rapid transit times within the system [28]. Therefore, besides the problem of groundwater quantity that is of concern for management authorities [15, 79], limited storage also implies that the dilution potential of the system may be threatened. The rapid transit of a portion of the pollution through fissured parts and the possible lower-

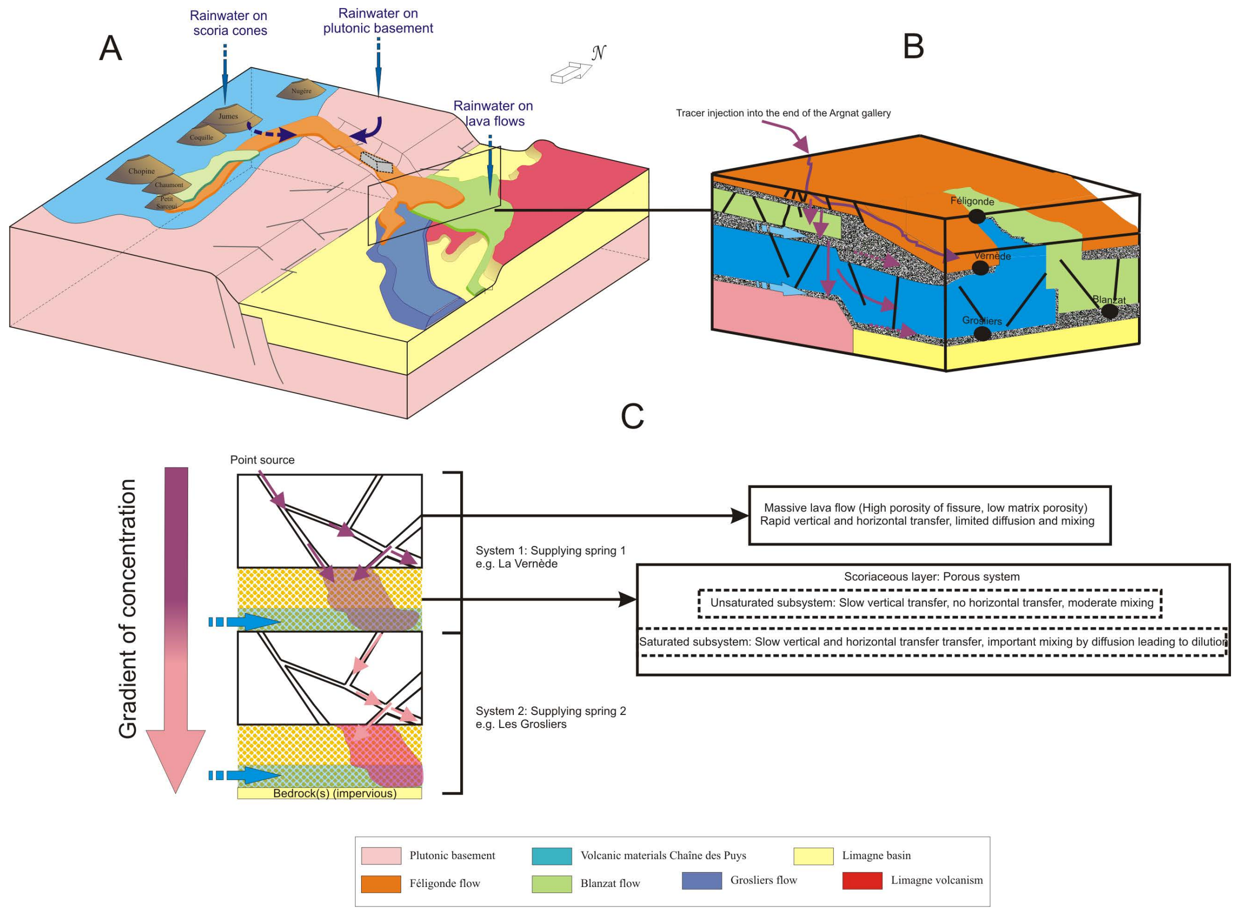

Figure 8: Conceptual schemes of the supply of springs located at the end of lava flows; A) Water supplies at the basin scale; B) Inferred tracer flowpaths from the tracer recoveries; C) Schematic representation of the processes affecting tracer transfers.

ing of groundwater recharge rates (with higher recharge rate favouring dilution) in temperate areas due to climate change impact [78, 79] should therefore both be considered to allow the sustainable management of the system.

This conceptual scheme also suggests that the position of the spring in relation to the water table is a key factor influencing its vulnerability. This is especially so in volcanic systems. Indeed, the superposition of massive lava flow and scoriaceous layers implies that the springs collecting the water circulation in scoriaceous layers will be less impacted by point source chemical pollution due to dilution processes. However, aquifer resilience after pollution should be longer due to slower groundwater transit within the scoria. From a quantitative point of view, however the discharge would unlikely stop, even in low recharge periods. In contrast, springs or catchments located in with a massive lava flow would be more sensitive to pollution (rapid transfer through fissures, less dilution) and to rapid water table lowering due to limited storage capacity.

The discussed vulnerability to pollution assumes steady-state conditions of discharge. However Legout et al. [80] have shown that in order to obtain a complete view of the chemical vulnerability of dual porosity aquifers, one should also take into account transient state processes. In particular, the degree of saturation of lava flows that can constitute temporary or permanent perched aquifers [5, 9, 13, 54] is a key consideration. A pollutant may be present alternatively in partially or totally saturated conditions along the flowpath. During low flow periods, the saturated zone is recharged by water flowing vertically through the slow-mobile porosity of the unsaturated zone and the fluctuation zone (temporarily saturated or unsaturated). During the water table increase period, rainwater percolates through the matrix (slow) porosity of the unsaturated zone as long as the rainfall input rate does not exceed the infiltration capacity of the matrix. Once this threshold is reached however, rainfall excess is rapidly transferred vertically through the fissures of the unsaturated zone.When the fluctuation zone is saturated, the flow direction usually changes from vertical to sub-horizontal [81] and the groundwater then flows mainly through the fracture and interacts progressively along the flowpath with the matrix porosity. As a result, even if chemical pollution was previously introduced, its impact on water quality would be more significant when the water table rises if the pollutant behaves as a perfect tracer (that is with no adsorption and high solubility). On the contrary, if the pollutant is adsorbed, then pollution should be partially or totally buffered. If the pollution arrived during a high recharge period (as for the presented tracer test), an important sub-horizontal transport would occur and a part of the pollutant would be evacuated rapidly. As volcanic systems may present a patchwork of perched systems, vertical transfers may occur and a great part of the pollutant may reach the main and permanently saturated zone, leading to dilution through diffusion within the scoriaceous layers analogous to fine-grained systems. When the water table decreases however, the pollutant transfer in the fluctuation zone may be mainly vertical and may occur through the matrix because of unsaturated conditions.

The analytical evaluation of BC's provided a preliminary insight into matrix porosity in massive lava flows. It appeared very reduced according to the low observed longitudinal dispersivities. This means that saturation may occur very rapidly if the fissure flow is not connected to scoriaceous layers (typically the superficial and basal brecciated zones, cf. [9, 18]), and that any contamination would be evacuated rapidly given the poor matrix porosity and low dispersivity of massive lava if contaminant flows predominantly within the massive parts. In contrast, the scoriaceous layers should saturate slowly and smoothly due to their higher porosity, and a contamination reaching these zones should be released only in the long-term. Although these insights should be confirmed by the implementation of other tracer tests during contrasted hydrological situations, they suggest that an optimal protection of volcanic aquifers should require the mapping of the relative proportion (for example in thickness) of massive lava flow versus scoriaceous material at the watershed scale. Such a strategy is reminiscent of Bulkhölzer et al.´s insights [82] that recommend classifying dual porosity formations according to the value of the mobility number (the ratio of the aquifer flow rate carried out through the porous block of the aquifer to the discharge carried out by through the fracture network). However in a volcanic context, such as in the studied area which mainly featured a'a flows, a preliminary classification according to the ratio of massive lava flow versus scoriaceous layer thicknesses could provide a useful management tooland may highlight specific areas that require prioritized protection.

5 Conclusion

This study used an artificial tracer test to investigate groundwater and pollutant transfer within volcanic flows. This led to the development of a conceptual scheme of the Argnat basin groundwater flow characteristics.

The tracer was found at all the monitored springs between 6 and 11 days after the injection, indicating a hydraulic connection between the injection and monitored points. The high flow velocities (maximum velocities range from 227 to 683 m/d) were similar to groundwater speeds in highly karstified systems. The BC's had, in most cases, multimodal features. The SFDM approach for the Féligonde and La Vernède's outlets indicated that rapid transport occurs with limited longitudinal dispersion. This is consistent with flows within fissure zones located in massive rocks or in consolidated scoria masses. The matrix effective porosities are low in these massive lava flows, ranging between 0.2 and 0.6%.

The tracer recovery rate was low with a value of 2% for all of the outlets. This result may be explained by the lack of observation points, but the most probable explanation according to previous studies, is that a large amount of the tracer can be stored in the slow mobile porosity i.e. in the scoriaceous layers or in the perched brecciated areas.

Moreover, while the piezometric heads evaluations suggested hydraulic discontinuity in the aquifer system of lava flows, the tracer test demonstrated the hydraulic link between the three tested volcanic flows. Thus, even if each of the water circulations is largely independent in this kind of environment, they are not completely isolated.

Although the volcanic groundwater systems of the Chaîne des Puys are considered to present stable discharge conditions [27], their outflows are sensitive to the effective infiltration deficit which has occurred in the area for the last twenty years.This could also impact the dilution effect in the case of pollution. In order to obtain a more adapted and sustainable management approach of these complex systems, three investigative approaches could be developed and adapted for volcanic settings: (1) a more systematic monitoring of recharge and discharge of the springs; (2) an evaluation of transport parameters at distinct hydrodynamic conditions (e.g. low-flow, high flow and recharge periods) to characterize the transient state and the impact of transient processes; and (3) a comprehensive mapping of massive lava flow and scoriaceous layer thicknesses to identify the most vulnerable zones of the watershed in both the short-term (e.g. springs connected to massive lava flow) and the long-term (e.g., springs mainly supplied by the extended brecciated zones).

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