Groundwater-surface water exchanges in an alluvial plain in southern France subjected to pumping: A coupled multitracer and modeling approach

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Abstract

Study Region:

The study was conducted in an alluvial plain between the Rhône and the Ouvèze Rivers (in the southeast of France) extensively exploited for drinking water. The research area is characterized by significant groundwater-surface interactions influenced by groundwater pumping activities.

Study Focus:

The aim of this study is to enhance the understanding of interactions between rivers and alluvial aquifers by combined multi-tracer and numerical modeling approaches. Over an 18-month period, groundwater temperature, piezometric levels, and river surface water levels were continuously monitored. Field campaigns focused on conductivity, stable isotopes of water, and radon-222 activity concentration in both groundwater and surface water. Radon-222 was used to quantify water exchanges between the river and the aquifer. A MODFLOW model, calibrated using piezometric data and PEST, was employed to simulate groundwater flow and reactive transport of radon-222 using MT3DMS.

New Hydrological Insights for the Region:

The study reveals that river water recharges the aquifer, with radon-222 data delineating this recharge zone. The methodology extended the interpretation of periodic groundwater temperature signals to isotopic signals, allowing the identification of dispersivity and Darcy's velocity. The Ouvèze River was found to contribute approximately 55% of the pumping water supply, alongside the Rhône. These findings provide valuable insights for sustainable water resource management, demonstrating the relevance of using natural tracers in scenarios where artificial tracers are impractical.

Keywords

Alluvial aquifers, River-aquifer interactions, Multi-tracer approach, Numerical modelling, Groundwater recharge, Radon-222 tracers, Stable isotopes, MODFLOW model, Rhône River, Ouvèze River, Groundwater temperature monitoring, Water resource management, Sustainable water management, Hydrological study

1 Introduction

When surface water and groundwater are connected, the abstraction of one of them can impact both resources and their interactions. Modifying the direction and/or rates of their respective exchange fluxes can influence the long-term quantity and quality of surface water and groundwater (Lu et al., 2018; Shu and Chen, 2002; Sophocleous, 2002; Winter, 1999; Winter et al., 1999), affecting ecosystem equilibrium (Woessner, 2000), and downstream users (Winter et al. 1998). Alluvial aquifers hydraulically connected to rivers are widely exploited and represent a major water resource. Although the proximity to the river can ensure significantly high aquifer pumping rates, water quality problems can be encountered for those pumping fields along the riverbanks (Bertln and Bourg, 1994; Hiscock and Grischek, 2002). Quantifying groundwater-surface water exchange is essential for understanding and joint management of surface water and groundwater resources (Fleckenstein et al., 2010; Shanafield and Cook, 2014), as well as for possible contamination management (Boano et al., 2010; Chapman et al., 2007; Chen, 2007; Lamontagne et al., 2005; Trauth et al., 2018).

The characterization of groundwater-surface water exchanges as well as methods to describe them, both in quantitative and qualitative terms has been widely studied (Bernard-Jannin et al., 2017; Lu et al., 2018; Brunner et al., 2009; Rivière et al., 2014; Cardenas, 2009). Quantification of exchange is generally based on differential gauging or Darcy flow estimates using piezometric records and hydraulic conductivity values (Dujardin et al., 2014; Keery et al., 2007; Schmidt et al., 2007; Xie et al., 2016). However, estimates of Darcy flows based on piezometric level data are difficult due to problems in obtaining reliable hydraulic heads and hydraulic conductivity from piezometers, especially in the case of a riverbed and due that it is integrative in space (Keery et al., 2007; Surridge et al., 2005). In addition, the hydraulic conductivity of riverbed sediments is likely to vary by several orders of magnitude in space (Calver, 2001), adding further uncertainty to the Darcy flux estimates. Differential gauging corresponding to river flow measurement at different location of the river to determine gains or losses linked to the connection with the aquifer. This method is commonly used to estimate groundwater-surface water exchange flow at the watershed scale and allows reliable and inexpensive measurement and is generally applied to rivers with low to moderate discharge (from 0.001 $m^3 s^{-1}$ to 500 $m^3 s^{-1}$; (Grapes et al., 2005; Konrad, 2006; McCallum et al., 2012; Opsahl et al., 2007; Xie et al., 2016). However, its application becomes challenging for high flow rivers like the Rhone, necessitating fixed gauging stations rather than manual flow measurements (Konrad, 2006; Xie et al., 2016).

Artificial tracers are chemical compounds not naturally present in the water and cannot always be used due to large river discharge or to the proximity to sensitive sites (industrial, urban), so the use of natural tracer has been widely developed. Thermal signal transfer is a relevant tool for assessing water transfers from rivers to unconfined aquifers that has attracted great attention over the past two decades (Constantz, 2008; Keery et al., 2007). The advantage of temperature as a tracer, compared to hydraulic approaches, is that thermal properties vary less than hydraulic properties, such as hydraulic conductivity, thus reducing the uncertainty of exchange flux estimates (Boano et al., 2010; Lautz, 2010). The interest of temperature monitoring relies on its ease of implementation with inexpensive data loggers, even over relatively long times and with fine time resolution (Constantz, 2008; Constantz and Stonestrom, 2003; Keery et al., 2007). Most studies focus on vertical

temperature transfer from the river to the aquifer at the riverbed (Anderson, 2005; Constantz, 2008; Ghysels et al., 2021; Rosenberry et al., 2016; Xie and Batlle-Aguilar, 2017). Exchange fluxes are then estimated, by fitting an analytical solution to vertical riverbed temperature profiles (Constantz, 2008; Schmidt et al., 2007), by analyzing time series of temperature data (Anibas et al., 2011; Rosenberry et al., 2016), or by means of coupled modeling of groundwater flow and heat transport (Anibas et al., 2011; Schmidt et al., 2007). Temperature is rarely used directly to quantify lateral (horizontal) exchange between streams and aquifers but can be used as a calibration variable to constrain numerical models (Bravo et al., 2002; Brookfield and Sudicky, 2013; Tang et al., 2017). In these cases, piezometers and temperature sensors are often installed near the river (Engelhardt et al., 2011; Gerecht et al., 2011; Musial et al., 2016).

Along with temperature, the use of radon-222isotope is considered one of the most effective and well-established tracing techniques in hydrogeological studies (Kurylyk et al., 2018; Rau et al., 2010; Ren et al., 2018). This natural radioactive tracer is generally used to quantify the groundwater flow towards surface bodies such as coastal lagoons or rivers (Cook et al., 2018, 2006; Dugan et al., 2012; Hoehn and Von Gunten, 1989; Stellato et al., 2008; Stieglitz et al., 2013, 2010). Radon-222 is a naturally occurring gaseous radioactive isotope with a short half-life (3.8 days). In groundwater, the activity concentration of radon-222is high, because it is produced by the decay of radium present in the soil minerals (Kraemer et al., 1998). Surface waters generally have low radon-222activity concentrations due to the negligible radon-222 content in precipitation, out gassing in the water column, and its short radioactive half-life (Bertln and Bourg, 1994; Cecil and Green, 2000; Cook et al., 2006; Dugan et al., 2012). This difference allows for radon-222 to be used as a tracer of groundwater discharge to surface water bodies (Hoehn and Von Gunten, 1989; Mayer et al., 2018; Stellato et al., 2008). Radon-222 mass balance calculations enable the quantification of radon-222 and therefore groundwater fluxes upon quantification of radon-222 concentration in groundwater (Cable et al., 1996; Cook et al., 2006; Corbett et al., 1997; Ellins et al., 1990). However, the situation of a losing river, i.e., surface water supplying the groundwater is less studied. In this situation, it is possible to obtain qualitative information on surface water groundwater exchange, when the boreholes are close to the river, and quantitative information, such as velocity and transit time, by analyzing the continuous radon-222 record (Close et al., 2014; Hoehn and Von Gunten, 1989).

Stable isotopes of water (^{18}O and ^{2}H) are commonly used as conservative tracers in hydrology. Isotopic methods rely primarily on the Meteoric Water Line (MWL), which expresses a linear relationship between $\delta\,^{2}H$ and $\delta\,^{18}O$ values due to isotopic fractionation during condensation of water vapor mass (Craig, 1961; Clark and Fritz, 1997) and represents the natural variations of the meteoric water composition before the influence of post-precipitation evaporation processes. The isotopic composition of groundwater has been widely used to determine the origins of groundwater recharge, estimate mixing proportions between different sources, quantify recharge rates (Clark and Fritz, 1997; Engelhardt et al., 2011; Fette et al., 2005; Gat et al., 1969; Gonçalvès et al., 2015; Séraphin et al., 2016; Sharma and Hughes, 1985; Vallet-Coulomb et al., 2017). The application of stable water isotopes also makes it possible to improve numerical modeling of groundwater-surface water interactions, in particular by distinguishing different sources of water or by evaluating the recharge (Binet et al., 2017; Long and Putnam, 2004; Perrin et al., 2003). Especially when contrasted signatures can be identified between a river and its accompanying alluvial

aquifer, as is the case for rivers fed at high altitude, the application of stable isotopes of water also makes it possible to trace the surface water – groundwater exchanges and to determine the transit times (Poulain et al., 2021a, 2021b).

Tritium (^3H) is another valuable tracer that could have been considered for this study, particularly due to the presence of nuclear power plants along the Rhône River, which release tritium at relatively high concentrations into the surface water. Tritium, with its longer half-life of approximately 12 years, has been used effectively in previous studies to investigate groundwater-surface water interactions, as demonstrated by e.g. Engel et al. (2024), Zill et al. (2023), and Schubert et al. (2020). However,this study focused on natural tracers that are less influenced by anthropogenic sources, with the aim of understanding the natural dynamics of river-aquifer interactions and to explore methods that could be broadly applicable in other regions where tritium may not be as prevalent or where nuclear facilities are absent.

The integration of spatial and temporal data from field and laboratory experiments with physically based numerical models is a current focus of research. This approach assists in uncovering important dynamics and enhances our comprehension of various processes. (Binet et al., 2017; Fleckenstein et al., 2010; Ghysels et al., 2021; Gilfedder et al., 2015; Jiang et al., 2019). Regarding groundwater-surface water exchanges, regional studies most often use MODFLOW type models with rivers represented by Cauchy type or prescribed hydraulic head boundary conditions (Cousquer et al., 2017; Di Ciacca et al., 2019; Morel-Seytoux et al., 2018, 2014; Rushton, 2007). However, calibration does not guarantee the reliability of a model, indeed several problems can arise as the possibility of multiple parameterizations (recharge, steady state hydraulic conductivity) to obtain the same result, the so-called equifinality issue (Bravo et al., 2002; Cousquer et al., 2018, 2017). In this context, the acquisition of constraining data (e.g., tracers) to increase the reliability of the variables and parameters (hydraulic conductivity, recharge, dispersivity…) is fundamental to improve the model quality (Cousquer et al., 2018; Delottier et al., 2017; Fleckenstein et al., 2010; Gardner et al., 2011). Although tracers can be individually used, the information obtained by combining different techniques (tracers) can validate the results and overcome the limitations of a single method (Dujardin et al., 2014; Gardner et al., 2011; Gilfedder et al., 2015; Sadat-noori et al., 2021; Stellato et al., 2008; Xie et al., 2016).

In this study, we combined a multitracer approach and numerical modeling to quantify the groundwater-surface water interactions at the local scale, in the context of groundwater exploitation by riverside pumping. A catchment field located on the banks of the Rhône River, in southeastern France, was chosen as the study site. This site has the particularity of being located between two rivers (Rhône and Ouvèze), thus likely prescribing hydraulic head values at river stages on each side if the rivers are connected to the exploited aquifer. As the pumping wells are located only a few dozen meters from the Rhône, there is a risk of pollution by the river. The main aim of this study is to accurately determine the origin and proportions of the various water sources feeding a well system on an alluvial plain. This study needs the following steps:

i) Identification of the water sources feeding the pumping wells using at least two distinct tracers (radon-222 and stable water isotopes) and the piezometric data (hydraulic head (m));

ii) Analysis of temperature seasonality in the piezometers and Rhône River to estimate pore velocity (u), based on known porosity;

iii)Use of δ18O seasonality in piezometers to constrain dispersivity, a crucial but often poorly known property;

iv)The use of steady-state flow modelling coupled with reactive transport (radon-222 with radioactive decay) to confirm the origins and determine the proportions of the pumped water mixture, using pore velocity and radon-222 signal for model validation.

Through these steps, the study aims to provide a comprehensive understanding of water dynamics in the alluvial plain, essential for sustainable management of water resources.

Piezometers and pumping wells were equipped with pressure and temperature sensors. In addition to continuous piezometric monitoring, various methods of evaluating the water exchanges between the alluvial aquifer and the river were implemented to obtain robust information on the nature and direction of the flows. The transfer of the seasonal (periodic) temperature signal from the Rhône River to the aquifer was monitored to quantify the lateral water exchange as well as the thermal diffusivity of the aquifer (measure of how heat flow occurs within the material). Several radon-222 and electric conductivity measurement campaigns were carried out to determine the nature of the exchanges in different operating situations (pumping or no pumping). The temporal monitoring of stable isotopes of water allows us to identify the different sources of water supplying the pumping wells and to characterize the mixing. By transposing the thermal periodic signal interpretation method and thus its analytical solution to the mass transfer problem, the stable isotopes of water time series analysis could enable quantifying the dispersivity of the medium. These different methods lead to a comprehensive understanding of the hydrodynamic behavior of the aquifer in a robust and repeatable way in the very common context of groundwater exploitation in an alluvial plain. All these data were aggregated and used to constrain a local hydrogeological model using MODFLOW and highlighting the vulnerabilities of the site regarding possible contamination coming from the rivers.

To our knowledge, we propose several new approaches to improve our understanding of alluvial aquifers in the context of river-groundwater exchange. While radon-222 is conventionally used for estimating groundwater discharge in rivers, it has been employed here to determine water flow from river to aquifer, characterized by the dilution of the aquifer's natural radon-222 signal by radon-depleted river waters. Previous studies typically focus solely on either gaining river or losing river scenarios. However, it is uncommon to altern gaining/losing scenarios at the same site. This unique situation, made possible by an exceptional cessation of pumping, facilitated the efficient use of radon-222 to identify the origin and proportions of the water supplying the pumping wells. To our knowledge, the use of radon-222 data in transport models is also rarely discussed and represents a further addition to improve and constrain numerical models. In addition, we propose an extension of the standard 1D interpretation of the periodic temperature signal to the periodic signal of stable water isotopes. Interpreting a natural tracer such as stable isotope of water, in the context where the use of artificial tracers is not feasible, theoretically allows assessing the poorly known dispersivity of the alluvial aquifer.

2 Study site, material and method

2.1 Field Site

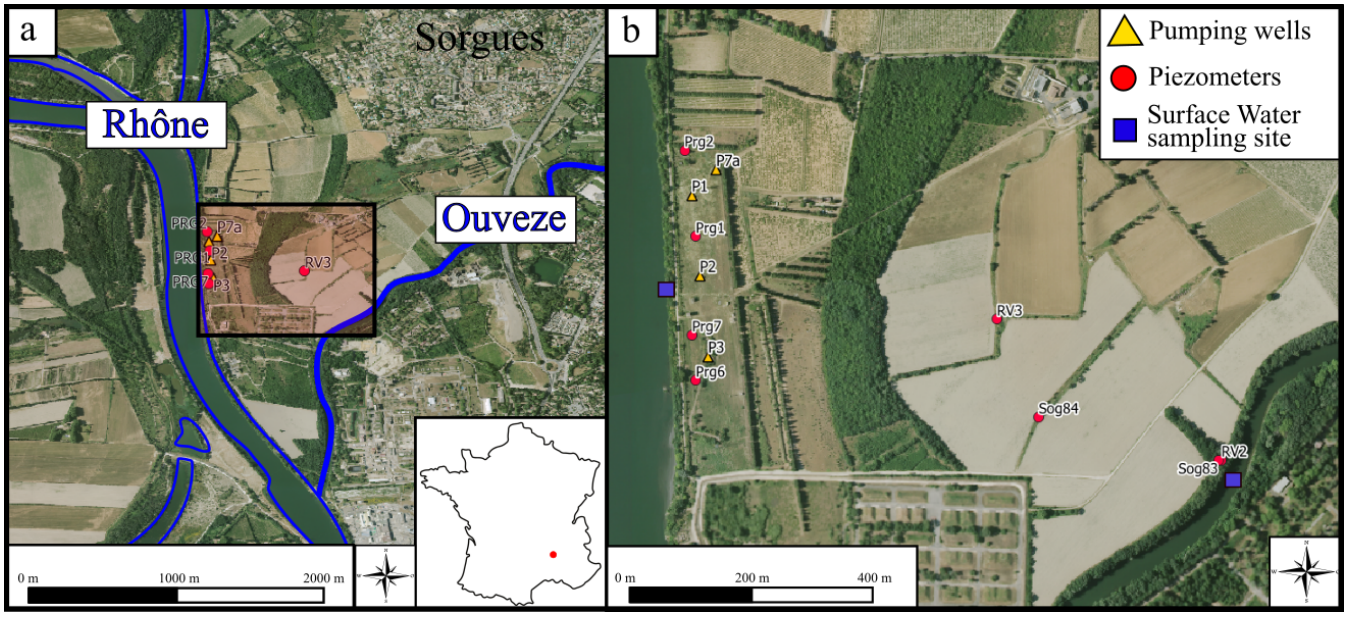

Figure 1 : a) Map of the study area the Jouve site (Vaucluse, France) located in the red inset b) the monitored pumping wells and piezometers symbolized by yellow triangles and red points respectively. The water surface sampling sites are symbolized by blue squares (© Google Earth 2022).

The study site located at Sorgues (Vaucluse, Southeast of France, Figure 1) is the domestic water extraction site of La Jouve composed of pumping wells aligned along the Rhône riverbanks. The pumping site is located upstream of the confluence of the Rhône (average monthly flow rate; 400 m^3 s^{-1}) and Ouvèze (average monthly flow rate; 20 m^3 s^{-1}). These two important surface water bodies result in a great local stability of groundwater levels. The site is equipped with piezometers allowing the monitoring of the water table and of four active pumping wells operating intermittently for a total extraction flow of 11,000 m^3 per day. The exploited unconfined alluvial aquifer corresponds to a plio-quaternary formation mainly composed of sand and pebbles deposited by the Rhône River. The aquifer substratum at the extraction site is composed of Miocene blue marls from between an elevation of 9 and 11m absl(Caridroit, 1968; Nofal, 2014). For the Rhône, the level is controlled upstream at the northwest by the Sauveterre dam which maintains the river stage at about 16.5 m absl.

2.2 Material

The pumping wells (P1, P7A, P3; see Figure 1) were equipped with temperature sensors (HOBO Water Temp Pro v2) in February 2019. The groundwater level of the pumping wells being provided by the pumping wells operator. The accuracy, resolution and response time of the probes are 0.2 °C, 0.02 °C and 5 min, respectively. Four piezometers on the pumping site, and one outside (200 m to the East, RV3 see Figure 1), have been also equipped with pressure and temperature sensors (OTT

Orpheus Mini) with an accuracy of 0.05% for water-level measurement and a resolution of 0.1 °C and an accuracy of 0.5 °C for temperature. The temperature and the river stage of the Rhône were also monitored. All probes have been configured to record measurements every 15 minutes.

Radon-222 sampling was conducted in surface water and groundwater during three campaigns: March 2020, June 2020, and April 2021. The first campaign focused on the Rhône riverbank and the pumping site during a period of pumping shutdown. The second occurred when pumping activity resumed and focused on the groundwater abstraction site. The third campaign focused on the Ouvèze riverbank and the aquifer outside the site in the same hydrodynamic context of the second campaign (pumping wells functioning and related pseudo-steady state). The radon-222 activity concentration of the samples was measured at the laboratory using electronic radon-in-air monitors Durridge RAD-7 coupled with the Durridge RAD H2O system,. The RAD H2O system uses a closed-loop air stripping method to transfer radon from the water sample into a measurement cell, where the radon concentration is then quantified using a RAD7 detector (Schmidt et al., 2010). The monitors count α decays of radon-222progeny, and radon-222 activity concentration is determined by discriminating ^{2}radon-222 progeny decays in specific energy windows. The radon-222 activity concentration in water is then calculated from the activity concentration measured in air with a known temperature correlation (Weigel, 1978). During the campaign additional electric conductivity measurements were made for all radon-222 samples with a multimeter field sensor.

Surface water and groundwater were collected between June 2019 and November 2020 for water stable isotope analysis (^{18}O, ^{2}H). Groundwater samples were collected in the pumping wells and piezometers by means of a submersible groundwater pump and access taps. For the piezometer a pumping time equivalent to the displacement of 3 borehole volumes (calculated using the borehole geometry and the piezometric levels) resulted in a constant value of electric conductivity (measured during sampling with a multimeter field sensor). The composition in $\delta^{18}O$ and $\delta^{2}H$ of the water samples were analyzed using a Picarro L2140-i wavelength-scanned cavity ring-down spectroscopy (CRDS), following the protocols described in Vallet-Coulomb et al. 2021 for post-measurement data processing. All samples were filtered at 0.45µm before analysis and analyzed twice. All data are provided in ‰ *vs* VSMOW, after normalization to the VSMOW2-VSLAP scale. Based on the performances obtained for samples of a laboratory standard included in each analysis runs as a quality assurance / quality control, the precision obtained was 0.03‰ and 0.09‰ for $\delta^{18}O$ and $\delta^{2}H$ respectively (n = 6 runs). The Rhône isotopic signature records were obtained by the Avignon databases (Poulain et al., 2021b, 2021a).

2.3 Methodological approaches

2.3.1 1D temperature analysis

Heat transfer in a half-space (river infinite porous medium) is assumed to be governed by a one-dimensional conduction-advection equation, i.e. considering both the heat transfer due to waterconvection andheat conductionthough the solid and the liquid phases. This equation is usually solved in the framework vertical transfers from the streambed downwards (Hatch et al., 2006; Stallman, 1965) (1).

$$\frac{\partial T}{\partial t} = Ke\frac{\partial^2 T}{\partial z^2} - \frac{nv_f}{\gamma}\frac{\partial T}{\partial z} \quad (1)$$

where T is temperature (°C) variable with time t (s), and depth z (m), Ke is the effective thermal diffusivity ($m^2\ s^{-1}$), $\gamma = \frac{\rho c}{\rho_f c_f}$the ratio of the heat capacity (-) of the streambed to that of the fluid, n is the kinematic porosity (-), and $v_f$is the vertical fluid velocity $m.s^{-1}$.

The heat transfer from the Rhône River to the aquifer was studied by adapting the one-dimensional (1D) solution to calculate the amplitude change of a periodic temperature signal (seasonal signal of the surface water) with the distance to the riverbed and the shift when this signal is propagating within the aquifer (Goto and Matsubayashi, 2009; Stallman, 1965). This analytical solution, in its standard form, assumes 1D uniform vertical flow, sinusoidal behavior of the surface temperature, and no change in the average temperature with depth of the response time to the temperature variation at the surface(Hatch et al., 2006). However, applying this solution can become challenging in the presence of complex thermal signals in the river or non-constant flow rates. In such cases, the best alternatives are numerical models based on finite differences or finite elements schemesthat simulate heat transport in porous media. These models, although more sophisticated, are less accessible due to their complexity and the need for specialized expertise in numerical modeling.(Munz and Schmidt, 2017). Here, the solutions of Goto (2005) and Stallman (1965) have been adapted to study the attenuation and phase shift in the case of a purely horizontal (x-axis) transfer from a riverbank and writes (2). An inversion using the temperature records of the piezometers allows quantification of two parameters: the flow velocity and the effective thermal diffusivityof the alluvial aquiferusing the following expression:

$$T(x,t) = \mathrm{A}\exp\left(\frac{vx}{2Ke} - \frac{x}{2Ke}\sqrt{\frac{\alpha+v^2}{2}}\right)cos\left(\frac{2\pi t}{P} - \frac{x}{2Ke}\sqrt{\frac{\alpha-v^2}{2}}\right) \quad (2)$$

where A is the amplitude of the temperature variations (-), P is the period of temperature variations (s), v is the horizontal velocity ($m.s^{-1}$)and $\alpha = \sqrt{v^4 + \left(\frac{8\pi Ke}{P}\right)^2}$

2.3.2 1D Stable isotope transport

The lateral transfer of surface water into the alluvial aquifer can also be studied using the periodic signal of the stable isotopes of water. For this purpose, the sinusoidal fluctuations of the isotope concentration can be studied by adapting to mass transport the previous solution for heat transfer. By parameter identification of the 1D dispersion advection equation (3) which writes:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - v_f\frac{\partial T}{\partial x} \quad (3)$$

and heat transfer equation (1), the analytical equation (2) was transposed to a mass transport problem giving (4):

$$C(x,t) = \mathrm{A}\exp\left(\frac{\frac{v_f}{n}x}{2D} - \frac{x}{2D}\sqrt{\frac{\alpha+\frac{{v_f}^2}{n}}{2}}\right)cos\left(\frac{2\pi t}{P} - \frac{x}{2D}\sqrt{\frac{\alpha-\frac{{v_f}^2}{n}}{2}}\right) \quad (4)$$

Eq. (4) was applied on the stable of water monitoring. Using the Darcy velocity obtained from temperature signal interpretation (eq. 2), eq. 4 allows identifying the dispersion coefficients (D).

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2.3.2 Coupled Flow and Radon-222 Transport Model

In order to integrate piezometric and radon-222 data to estimate the site's vulnerability to contamination and quantify sources, a model was built using MODFLOW. The numerical codes used for groundwater flow and transport modeling are MODFLOW (Mcdonald and Harbaugh, 1988) for groundwater flow and MT3DMS (Zheng and Wang, 1999) for advective-dispersive
250 transport integrated into the PROCESSING MODFLOW X software. MODFLOW solves the diffusivity equation using a finite difference method. Thus, for horizontal 2D flows in a free, heterogeneous and isotropic aquifer, the diffusivity equation writes:

$$\omega_s \frac{\partial h}{\partial t} + Q(x,y) = \frac{\partial}{x}\left(T(x,y)\frac{\partial h}{\partial x}\right) + \frac{\partial h}{\partial y}\left(T(x,y)\frac{\partial h}{\partial y}\right) \quad (5)$$

With ω_s (-) the specific yield, h (m) the hydraulic head, $T(x, y)$ ($m^2\ s^{-1}$) the aquifer transmissivity, $Q(x, y)$ ($m\ s^{-1}$) source or sink term.

255 MT3DMS was used for radon-222 transport calculations. MT3DMS allows the simulation of dissolved species concentrations by considering advection, dispersion, molecular diffusion, and chemical reaction (Zheng et al., 2012; Zheng and Wang, 1999). MT3DMS thus relies on the flow solutions of MODFLOW to solve the advection-dispersion equation:

$$\frac{\partial(\theta C)}{\partial t} = \frac{\partial}{\partial x_i}\left(\theta x_{ij}\frac{\theta C}{\partial x_j}\right) - \theta v_i \frac{\theta C}{\partial x_i} + q_s C_s + \sum R_n \quad (6)$$

where C ($kg\ m^{-3}$) the concentration of the dissolved species, θ (-) the aquifer porosity, x_i (m) the distance belong an axe,
260 D_{ij} ($m^2\ s^{-1}$) the dispersion coefficient, v ($m\ s^{-1}$) the pore velocity, q_s(s^{-1}) the volumetric flow rate per unit volume of the source or sink, ΣR_n ($g\ m^3\ s^{-1}$) the chemical reactions term.

The modeled surface area is 3 km^2 and covers the Jouve site and the two rivers, the Rhône and the Ouvèze. The boundary conditions (BC) of the model are prescribed hydraulic head conditions: the Rhône at the western limit set at 16.5 m, the Ouvèze at the east set at 18.2 m and a northern limit at 17 m, considered time invariant according to the piezometric monitoring of the
265 pumping field operator. The northern BC is set at 17 m by analysis of previous operator piezometric maps indicating a zone around 17 m due to the influence of the Rhône and Ouvèze rivers on the alluvial groundwater table. Unfortunately, there is no geological boundary in the vicinity that could be used as an alternative BC. The model is composed of 2600 rows and 2200 columns with 5m wide grid cells. The model includes one aquifer layer limited by a lower surface corresponding to the alluvial substratum and an upper surface corresponding to the topography. An annual recharge of 300 mm years by direct rainwater
270 infiltration was introduced based on previous estimates in the Avignon region (Nofal, 2014). The hydraulic conductivity field is considered heterogeneous, so the study area was divided into 10 zones, each with a constant hydraulic conductivityto becalibrate. The calibration to adjust the hydraulic conductivity field of the model to reproduce the measured piezometric levels (10 observations points) was performed at steady-state using the PEST suit, a standard gradient-based optimization algorithm (Doherty, 2004). Initial hydraulic conductivity values were obtained by interpolation using data from the operator's

hydraulic test reports (value of 1.10^{-2} to 1.10^{-4} m s^{-1}). Radon-222 transport was integrated into the model by including the radioactive half-life of radon-222 (3,823 days) in MT3D and by setting the rivers at a constant concentration of radon-222 measured (0.2 Bq L^{-1}). This value of 0.2 Bq. L^{-1} is consistent with the measurement campaign with radon-222 activity concentration in both rivers ranging between 125 and 225 Bq/m³ . A dispersivity of 10 meters was used based on usually found values in the literature for sandy alluvial aquifers at this scale (Schulze-Makuch, 2005). The aquifer radon-222 source term, representing the background radon-222 concentration, was determined by simulating the natural production of radon-222 within the aquifer material. This was conveniently achieved using the flow injection wells package in MODFLOW, where each cell of the aquifer was assigned a low, uniform injection rate (to avoid impacting the piezometric levels)at a constant radon-222 concentration. The concentration of radon-222 injectedwere uniformly applied given the lack of specific information on geological heterogeneities. These parameters were then calibrated using the PEST software to best match the observed radon-222 activity concentrations in the aquifer. This approach allowed to reproduce the natural radon-222 production and ensure that the model reflects the measured radon-222 activity concentration.

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3 Results

3.1 Piezometric level variations

The daily piezometric level variations at the site (Figure 2) are controlled by the pumping cycles of the four wells (P1, P2, P3 and P7A). Over a period of continuous pumping, from January 2019 to February 2020, the pumping maintains the water level between 15.5 m and 14.6 m, except during flooding events of the Rhône River. A pseudo-steady state is therefore established at the site caused by periodic pumping. River stage variations of the Rhône influence the water table, particularly during periods of flooding marked by a sharp increase in the level of the Rhône (Figure 2). When the site is operative, the water table is permanently below the Rhône River stage, even during short pumping stops and floods. The conditions are therefore favorable for an exchange from the Rhône to the alluvial aquifer (Figure 3A). During periods of flooding, the Rhône River rises to an average value of 16.9 m. Higher values were observed in October and December 2019 with sharp increases to 18.9 m and 19.4 m respectively as a result of water releases from the Sauveterre dam (Figure 2). Site maintenance operation led to a prolonged pumping stop (6 weeks during spring 2020, red period, Figure 2). During this period, the water table rose progressively until reaching a stable level above the Rhône River stage, and the exchanging flow between the Rhône and the alluvial aquifer likely reversed. This case corresponds to the natural hydraulic condition leading to a flow from the Ouvèze to the Rhône (Figure 3B). The connection between the alluvial aquifer and the Ouvèze is suggested by the piezometric map but requires further confirmation in the following sections.

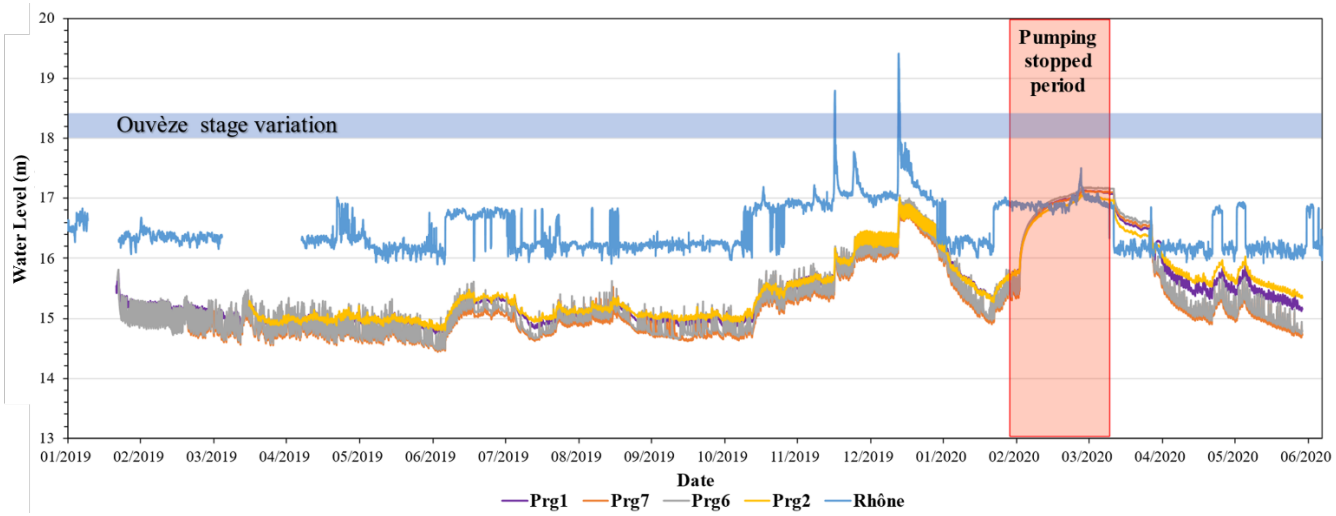

Figure 2 : Piezometric level time series at the study site of la Jouve (Prg1, Prg7, Prg6, Prg2) and Rhône River stage time series at 15 min intervals.

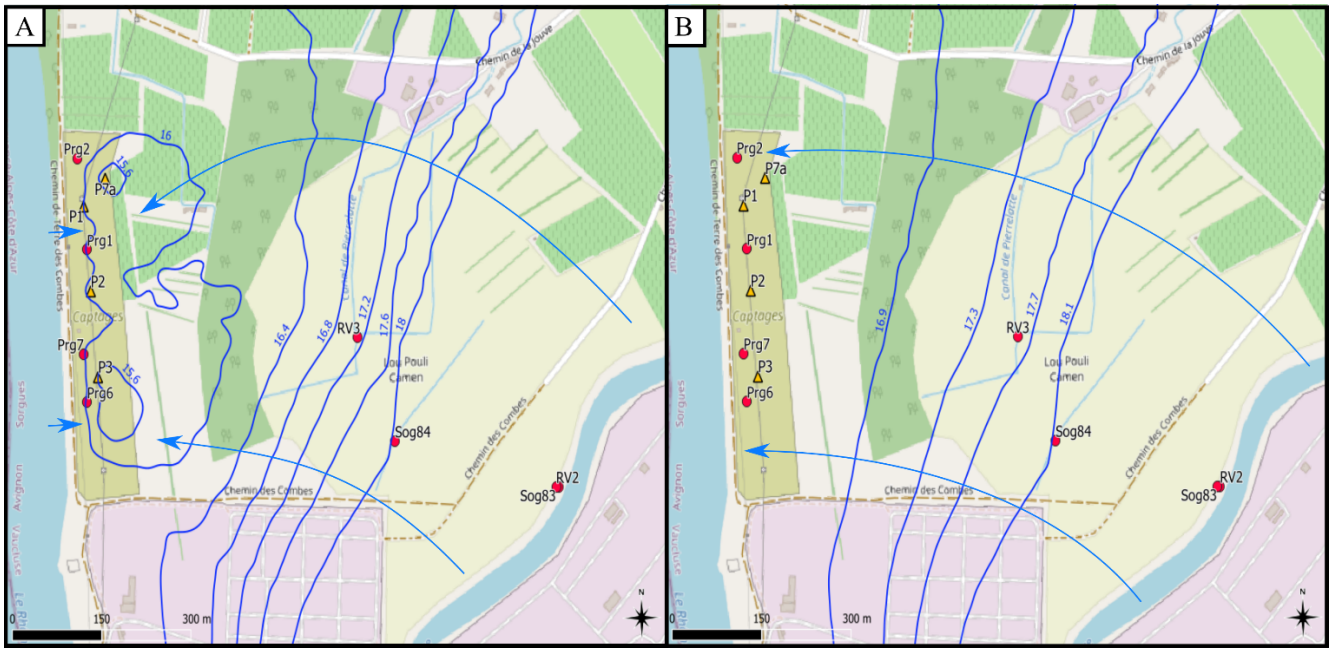

Figure 3 : A) Piezometric map obtained by interpolation of piezometric level using the inverse distance to a power method (campaign during October 2018). B) Piezometric map obtained by interpolation of piezometric level using the inverse distance to a power method (Pumping Stopped March 2020). © OpenStreetMap contributors 2022. Distributed under the Open Data Commons Open Database License (ODbL) v1.0.

3.2 Radon-222 and Electrical conductivity data

During the first field campaign, when the pumping wells were stopped, the radon-222 signature of the Rhône was about 0.2±0.03 Bq $L^{-1}$and at the Prg2 piezometer, the value was at 15±0.35 Bq L^{-1}(Figure 4). During the second campaign, pumping

was restored and the usual piezometric levels were recovered; the same piezometer, Prg2, showed radon-222 activity concentrations four times lower at 3.0±0.24 Bq L^{-1}. The other piezometers, close to the Rhône, showed similar low values, while the pumping wells further away were at values of between 10.0 Bq L^{-1} and 8.0 Bq L^{-1}. These latter values were close to those found during the first campaign at the RV3 piezometer outside the site. During the third campaign, the hydrodynamic conditions on the site were the same as during the second campaign (pumping in operation). The piezometers near the Ouvèze had low radon-222 values at about 3.0±0.12 Bq L^{-1} compared to 11.0±0.24 Bq L^{-1} at a downstream distance of 100 m away from the Ouvèze in the aquifer. During the first and the second campaigns, the radon-222 signatures for the Rhône were similar, and the same signature was observed for the Ouvèze during the first and third campaigns.

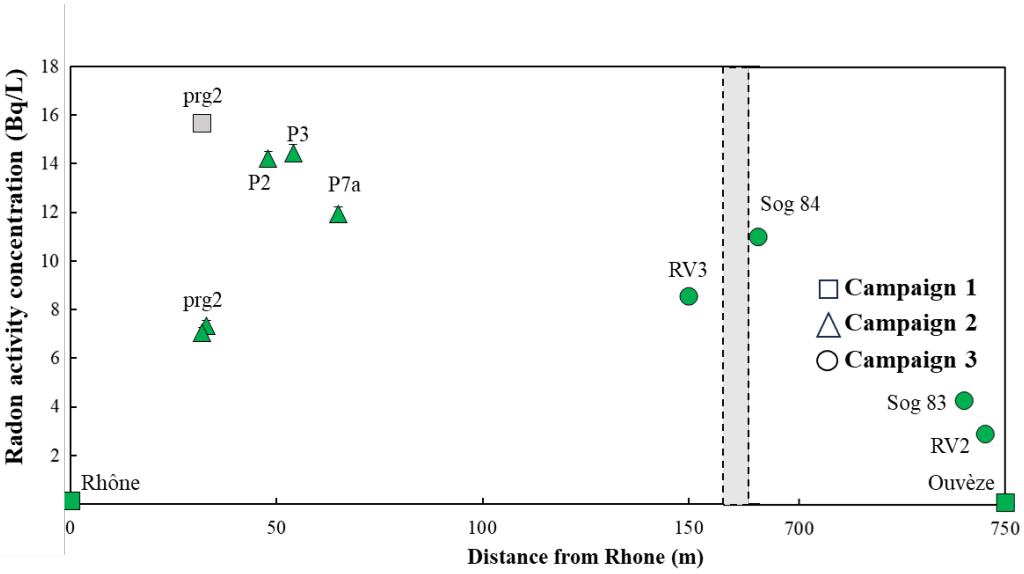

Figure 4 : Radon-222 activity concentration as a function of the distance from the Rhône riverbank (Rhône at 0 m and Ouvèze at 750 m). The colors indicate the pumping state of the site (green for activated pumping, grey for stopped pumping)

In comparison, the electrical conductivity of water samples measured during the second and third campaigns showed a similar behavior as radon-222 during pumping, with almost constant values in the aquifer at about 680 mS cm^{-1}, the Rhône at 350 mS cm^{-1} and the Ouvèze at 570 mS cm^{-1} (Figure 5). The wells and piezometers showed intermediate values between those observed in the aquifer and in the Rhône, with decreasing values for the wells and piezometers closer to the Rhône.

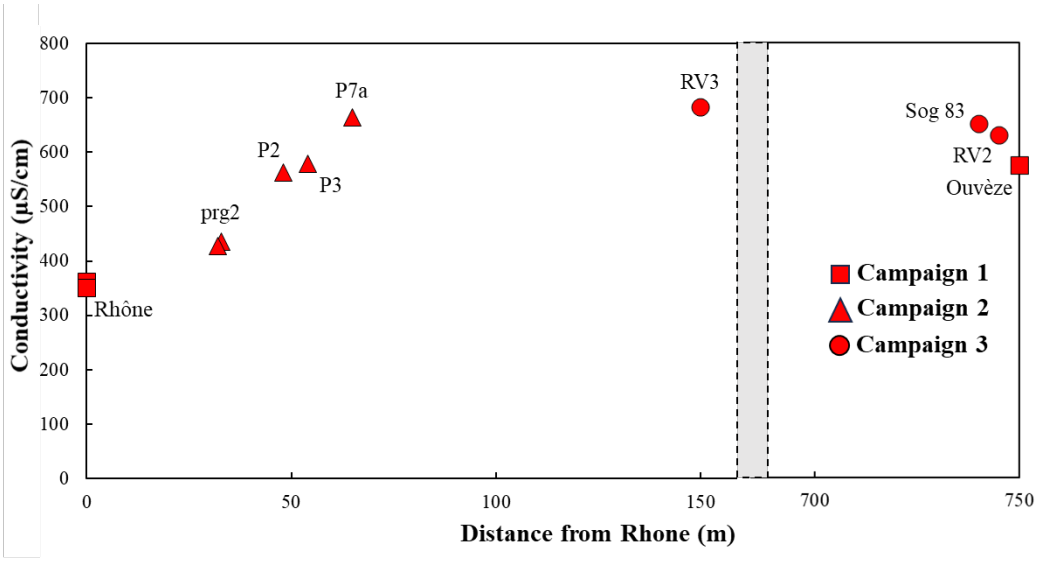

Figure 5 : Electrical conductivity of groundwater and surface water as a function of the distance to the Rhône (Rhône at 0 m and Ouvèze River at 750 m).

3.3 Stable isotopes of water

The two rivers delimiting the study area, the Rhône and the Ouvèze, present depleted isotopic compositions compared to local precipitation, attributed to the altitudes of their respective catchments (Figure 6). The groundwater isotopic compositions are spread around the global meteoric water line (GMWL), in the range of the composition of these two rivers, with a small influence of evaporation for some of the samples. As previously described, the sampling protocols were carefully implemented, and we are confident that the sample compositions were representative of the groundwater composition (sufficient purging phase for piezometers, and a continuous flux for pumped wells). The influence of a direct evaporation of groundwater is possibleparticularly at the riverbanks where the depth approaches zeroand the particularly dry Mediterranean climate of the area (Coudrain-Ribstein et al., 1998). This effect is particularly relevant on the eastern side of the study area near the Ouvèze River, where, even at 150 meters from the river, the water table remains less than 2 meters below the surface, indicating a shallow depth over a significant portion of the area.

Considering the composition of local precipitation, the potential influence of a meteoric groundwater recharge remains undetectable. The piezometers located close to the Rhône (Prg: 42 m, Prg6: 33 m and Prg7: 35 m) present isotopic compositions fully compatible with a lateral recharge coming from the Rhône River, and further modified by the influence of evaporation. The spread of the data can be explained by the seasonal variation of the Rhône composition, and a variable contribution of evaporation.

The distances between the pumping wells and the Rhône River are slightly higher (P1 : 56 m, P2 : 58 m, P7a : 97 m), and their compositions are slightly enriched compared to that of these piezometers. This could be explained by a small contribution of the Ouvèze River, in addition to that of the Rhône. This will be discussed latter.

On the other hand, the composition of the piezometer RV3, located in between the two rivers, points towards a dominant contribution of the Ouvèze River.

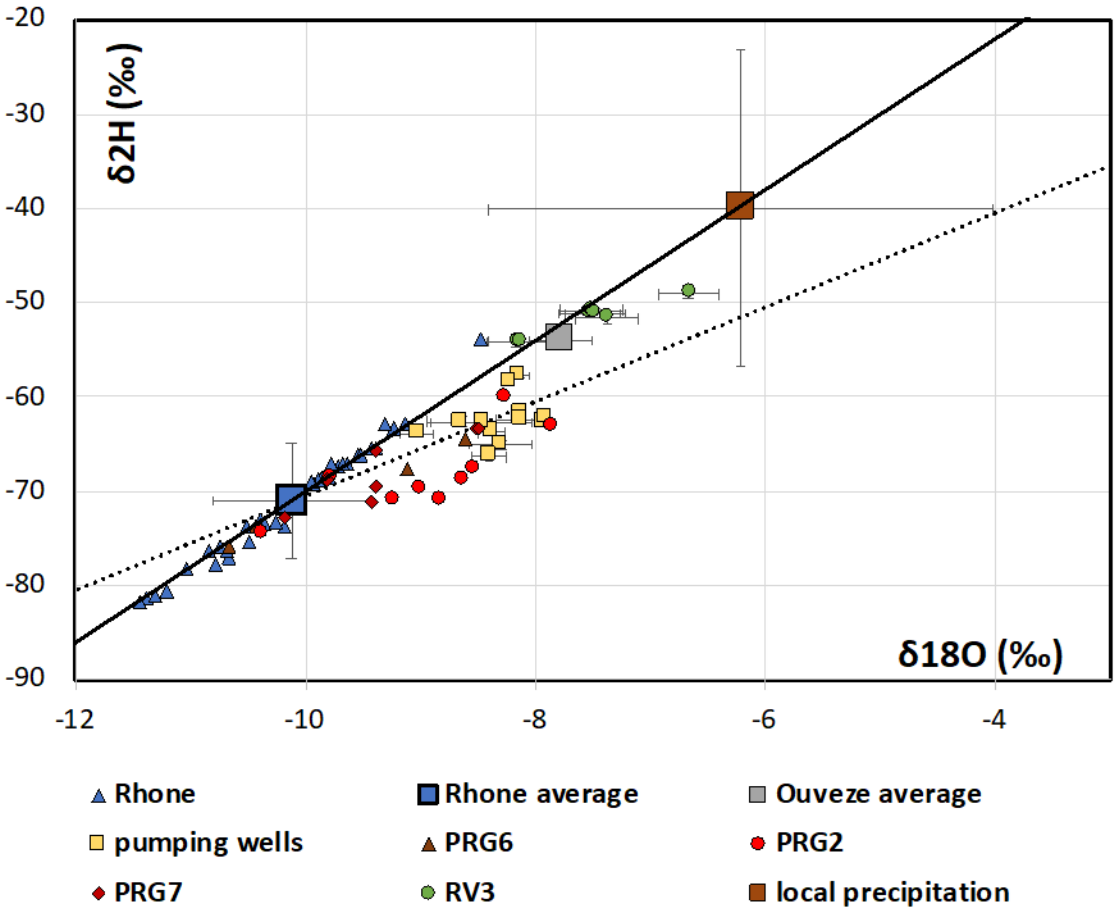

Figure 6 :Isotopic compositions of local precipitation (long term weighted average of the Avignon GNIP station), surface water (Rhône and Ouvèze rivers) and groundwater sampled in piezometers and pumping wells (average values of each sampling campaign). The Rhône's isotopic data are obtained from (Poulain et al., 2021a). Error bars represent the standard deviations of average values. The black line represents the global meteoric water line. The dotted line represents a theoretical evaporation line crossing the Rhône average composition, and assuming a slope of 5.

3.4 Interpretation of Temperature Periodic Signals

Temperature monitoring over the same period showed a periodic variation at all wells and piezometers as well as in the Rhône. The variations in the piezometers showed a significant attenuation as well as a phase shift with respect to the Rhône River temperature signal. During the period when the pumping wells were stopped, discontinuities appeared in the data due to the change in the hydraulic regime. Temperature data for the pumping period can then be modeled using the 1D solutions described in section 2.3 (Figure 7A).

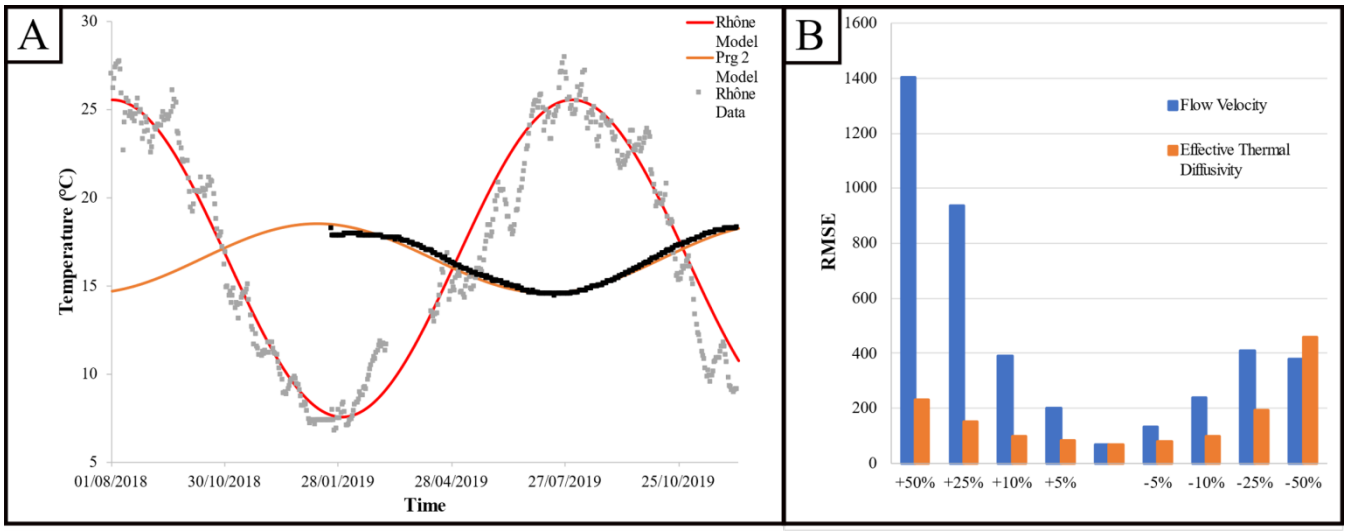

Figure 7 : A) Temperature monitoring of piezometers Prg2 and the Rhône River and corresponding fitted 1D analytical expressions. B) Sensitivity analysis of Prg2 parameters

Upon calibration, the 1D analytical expression for temperature provides the thermal front velocity and by extension the fluid flow velocities. The pore velocity values between 4.7 10^{-6} m s^{-1} and 8.7 10^{-6} m s^{-1} were obtained with a porosity of 10% (value from the operator). The lower velocity was obtained for Prg1 while the three other piezometers led to velocities in the order of 0.8 10^{-5} m s^{-1} (Table 1). The flow velocity appears as the more sensitive parameters, while the thermal diffusivity can vary to 10 % without impact the numerical result (Figure 7B). The flow velocity values are smaller than the first estimate made by calculating Darcy's flux based on the site hydraulic conductivity data (average value of 1.3 10^{-3} m s^{-1}) and the hydraulic head gradient and leading to values ranging from 1.10^{-4} to 3.10^{-4} m s^{-1}. This difference can be explained by the fact that the heat transport accounts for the entire continuum between the river and the aquifer with possible heterogeneity, while the Darcy method considers a homogeneous aquifer. Thus, the lower velocities obtained by this method can be explained by the presence of finer materials along the banks or a partial clogging of the riverbed, reducing the hydraulic conductivity.

Table 1 : Calculated pore fluid velocity and thermal parameters based on the 1D periodic temperature analytical solution, and distance between the piezometers and the Rhône River

Piezometers	*Distance (m)*	*Pore velocity (m s^{-1})*	*Ke(m s^{-1})*
Prg2	42	$8.71*10^{-6}$	$7.29*10^{-7}$
Prg7	35	$6.75*10^{-6}$	$8.44*10^{-7}$
Prg6	33	$8.10*10^{-6}$	$5.13*10^{-7}$
Prg1	45	$4.67*10^{-6}$	$5.34*10^{-7}$

3.5 Fluid flow and transport modeling of the alluvial aquifer at the study site

3.5.1 Fluid flow modeling and calibration

The hydraulic conductivity calibration consists in adjusting the values of the model to reproduce the measured piezometric levels. The PEST package was used to calibrate the model using the piezometric level during the site activity (pseudo-steady state). After calibration the model reproduce the groundwater level (RMSE=0.018 m) and provides hydraulic conductivity values between 1.10^{-2} and 1.10^{-4} m s^{-1} for the aquifer and lower value of 5.10^{-5} for the Rhone riverbank (values consistent with the operator reports).

Modeling under pumping conditions results in a calculated flow from the Ouvèze to the well field, as well as from the Rhône to the well field. In accordance with the piezometric monitoring, the piezometric depression is deeper in the southern part of the site during pumping. When pumping is active, the site is fed on the east side by the aquifer itself fed by the Ouvèze and on the west side directly by the Rhône (Figure 3A). Without pumping, the natural piezometric level is consistent with the piezometric monitoring during the pumping stop period with a reversal of the Rhône-aquifer exchange at the site. In fact, in this configuration, the flow is uniformly directed from the Ouvèze to the Rhône, including at the extraction site (Figure 3B). The calibrated model allows to determine the water budget of the system with 56%of the water exploited at the pumping wells comes from the Ouvèze, 38% from the Rhône, and 6% from the recharge associated with rainfall.

3.5.2 Radon-222 Transport Modelling

Introducing the reactive transport of the radon-222 enables producing a profile of the aquifer from the Rhône to the well. Figure 8 shows that the MT3D transport model accurately reproduced the observations obtained during the different campaigns under the same hydrodynamic conditions, with a gradual increase in radon-222 activity concentration in the aquifer during pumping, due to the progressive increase in radon-222 concentration in the water coming from the Rhône due to gas production within the aquifer. The radon-222 activity concentration increases to a maximum of about 12.0 Bq L^{-1} at about 60 m of the Rhone River.

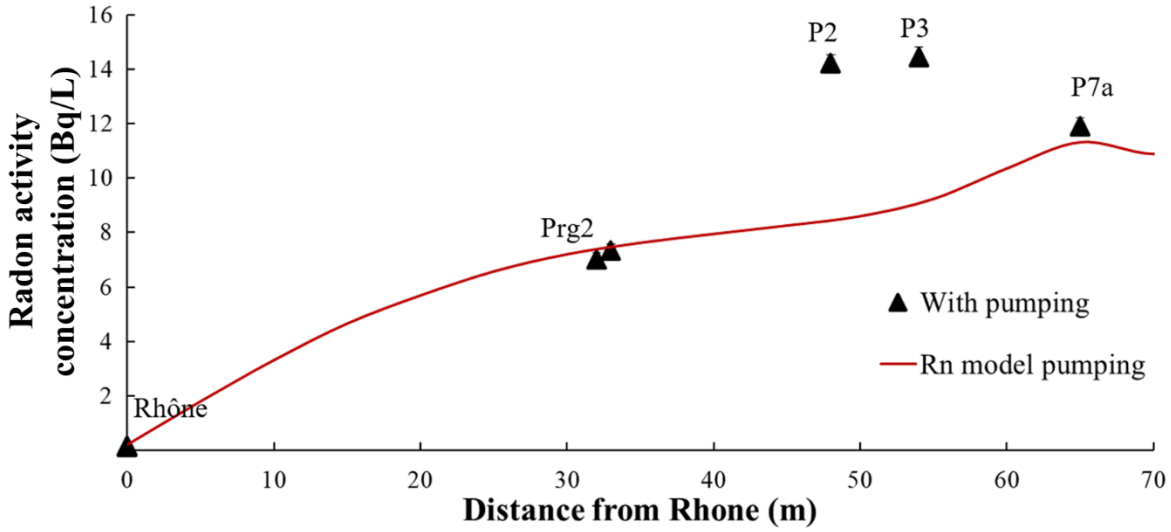

Figure 8 : Comparison between measured and simulated radon-222 activity concentrations in the Rhône River, piezometers and pumping wells in pumping conditions.

4 Discussion

The groundwater is characterized by variable radon-222activity concentrations between 8.0 Bq $L^{-1}$and 15.0 BqL^{-1} which can be explained by some heterogeneity in internal radon-222 production due to petrographic variability. During the pumping period, the radon-222 measurement indicated groundwater recharge from both rivers, i.e., the Rhône and the Ouvèze, thus inducing low radon-222activity concentrations during pumping. These supplies are confirmed by radon-222 in the piezometers near the Rhône and the Ouvèze, even if due to the progressive enrichment of radon-222 in the aquifer, the activity concentration returns to an equilibrium value in the order of 12.0 BqL^{-1} from 50 m downstream. As the wells are located between 50 and 90 meters of Rhône, at this distance the dilution effect of radon-222 activity concentration is no longer visible in the wells, contrary to the piezometers. For the Ouvèze, as for the Rhône riverbank, the lower radon-222 signal caused by the intrusion of the river water with a low radon-222 concentration disappears moving away from the river. We can deduce that the pumping wells producing the piezometric depression have a multiple supply with one part coming from the Rhône River and another one from the groundwater supplied by the Ouvèze. This hypothesis is supported by the piezometric contours that shows a flow from the Ouvèze towards the Rhône either in pumping or no pumping conditions. However, the radon-222 activity concentration does not allow for total quantification of recharge.

Our findings align with the work by Schubert et al. (2006), who also used radon-222 as a tracer to estimate the infiltration of surface waters into aquifers. Their study demonstrated that radon-222 is a valuable tool for identifying the movement of surface water into groundwater systems, particularly when the surface water has significantly lower radon-222 concentrations than groundwater. Similarly, we effectively traced the dilution of the radon-222 activity concentrations in the aquifer due to river water infiltration. The gradual return to equilibrium radon-222 concentrations further supports the idea of progressive radon-

222 enrichment as the river water moves through the aquifer. However, unlike Schubert et al., our study also considers the impact of pumping activities, which introduces additional complexity in interpreting radon-222 signals. This comparison highlights the versatility of radon-222 as a tracer and its applicability in various hydrological settings, including those influenced by anthropogenic activities like pumping.

The dual supply to the pumping wells is well supported by the $\delta^{18}O$-$\delta^{2}H$ data, which indicate a mixture of water from both the Rhône and the Ouvèze. The piezometers near the Rhône exhibit isotopic compositions consistent with lateral recharge from the Rhône, further modified by evaporation.In contrast, the isotopic compositions of the pumping wells are slightly enriched compared to those of these piezometers, suggesting an additional contribution from the Ouvèze. This isfurther confirmed by the isotopiccomposition of the piezometerRV3, located between the two rivers along the flow line from the Ouvèze, which indicates a dominant contribution from the Ouvèze.

In this first approach, the annual recharge due to rain was considered as negligible with a value at 300 mm by year for an area of 3 km² and a total pumping of 12000 $m^3 d^{-1}$. Some piezometer and pumping well data points exhibit signs of evaporation. Water from the wells was collected via an access tap connected to the pumping system, while piezometer water was collected using a pump with a purge corresponding to 3 times the piezometer volume. On the measurement chronicle covering a seasonal cycle, the Rhône does not show an evaporative signal. Considering the estimation of the recharge, this evaporative signaturecould be due only for a small part to the recharge. This evaporative signature is therefore probably produced by direct evaporation from the aquifer in a Mediterranean climatic context, as the water table rises closer to the surface near the rivers, particularly at the riverbanks where the depth approaches zero (Gao et al., 2022). This effect could be more pronounced on the eastern side near the Ouvèze River, where, even at a distance of 150 meters from the river, the water table remains less than 2 meters below the surface.

Although the isotopic phase shift could theoretically be analyzed using Equation (4), the reliability of the seasonal signal in the aquifer is questionable due to evaporation and low sampling frequency. Nevertheless, this method could be generally applicable in alluvial plains under pumping conditions if a seasonal isotopic signal is present in the river. It offers a fast, cost-effective way to assess dispersivity (a weakly constrained variable in hydrogeology which is the dispersivity) using a natural tracer, especially where artificial tracers are not feasible due to the proximity to sensitive sites (industrial, urban).

The water supply to the alluvial aquifer by the Ouvèze, suggested by piezometric data, is then clearly confirmed by radon, isotopic data, with the radon-222 activity concentrations of the piezometers close to the Ouvèze showing a decrease in activity concentration, as for the Rhône, and a similar isotopic composition between the piezometer RV3 located outside the site (to the east) and the Ouvèze.

The use of a simple 1D solution for the temperature record allowed determiningthe flow velocitiesby numerical inversion by reproducing the records. Hence, based on three piezometers the pore velocity was found in the order of 10^{-5} m s^{-1}. After calibration using PEST, constrained by the piezometric levels monitoring, the numerical model reproduced the natural flow, i.e., with and without pumping, with velocities consistent with those deduced from temperature signal interpretation. The transport modeling of the radon-222 supports the feeding of the alluvial aquifer by the Ouvèze, as shown by the intrusion of

radon-depleted water sampled at the piezometers near the river, as well as the pumping of the site by the Rhône. Simulated groundwater balance analysis makes it possible to determine a full quantification of water supplied to the site's pumping systems. Regarding uncertainty, the groundwater balance validated by the Radon transport calculations is obtained by calibrating a steady-state flow simulation. This steady-state calculation depends on hydraulic conductivity and recharge. Modeling uncertainty is primarily caused by the standard equifinality problem i.e., the fact that several permeability-recharge field "pairs" could lead to the same quality of result (agreement between simulated and observed piezometric levels). As recharge represents a small fraction of the groundwater balance (less than 10%), the equifinality problem is very limited here. Therefore, the calibrated permeability field (using measurements as initial guesses) is very robust in a context where the piezometry is largely controlled by the rivers stages used as prescribed boundary conditions. It is therefore difficult to carry out a sensitivity analysis (and therefore an uncertainty analysis) that would consist in varying the permeability field, as this would severely degrade the agreement with the observed piezometric levels. Overall, and despite an unquantified but likely limited model uncertainty, the major part of the water exploited at the pumping wells is sourced from the Ouvèze (around 60%), with a significant portion also coming from the Rhône (around 40%), and a smaller contribution from local recharge associated with rainfall. This calculated groundwater budget is consistent with the isotopic analysis, confirming an important contribution of the Ouvèze. The simulations confirm a clear vulnerability of the site with respect to possible contaminations from the Rhône and the Ouvèze. The Rhône is extremely close to the pumping wells and remains the main threat regarding potential contamination issues.

5 Conclusion

In this study, the ability of a multi-tracer approach to quantify groundwater-surface water exchanges in a riverside pumping context connected to the aquifer was investigated. The objective was to quantify the groundwater-surface water interactions at the local scale using a set of tracers in order to obtain robust information to feed a numerical model. This river-aquifer exchange system was simulated using the MODFLOW model under natural and pumping conditions.

Radon-222 has been used to trace the entry of river water with low radon-222 activity concentration into the aquifer, thus diluting the natural signal of the aquifer. This rarely used method allows tracing the exchange from the river towards the aquifer, whereas radon-222 is mainly used to determine the groundwater discharge into surface water bodies. This method enables identifying the entry of surface water until a distance from the riverbank of about 50 meters.

The 1D transport model, which uses a periodic input signal, allows to analyze the horizontal heat flux in the aquifer.An inversion using the temperature records from the piezometers allows quantificationof the flow velocity and the effective thermal diffusivity of the aquifer. The combination of the piezometric, radon-222 and isotope monitoring showed that both the Rhône and the Ouvèze rivers recharge the aquifer.

The hydrogeological modeling allowed reproducing the set of observations and to precisely determine the water balance at the pumping site with a dominant contribution from the Ouvèze (around 60%) and secondary from the Rhône (around 40%). This

water balance highlights the vulnerability regarding possible pollutants from the two rivers. It is interesting to note that it may appear somewhat counterintuitive that the smaller river makes greater contributions to the aquifer recharge than the larger river, and indeed the water management company held that belief before this study. Despite the lower contribution of the Rhône , the greater proximity represents an important risk due to the shorter time to reach the wells and therefore a lower chance of retention or degradation of pollutants.

Beyond our case study, this study confirms the great interest of the radon-222 tracer in the context of groundwater-surface water exchanges when piezometers close to the riverbank are available, whatever the direction of the exchange. The use of different natural tracers such as temperature, isotopes or other natural tracers allows for a more accurate estimation of the hydraulic parameter and the creation of a more robust model and is especially interesting where the use of artificial tracers is not an option. Integrated tracer-modeling studies can thus help improve the understanding of aquifer functioning and contribute to a sustainable exploitation strategy.

Author contribution

J. Texier prepared the manuscript with contributions from all co-authors. J. Texier perform the MODFLOW model with contributions from J. Gonçalves. Field work and data acquisition performed by all co-authors. Isotopic data analysis performed by J. Texier and C. Vallet-Coulomb. Radon-222 data analysis performed by J. Texier and T. Stieglitz.

Competing interests

The authors declare that they have no conflict of interest.

Acknowledgments

The Agence de l'Eau Rhône-Meditérannée-Corse, the Syndicat Rhône-Ventoux and SUEZ are acknowledged for their financial support. The modeling software use is Processing MODFLOW X ver.

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