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SPECIAL ISSUE

Hydrodynamical changes and their consequences on groundwater hydrochemistry induced by three decades of intense exploitation in a Mediterranean Karst system

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Abstract The Lez karst spring, located in the Mediterranean basin (southern France) supplies drinking water to the metropolitan area of Montpellier (France) since the nineteenth century. Since 1981, an intense pumping is being performed directly in the main conduit with a maximum exploitation flow rate of about 1,700 l/s. To improve the understanding of groundwater origins and circulation dynamic in this karst system, as well as the impact of three decades of intense water exploitation, groundwater samples have been collected during various hydrologic conditions since March 2006. A previous hydrochemical study on the Lez karst system had been carried out before the installation of the pumping plant. This dataset was compared to the present one to identify possible changes on groundwater hydrochemistry. The results of this analysis indicate the existence of historical changes in water hydrogeochemistry and evidence a decrease of the deep compartment participation to the Lez spring outflow. This change in spring water hydrogeochemistry may be attributed to the intense pumping of the karst system and, in the absence of noticeable climatic changes, expresses the direct consequences of anthropogenic forcing on the overall functioning of the aquifer. This study aims to analyze the differences, to understand the water chemistry changes and to better foresee the aquifer evolution for the future.

Keywords Karst · Groundwater · Natural tracing · Hydrochemistry · Water transit

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Introduction

Karst aquifers supply about a quarter of the world population with drinking water. They represent over 35% of the French territory and supply up to 30% of the total potable water (Plagnes and Bakalowicz 2002). Fissured carbonate aquifers host huge resources of high-quality groundwater (groundwater) and optimizing the groundwater protection and management is a priority in both industrialized and developing countries (COST ACTION 65 1995).

The chemistry of karst springs is normally related to the physical characteristics and helps distinguishing different types of hydrodynamical behaviour in the drained karst systems (López-Chicano et al. 2001; Moral et al. 2008; Mudarra and Andreo 2011; Rosenthal et al. 2007; Belkhiri et al. 2011). Hydrodynamical characteristics such as recharge, thickness of infiltration zone, aquifer geometry and its fissured structure can significantly influence the groundwater transit time and water–rock interactions, as well as the hydrochemistry of waters draining a karst (López-Chicano et al. 2001; Jiang et al. 2009; Moore et al. 2009; Stuart et al. 2010).

The Lez spring, the main outlet of the Karst System under study, supplies potable water to the metropolitan area of Montpellier (France) since the nineteenth century. Since 1981, an intense pumping (active management of the aquifer) is being performed directly in the main conduit, from an excavated pumping plant located 48 m below the Lez spring level, with a mean pumping rate of 1,300 l/s and a maximum pumping rate of 1,700 l/s.

The Lez spring has been regularly monitored since 2006 by the Laboratory HydroSciences Montpellier, for measuring chemical and physico-chemical parameters. From this survey, chemical data interpretations allowed a significant progress for the comprehension of water circulation in the Lez aquifer, and a new hydrodynamic conceptual model was proposed for this aquifer (Bicalho et al. 2010, 2011).

The present paper analyses and discusses the changes induced in the chemical composition of water at the Lez spring as a result of intense exploitation since 1981, in comparison with the hydrochemical monitoring achieved in 1973-1974 (Marjolet and Salado 1976), before the intense pumping starts.

Few works are reported in the literature relating to the evolution of the aquifer behaviour before and after several years of intense exploitation. Rosenthal (1988) investigated the hydrochemical changes induced by two decades of intense exploitation of groundwater in a multiple layered aquifer system in Israel, provoking the ascension of brines and the salinization of waters.

The Lez aquifer unifies many advantages that favour the advance of karst hydrogeology science, like a large monitoring network and an important historical dataset during periods with distinct hydrodynamic conditions. Former studies have shown this karst system to be a complex and heterogeneous system in terms of structure, organization, and functioning (Marjolet and Salado 1976; Joseph et al. 1988; Thierry and Bérard 1983; Fleury et al. 2009; Karam 1989). However, after years of scientific investigations on the Lez karst system functioning, the dynamics and origin of groundwater remain relatively unknown.

Materials and methods

Hydrogeological and geological settings

The Lez spring is one of the main karst springs in France and is located 15 km north of Montpellier (France). This outlet is a Vaucluse-type spring, i.e. with a main conduit developed below the spring level, and is located on a major regional fault. This spring (65 m a.s.l.) is the source of the Lez River that flows over 28 km to the Mediterranean Sea. This river has three main tributaries fed by temporary karst springs belonging to the Lez karst system.

The Lez spring is the main perennial outlet of the system, with groundwater discharge that can reach about 15 m^3/s during rainfall events, with an annual average discharge of 2.2 m^3/s. It used to be a perennial spring; however, since 1981 water has been directly withdrawn from the main conduit to supply water to Montpellier and its metropolitan region. The pumping rate sometimes exceeds the natural water discharge in order to secure water supply throughout the year. Consequently, during low-water period, the spring dries up. Ecological water discharge (160 l/s) at the Lez River is ensured during this period by a partial deviation of the pumped water to the river.

The hydrogeological basin of the Lez spring has an area of about 380 km^2 (Thierry and Bérard 1983), and is located between the Hérault and Virdoule river valleys (Fig. 1). As a large part of the hydrogeological basin is relatively impermeable, due to the presence of marls and marly-limestones of the Valanginian, the Lez spring diffuse recharge area covers only 150 km^2 (Marjolet and Salado 1976). The Lez karst system discharges also at several seasonal outlets, Lirou, Restinclières, Fleurettes and Gour Noir (Fig. 1).

The diffuse recharge area over the basin corresponds to the Jurassic limestone outcrops located by the western and northern limits of the basin. Localized infiltration occurs through fractures and sinkholes along the basin and through the major geological fault of *Corconne-Les Matelles*, in the northern part of the basin. A certain number of fractures are also known to exist only just upstream from the Lez spring (Dubois 1964).

The cross section of the terminal conduit that reaches the Lez spring is greater than 10 m^2. The conduits dimensions result from an intense karstification that reached several hundred of meters during the Messinian Salinity Crisis (Joseph et al. 1988) and affected the limestone formations during the Plio-quaternary.

The lithology of the Lez karst system corresponds to massive limestone of the Upper Jurassic (Argovian to Kimmeridgian) and of the lower part of the Early Cretaceous (Berriasian), with 650–1100 m thickness. The marls and marly-limestone of the Middle Jurassic (Oxfordian) constitute the lower boundary of the aquifer. The marls and marly-limestone of the Early Cretaceous (respectively, Lower and Upper Valanginian) constitute the upper boundary of the aquifer. The major tectonic events that influenced the Lez aquifer were the Hercynian/Variscan orogeny, the Pyrenees formation, and the opening of the Lion Golf (Bousquet 1997). Accordingly, the Lez karst system is referred to as a partly confined system.

Hydrological settings

The average annual rainfall calculated over the last 40 years at Valflaunès raingauge (Météo France) was 942 mm (Fig. 2). The total annual rainfall is highly variable from year to year: 1989 was the driest year during this period, with 474 mm, and 1972 was the wettest year, with 1,620 mm of rainfall. During the studied period, the annual rainfalls were 810 mm for 2006–2007, 849 mm for 2007–2008, 1,266 mm for 2008–2009 and 666 mm for 2009–2010. For the 1970–2010 period, the intra-annual rainfall distribution was: 37% during autumn, 27% during winter, 22% during spring and 13% during summer. The first recharge events (rainfall events) of the hydrological cycle (or hydrological year: from September to September) start normally between September and October.

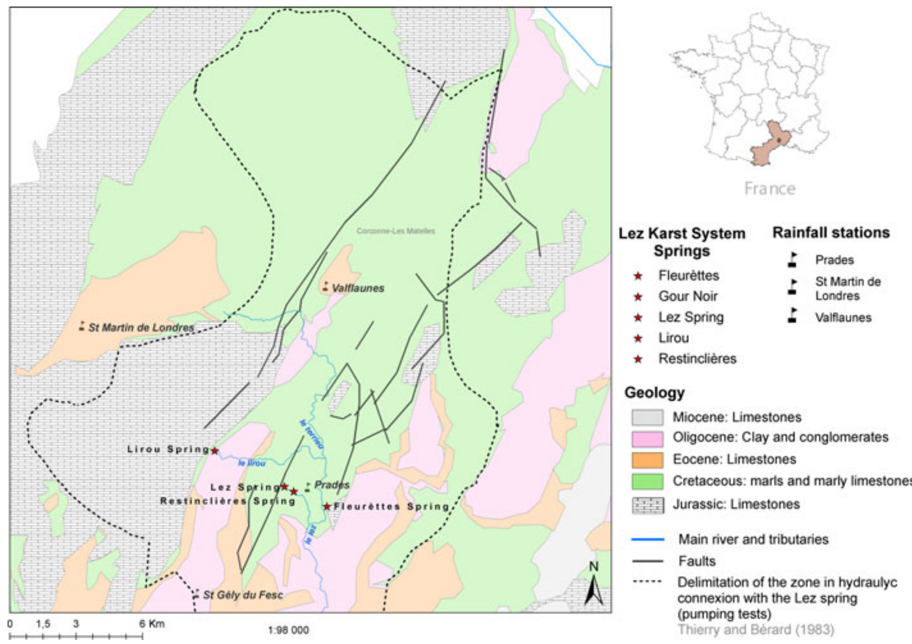

Fig. 1 Hydrogeological map of the Lez karst system with sampled springs, wells and rain gauges location

Sample collection and analysis

Data were obtained by regular samplings achieved by HydroSciences Montpellier (HSM) from 2006 to 2010. Temperature (T), turbidity, Electrical Conductivity (EC $T_{ref} = 25°C$) and groundwater level were monitored using an hourly time-step at the Lez spring with an automatic datalogger (CTD diver, SDEC), at the observation well of Veolia pumping station (Lez well), located in the proximity of the Lez spring. HSM performed T, EC, and groundwater level measurements with an hourly time-step at the Lez spring spillway and at the Lirou spring (CTD diver, SDEC). Rainfall data were obtained from three meteorological stations of Météo France: Valflaunès, Saint-Martin-de-Londres, and Prades (Fig. 1).

Samples were collected from the Lez spring twice a month, from March 2006 to May 2010, and daily during the floods that occurred within this period. Temperature, pH and EC were measured in the field using a pH meter and conductimeter (WTW 330 i). Total alkalinity was measured within a day, by acid titration with HCl 0.05 N. Chemical and bacteriological analyses were performed at the laboratory. Major elements (Br^-, Cl^-, NO_3^-, SO_4^{2-}, Ca^{2+}, Mg^{2+}, Na^+, and K^+) were analyzed by ionic chromatography (DIONEX® ICS 1000) in filtered samples (0.22 μm) offering an analytical precision with a margin of error of less than 5%. Trace elements (Li, B, Al, V, Cr, Mn, Co, Ni, Cu, Zn, As, Rb, Sr, Mo, Cd, Ba, Pb and U) were analyzed using Q-ICPMS X series II Thermo Fisher® at the AETE technical platform of University of Montpellier 2 in

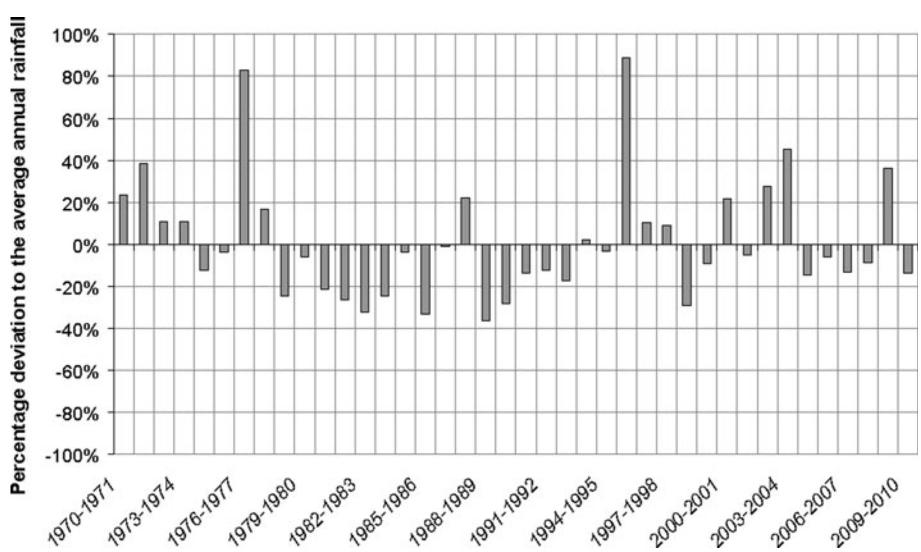

Fig. 2 Percentage deviation to the average annual rainfall calculated from 1970 to 2010 (Valflaunès raingauge)

filtered samples (0.22 μm), offering an analytical precision with a margin of error of less than 8%.

Marjolet and Salado (1976) performed sampling on the Lez spring from March 1973 to May 1974, with a 5–10 days interval during low stage and daily samplings during floods. T was measured in the field; pH and EC were measured in the laboratory. The chemical analyses were performed at the Service of Water Analysis of CERGA (Centre d'Etudes et de Recherches sur la Géologie et ses Applications). Ca^{2+} and Mg^{2+} were measured by complexometry with EDTA (±1%). Total alkalinity was measured by acid titration with H_2SO_4, N/10, (±1%). Sulphates were measured by nephelometry after addition of $BaCl_2$, (±3–5%). Na^+ and K^+ were measured by flame photometry, (Na: ±1–2%; K: 10%). Cl was measured by the Mohr method, by addition of $AgNO_3$, (±1–2%).

Data treatment analytical methods

Discriminant Factorial Analysis (DFA) was used to interpret geochemical data sets. The multivariate statistical technique identifies individual groups of samples and tests the membership of each sample to a defined group, as a function of the values for a number of considered variables. It minimizes intra-group variance and maximizes intergroup variance (Dagnelie 1975; Saporta 1990). The reduced dimension obtained by such a statistical technique permitted reaching a simplified multi-criteria representation, highlighting the main trends for the datasets. Aqueous speciation and fluid-mineral equilibrium were calculated with Diagrams software, version 5.0 (Simler 2009).

Results and discussion

Characterization of the different groundwater types flowing at the Lez spring

DFAs were performed with the data set in order to identify the water types flowing at the Lez spring and for interpreting their chemical characteristics (Fig. 3). The explicative or independent variables assumed were EC, major ions (Cl^-, Mg^{2+}, K^+, SO_4^{2-}, HCO_3^-, Na^+ and Ca^{2+}), trace elements (Li, B, Rb, Ba and Sr), pCO_2 and calcite SI. Five water-types were identified: (1) "*High waters*", that discharge during high stage period and are associated to high groundwater level and EC oscillations; (2) "*Low waters*", that discharge during low stage period and are associated to low groundwater level and stable and high EC; (3) "*Dropping waters*", that discharge during the transition between high and low stage period and are associated to groundwater level decrease and EC increase. Extremely high or low EC values are referred to as: (4)"*Piston-flow waters*" (EC > 780 μS/cm) and (5)"*Dilution waters*" (EC < 600 μS/cm) (Bicalho et al. under review; Bicalho et al. 2010).

The F1-axis for the first DFA (major elements) represents the mineralization with Cl, Na and Mg. The F2-axis is defined positively by Ca and partially by SO_4 (Fig. 3). "*Piston-flow waters*" and "*Low waters*" present a good discrimination. The second DFA (major and trace elements) shows a better discrimination between the five groundwater-types. A remaining zone of overlapping between "*High waters*" and "*Dropping waters*" is still observed. The F1-axis is positively weighted by EC, Cl, Na, Mg, Ba, K, Rb and Li. The F2-axis is negatively weighted by Sr and SO_4. The trace elements help to better characterize the "*Piston-flow waters*", associated with a marked mineralization in Cl, Na, Li, Mg, Cr, Rb and Ba. The "*Low waters*" are the second high-mineralized waters compared to those elements, but SO_4 and Sr concentrations are lower than in "*Piston-flow waters*". The "*Dilution waters*" remain the less mineralized waters.

Lez typical autumn floods: 1973 and 2008

The first comparing analysis between the current chemical characteristics of the Lez spring and the ones reported 35 years ago is focused on the autumn floods (Fig. 4). These floods, occurring at the beginning of the rainy season, have been observed each year and showed only variations in intensity (EC values and element concentrations peaks). The cumulated rainfalls that triggered those floods were, respectively, 86 mm in 1973 and 200 mm in 2008. In terms of annual rainfall, both 1973 and 2008 have rainfall surplus: about 10% in 1973 and about 35% in 2008. The maximum observed Cl concentrations were, respectively: 112 mg/l in 1973 and 80 mg/l in 2008.

The rainfall recharge causes a transient pressure pulse to travel the system resulting in a discharge spike at the spring (Desmarais and Rojstaczer 2002). Indeed, intense rainfall recharge causes the motion of more mineralized waters towards the spring, due to a piston flow effect. This phenomenon happens only during extreme hydrological situations and has been also observed in some karst systems (López-Chicano et al. 2001; Emblanch et al. 1999; Desmarais and Rojstaczer 2002).

In Oct. 1973, the historic-highest Cl concentration (120 mg/l) ever measured was observed at the Lez spring, during a rainfall event of 88 mm. This flood event was triggered by the smallest rainfall event in terms of height between the three analysed floods. Nevertheless, considering that there was no intense pumping in 1973, the total water-head on the aquifer was considerably higher, in spite of the smaller height of the rainfall event. Therefore, the

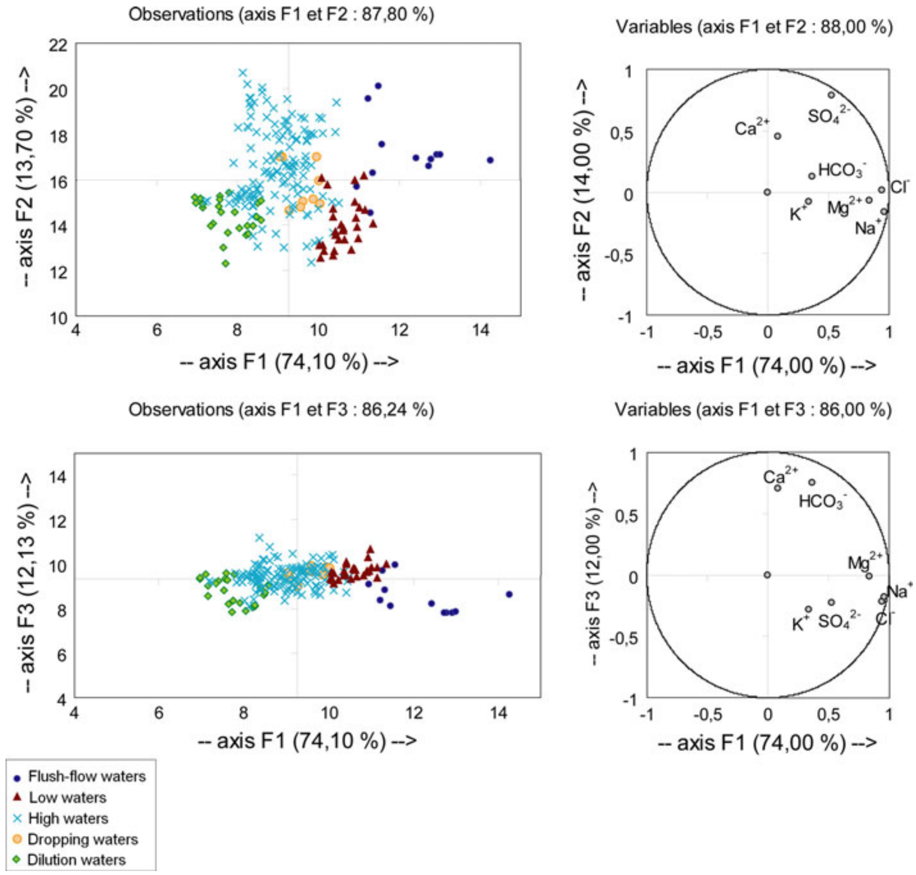

Fig. 3 *Top* DFA for the Lez spring data using major elements. *Left* sample space; *right* variable space. *Bottom* DFA of the Lez spring data using major and trace elements. *Left* sample space; *right* variable space (Bicalho et al. under review; Bicalho et al. 2010)

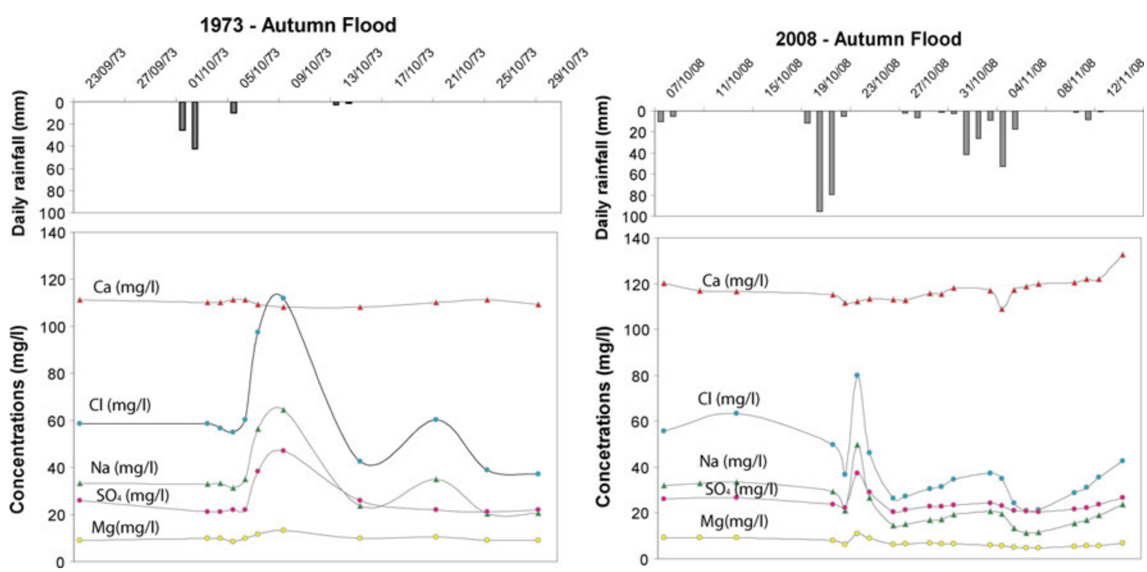

Fig. 4 First autumn flood events of the years 1973 and 2008, considering variations for: Ca, Mg, Na, K, Cl and SO_4. Cl maxima were, respectively, 112 mg/l in 1973, 80 mg/l in 2008 and 98 mg/l in 2009

saturation state in the aquifer was more easily reached in 1973 than in 2008 or 2009. Nowadays, the first rainfall events recharge the karst aquifer not only after a long period of dryness but also of intense pumping. At present, the karst aquifer is more hydraulically depleted and needs greater amounts of rainfall than it used to before the beginning of the intense exploitation, to reach the same piezometric levels.

Chemical changing description using Discriminant Factorial Analysis

In order to search for evidences of changes on water chemistry before and after intense pumping, the hydrological cycles of 1973–1974, 2006–2007, 2007–2008 and 2008–2009 were analysed using DFA. Two distinct DFA were carried out, both considering EC, pCO_2 and concentrations in Cl^-, Mg^{2+}, K^+, SO_4^{2-}, HCO_3^-, Na^+ and Ca^{2+} as independent variables. The dependent variable for the first DFA was the different hydrological cycles. Observation plots on axes F1 and F2, which represent 97.53% of the total variability, individualize the water year 1973–1974 from the other years (Fig. 5).

The negative direction of F1 axis represents mineralization with HCO_3 and Ca which is the predominant mineralization for the 2006–2007, 2007–2008 and 2008–2009 waters. The positive direction of F1 axis represents the mineralization with K, Na and Cl that characterizes the 1973–1974 waters. F2 axis is characterized by Mg concentrations and pCO_2 inversely related.

For the second DFA (Fig. 6), the water-type identification used for the current Lez waters was applied to the samples of 1973–1974 (Bicalho et al. 2009; Bicalho et al. 2010). The "water-year-type" was thus considered like the dependent variable for the calculation of the additional DFA. The global difference observed is that the whole system seems to have moved toward a new geochemical composition (Fig. 6). The most mineralized waters, before and after intense pumping, are still the *Piston-flow waters*, which flow during the autumn floods. However, this water-type for "after pumping" conditions shows lower concentrations in Cl, Na, Mg, K and SO_4 and presents higher concentrations in Ca and HCO_3 than "before pumping" conditions.

Conceptual model of a possible hydrodynamical change before and after pumping

The hydrological conditions determine the different compartments participation to the mixing that composes the waters of the Lez spring outflow (Bicalho et al. under review). The intense exploitation causes a general decrease of the hydraulic head within the aquifer which does not reach any longer the hydraulic head that it used to, even for similar rainfall recharges. The participation of the waters from the deep compartment to the Lez spring outflow is related to the occurrence of strong hydraulic head during the heavy autumn rainfall recharges (Bicalho et al. under review; Bicalho et al. 2010). Indeed, intense pumping apparently led to the decrease of the deep compartment participation to the Lez spring water flow.

Figure 7 presents an adaptation of the conceptual model of Bicalho et al. (2011) containing the hypothetical hydrodynamical differences that characterize the system for both years 1973 and 2009. The average concentration variations were calculated according to the average values of concentrations in Ca and Cl for low stage waters and high-mineralized waters (piston-flow waters) for both datasets (before and after pumping).

The origins of waters marked by a Ca–HCO_3 mineralization are associated to the main and shallow aquifers (Jurassic, Berriasian and Valanginian layers) (Marjolet and Salado 1976; Bicalho et al. under review) while the origins of Cl, Na, Mg and SO_4 are mainly associated to deep layer with evaporitic fingerprinting (Bicalho et al. under review). The intense exploitation generated a new hydrodynamical equilibrium which induced a different water chemical composition, by changing the participations of the different compartments to the spring waters mixing, according to the hydrological conditions. Some of the most visible changes

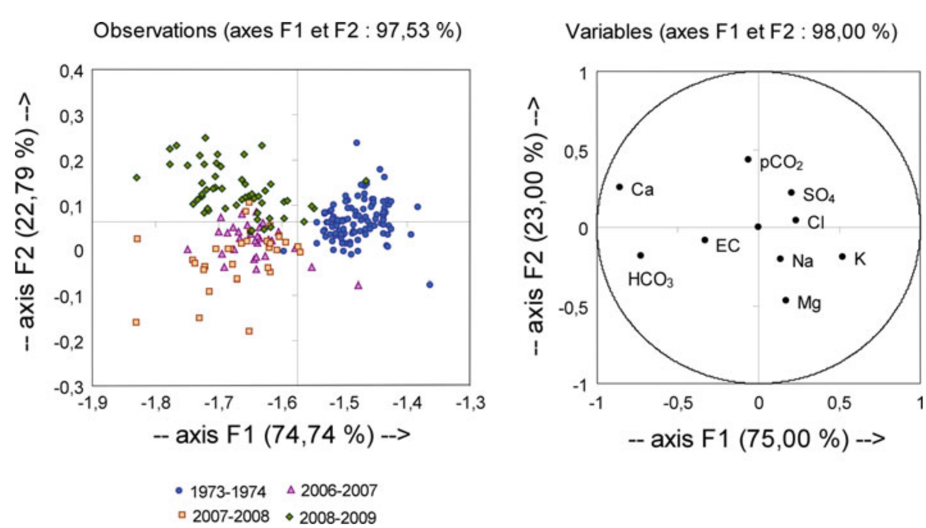

Fig. 5 DFA for the Lez spring data for the years: 1973–1974, 2006–2007, 2007–2008 and 2008–2009. *Left* sample space; *right* variable space

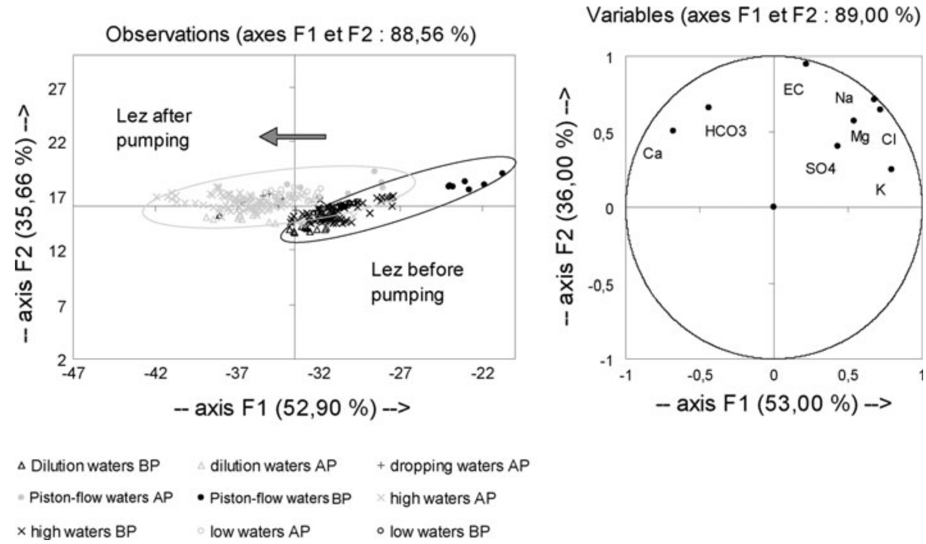

Fig. 6 DFA for the Lez spring data: Low water Before Pumping (BP), piston-flow water BP, Dilution waters BP, High waters BP, Low waters (2008), Piston-flow waters, Dilution waters, Dropping waters and High waters. *Left* sample space; *right* variable space

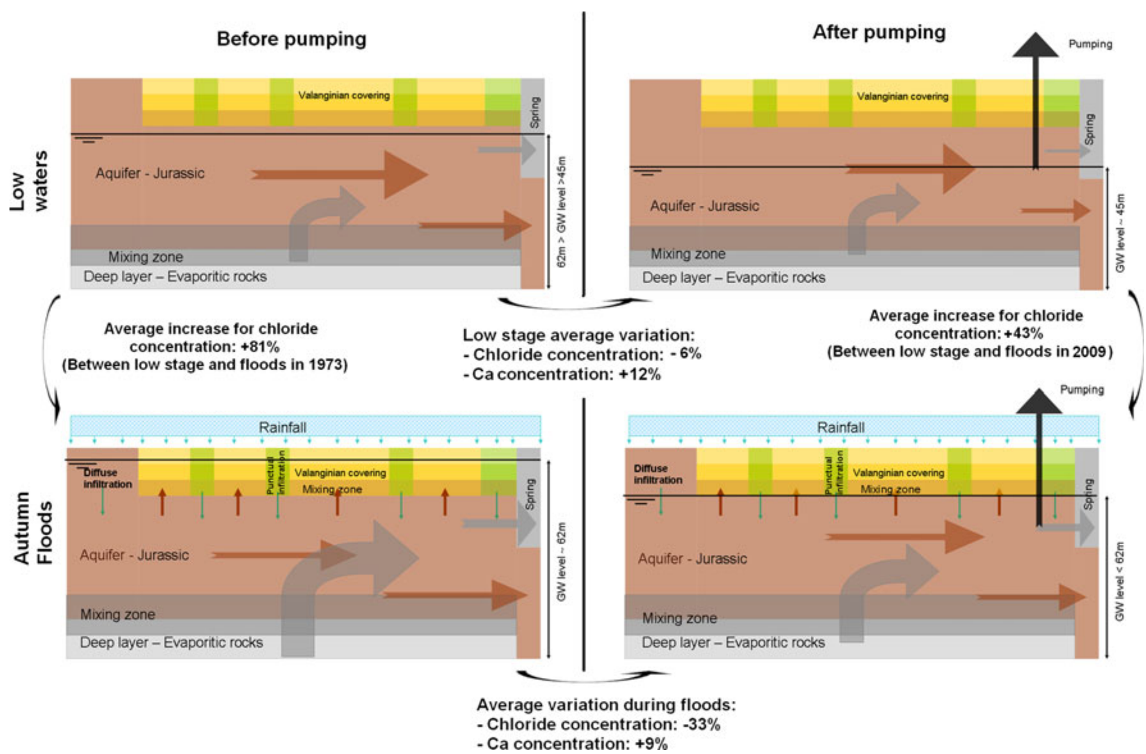

Fig. 7 *Left* Conceptual model of groundwater circulation within the Lez aquifer before pumping. *Right* Conceptual model of groundwater circulation within the Lez aquifer after pumping

on water chemistry are: Na–Cl mineralization decrease and Ca–HCO_3 mineralization increase, after the intense pumping started.

The average chloride concentration decrease during autumn floods is about 33% between 1973 and 2009. During the low stage season, Cl concentration differences between both scenarios are less pronounced. However, higher concentrations for Ca and HCO_3 show that the participation of those waters increased after the exploitation has started. The increased participation of the shallow aquifer waters to the spring flow was already suspected by Marjolet and Salado (1976).

The waters from 2006 to 2009 presented a higher pCO_2 than in 1973–1974, potentially leading to a more important karstification within the aquifer which could, in the long term, cause an augmentation of conduits diameter in the carbonate matrix. Under-saturated waters are observed when recharge is intense and when a very rapid draining of recently infiltrated waters takes place (López-Chicano et al. 2001). This could indicate that water circulation "after-pumping" happens in compartments rather shallower than "before-pumping", due to the decrease of the water residence time in the aquifer.

Conclusion

The distinct chemical characteristics, observed for each water type flowing at the Lez spring, suggest the probable occurrence of multiple origins and different lithologies influence on the groundwater mixing. High and low mineralized groundwaters seem to be, respectively, related to deep groundwater rising and to superficial infiltration waters. The hydrological conditions induce different proportions of the end-member participation on groundwater mixing of the Lez spring.

Despite differences between analytical methods, it is obvious that a significant chemical evolution of water appears after the beginning of the intense exploitation of the Lez spring. The global decrease of the total hydraulic head within the aquifer, especially in the proximity of the Lez spring, has modified the general groundwater circulation in the aquifer. This consequently changed the groundwater mixing induced by the participation of different compartments to the spring flow.

Apparently, the intense pumping mobilizes water from shallower levels in the aquifer, in flow paths probably related to shorter residence times, which has a direct influence over the aquifer vulnerability. The current higher global pCO_2 levels observed in the spring waters induce a Ca–HCO_3 mineralization increase, and could indicate that karstification processes is today more intense than it was before the intense aquifer exploitation has started. This could, in the long term, cause an augmentation of conduits diameter in the carbonate matrix.

The intense pumping has probably led to a decrease of the deep compartment participation to the Lez spring water flow; as a result, Cl concentration has decreased since the exploitation has started. Those results highlight the direct consequences of anthropogenic forcing on the overall functioning of the aquifer.

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