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## Climate change impact on the mineralization of karst groundwater in a Mediterranean context

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### Abstract

The aim of the current study is to investigate the geochemical evolution of groundwater mineralisation and the inter-connexion between shallow karst aquifer and deep aquifer groundwater. The studied aquifer system is considered as the principal outlet of the south-eastern end of the northern Pyrenean overthrust, with a volumetric flow reaching 2000 m<sup>3</sup>/h. Water at the emergence is characterized by high mineralisation and temperature relative to groundwater at the regional scale. An eight years survey was carried out at the outlet based on a geochemical and isotopic approach. Surprisingly spring water mineralisation increases with increasing precipitations. <sup>87</sup>Sr/<sup>86</sup>Sr ratios of sampled water range from 0.7085 to 0.7090. This ratio contrasts with that of the aquifer matrix constituted by Mesozoic marine carbonates and underlying evaporites. Furthermore, these ratios are significantly higher than in groundwater impacted by current or ancient seawater at the regional scale. These high <sup>87</sup>Sr/<sup>86</sup>Sr ratios may reflect the dissolution of <sup>87</sup>Sr rich minerals such as the Triassic salty clay deposits present at depth. The good correlation between <sup>87</sup>Sr/<sup>86</sup>Sr and TDS confirm the hypothesis of a contribution of deep saline and <sup>87</sup>Sr rich water. The temporal variations in mineralization and Sr isotopic signature may be explained by high recharge inducing a higher hydraulic pressure on the deep aquifer and an increased contribution of deep saline groundwater to the spring water. The data acquired on this aquifer system allow predicting the evolution of mineralization with climate trends where decreasing recharge would induce a decreasing salinity at the spring outlet.

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*Keywords:* salinization; coastal aquifer; climate change; strontium isotopes; Triassic deposits

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### 1. Introduction

In the context of the global climate change, increasing water demand and the progressive reduction of water resources in the Mediterranean basin<sup>1-3</sup>, understanding the salinization process is essential to the management of

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groundwater resources especially in coastal environments. Indeed, as reported in the annual IPCC report<sup>4</sup> and other studies<sup>5-8</sup>, climate scenarios for the Mediterranean basin clearly indicate decreasing precipitation rate, increasing temperature and a rise in sea level.

In the Mediterranean context, previous studies investigated the origin of salinity in coastal karst aquifers<sup>9-13</sup>. The influence of deep saline water has been demonstrated on isolated coastal karst aquifer located along the Gulf of Lion<sup>11</sup>, resulting from interaction of paleo-seawater with the carbonate matrix. In a similar hydrogeological and geological context demonstrated the influence of deep saline thermal waters on coastal aquifer<sup>9</sup>. More recently, the influence of deep saline water on the shallow karst aquifers at the northern Pyrenean thrust was demonstrated<sup>10</sup>.

In this context, the aim of this study is to investigate the potential impact of climate change variations during the last decade on the mineralization of groundwater at the karst outlet. Through a long term monitoring investigation (between 2001 and 2015) of a major karst system located in the Gulf of Lion continental margin, we assessed the evolution of mineralization at the outlet in relation with the precipitation amount.

The studied karst system is located in an overthrust area composed by Mesozoic carbonate formations. In the studied area, an impermeable clayey-marly unit separates two Jurassic limestone formations, referred to as the upper and lower karst units. The aquifer formations are underlined by the evaporitic levels of the Triassic formations and are covered by a thick impermeable Tertiary formation. Autumn and winter precipitations constitute the main recharge source to the karst system through fissured and fractured limestone largely outcropping in the studied context.

## 2. Material and methods

Samples were filtered in the field through a 0.45 µm MF-Millipore membrane filter on a Sartorius poly-carbonate filter holder, then stored below 4 °C in 60mL HDPE bottles pre-cleaned with hot 10% HNO<sub>3</sub> and deionized water. <sup>87</sup>Sr/<sup>86</sup>Sr ratios were determined at the GIS laboratory, using a TRITON Ti thermal ionization mass spectrometer with an internal normalization for isotope fractionation to an <sup>86</sup>Sr/<sup>88</sup>Sr ratio of 0.1194. Chloride concentration was obtained from the national database (ADES). Rainfall records are provided by Météo-France for the period between 2001 and 2014.

## 3. Results and discussion

The studied spring water shows a relatively high temperature at the emergence, significantly higher (2 to 4 °C) than the average annual temperature of regional karst water. The mineralization, ranging from 800 to 1200 mg/L, is also higher than that of the local and regional karst water<sup>10</sup>. Chloride concentrations vary between 135 to 351 mg/L. Surprisingly, the mineralization of the spring water is higher during wet period and lower during dry period. The variation in the mineralization can be observed both at a seasonal and inter-annual scale. This is illustrated in Fig. 1 (a and b) showing a 4-year running mean of precipitation and chloride concentration record at the studied spring water. The low mineralization level is associated with low <sup>87</sup>Sr/<sup>86</sup>Sr ratio close to that of the local karst groundwater, while, higher mineralization is associated with high <sup>87</sup>Sr/<sup>86</sup>Sr ratio (Fig. 2b). The high <sup>87</sup>Sr/<sup>86</sup>Sr probably results from a long term water-rock interaction with clayey Triassic deposits at depth in high temperature conditions. To confirm the signature of the deep end-member, experimental batches were carried out to recreate the deep groundwater end-member<sup>10</sup> by bringing to equilibrium shallow groundwater with Triassic Marls at varying temperature (Fig. 2a). The results display an increase in <sup>87</sup>Sr/<sup>86</sup>Sr ratio with increasing temperature from 0.7082 at 25°C to 0.7092 at 150 °C<sup>10</sup>.

This may suggest the release of <sup>87</sup>Sr from k-feldspars or mica mineral contained in deep Triassic formations. The <sup>87</sup>Sr/<sup>86</sup>Sr ratios obtained at high temperature is coherent with the deep groundwater end member (Fig. 2a).

The simultaneous variation of the mineralization (as Cl concentration) at the spring water with the amounts of precipitation allows proposing a mechanism for the groundwater mineralization. The increasing chloride concentrations and <sup>87</sup>Sr/<sup>86</sup>Sr ratios with increasing recharge suggests the mobilization of salts water from the lower levels of the aquifer bearing formations.

The studied aquifer system is shown as the combination of two circulation types: i) a shallow water circulation system in the upper karst unit, underlined by impermeable clayey marls, and ii) a deep water circulation system in the lower karst unit underlined by the salty Triassic deposits. Uprising flow the lower karst unit to surface may be driven by a high hydraulic pressure on the lower karst unit due to high precipitation on the limestone plateau. Uprising return flow would be favoured through major faults, fractures and thrust faults.

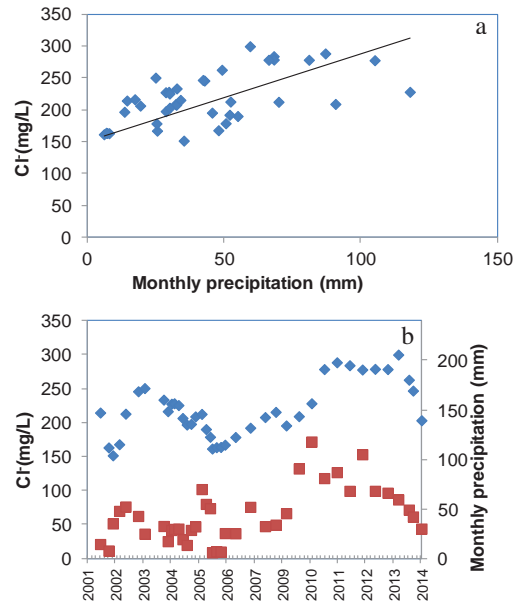


Fig. 1. (a) Correlation of chloride (blue diamond) concentrations with precipitation (Red Square); (b) variations of chloride and precipitations between 2001 and 2014.

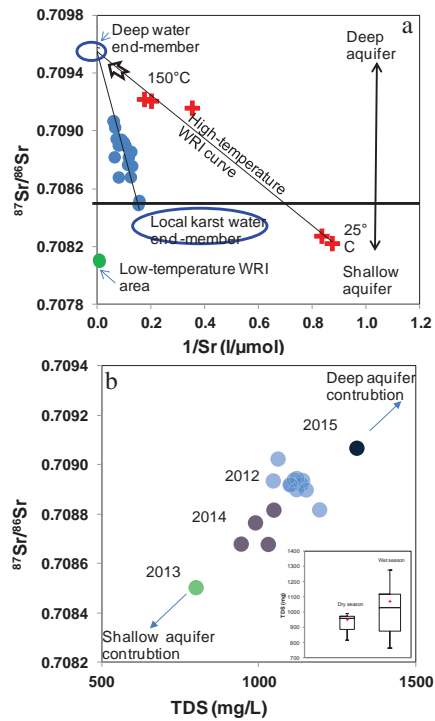


Fig. 2. (a) Sr mixing diagram showing the increase of  $^{87}\text{Sr}/^{86}\text{Sr}$  of the studied aquifer (blue round) toward a radiogenic end member similar to that obtained by experimental batch at high temperature<sup>10</sup> (Red cross), (b)  $^{87}\text{Sr}/^{86}\text{Sr}$  evolution with the TDS values (dates of sampling were reported in diagram).

#### 4. Conclusion

In the studied aquifer system, the spring water TDS values increase with increasing precipitation and show a positive linear tendency with the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios. The leaching of the salty clayey Triassic deposits at depth and high temperature seems to be the major process contributing to the mineralisation of the spring water which is affected by a mixing process with shallow aquifer system. Accessorily, the studied aquifer system provides information on geochemical processes on water rock interaction at deep within aquifers. Finally, the linear relationship that exists between the groundwater mineralisation and precipitation amount will allow predicting the diminution of mineralisation of groundwater in this aquifer system as a consequence of the planned diminution of meteoric recharge in the next 50 years.

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