



Strontium isotopes as tracers of water-rocks interactions, mixing processes and residence time indicator of groundwater within the granite-carbonate coastal aquifer of Bonifacio (Corsica, France)



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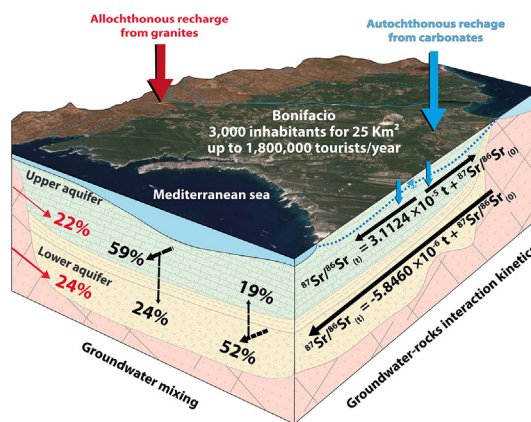
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HIGHLIGHTS

- Strontium and major ions are used to define groundwater mineralization processes.
- Granites and calcarenites are complex sources of Sr^{2+} .
- The $^{87}\text{Sr}/^{86}\text{Sr}$ vs $\delta^{18}\text{O}$ approach is relevant to assess groundwater mixings.
- The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio is suitable to quantify water-rocks interaction kinetics.

GRAPHICAL ABSTRACT



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ABSTRACT

This study aims at identifying the water-rock interactions and mixing rates within a complex granite-carbonate coastal aquifer under high touristic pressure. Investigations have been carried out within the coastal aquifer of Bonifacio (southern Corsica, France) mainly composed of continental granitic weathering products and marine calcarenite sediments filling a granitic depression. A multi-tracer approach combining physico-chemical parameters, major ions, selected trace elements, stable isotopes of the water molecule and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios measurements is undertaken for 20 groundwater samples during the low water period in November 2014. 5 rock samples of the sedimentary deposits and surrounding granites are also analysed. First, the water-rock interactions processes governing the groundwater mineralization are described in order to fix the hydrogeochemical background. Secondly, the flow conditions are refined through the quantification of inter aquifer levels mixing, and thirdly, the kinetics of water-rock interaction based on groundwater residence time from a previous study using CFCs and SF_6 are quantified for the two main flow lines. A regional contrast in the groundwater recharge altitude allowed the oxygen-18 to be useful combined with the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios to differentiate the groundwater origins and to compute the mixing rates, revealing the real extension of the watershed and the availability of the resource. The results also highlight a very good correlation between the groundwater residence time and the

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spatial evolution of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, allowing water-rock interaction kinetics to be defined empirically for the two main flow lines through the calcarenites. These results demonstrate the efficiency of strontium isotopes as tracers of water-rock interaction kinetics and by extension their relevance as a proxy of groundwater residence time, fundamental parameter documenting the long term sustainability of the hydrosystem.

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1. Introduction

Natural groundwater chemical composition in coastal areas is at first order generally controlled by (1) marine influences through seawater intrusion, (2) sea sprays and (3) water-rock interaction (Alcalá and Custodio, 2008; Cary et al., 2014; Dazy et al., 1997; Mongelli et al., 2013). The spatial evolution of groundwater chemistry depends mainly on groundwater origins, boundary conditions, and chemical processes operating along the flow within the aquifer (Appelo and Postma, 2005). Studying the groundwater mineralization processes is fundamental to understand the aquifer functioning and its vulnerability to human activities (Asmael et al., 2014; Djebebe-Ndjiguim et al., 2013). It also provides information on hydraulic connexions between and within aquifer compartments. As many aquifers are highly heterogeneous, such approaches thus contribute to better constrain the different components of the water budget of the hydrogeosystem.

To better constrain the groundwater origins and the water-rocks interactions processes, many isotopes such as radium or chlorine isotopes may be employed (Khaska et al., 2013; Vinson et al., 2012). Strontium isotopes are also widely used in hydrogeology to characterize mineralization processes conditioned by water-rock interactions with magmatic rocks (Casanova et al., 2001; Negrel, 1999; Negrel, 2006; Negrel et al., 2010) or with carbonates (Armstrong et al., 1998; Barbieri et al., 2005; Bishop et al., 1994; Dogramaci and Herczeg, 2002; Ettayfi et al., 2012; Frost and Toner, 2004; Vinson et al., 2012). It has also been successfully employed to describe mixing processes between different groundwater flow lines availing the distinctive strontium signatures of the minerals in the host rocks (Bishop et al., 1994; Bullen et al., 1996; Cartwright et al., 2007; Casanova et al., 2001; Franklyn et al., 1991; Kloppmann et al., 2001; Marques et al., 2012; Negrel et al., 2002; Petelet-Giraud et al., 2003; Petelet-Giraud et al., 2006; Xie et al., 2013). Strontium isotope ratios in rocks vary principally according to the Rb/Sr ratio and the age of parent material because ^{87}Sr is released by beta-decay of ^{87}Rb (Kendall and McDonnell, 1998). As an example, strontium from marine sediments displays generally low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (Brass, 1976; Elderfield, 1986) whereas $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from silicates are generally higher because minerals such as biotite, feldspar and plagioclase have high Rb/Sr ratio (Cook and Herczeg, 2000; Drever, 2005). Examining the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and Sr^{2+} concentrations in groundwater systems is thus a powerful approach for understanding geochemical interactions with the aquifer matrix. Many sources are known for strontium in groundwater and atmospheric deposition is not recognized as a major one since the concentrations found in rainwater are generally low (from 0.3 to 10.8 $\mu\text{g}/\text{L}$) (Herut et al., 1993; Mongelli et al., 2013; Negrel and Roy, 1998; Pearce et al., 2015). Nevertheless, the aerosol sources may influence the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of rainwater, which is variable and may be higher in presence of silicate rocks dusts or lower in presence of carbonate dusts (Bakari et al., 2013; Negrel and Roy, 1998). This input signature is however quickly replaced by the rock signatures of the unsaturated zone during aquifer recharge. The large variation of the $^{87}\text{Sr}/^{86}\text{Sr}$ in groundwater can consequently mostly be explained by the nature of the soil and the aquifer lithology.

The strontium signature evolution in groundwater along flow paths depends so on the aquifer lithology, the rocks alteration levels and the intensity of water-rock interaction, which is conditioned by a time factor (Lasaga et al., 1994). In 1996, Bullen et al. performed a column experiment that simulated the effects of changing residence time of water

in a sand aquifer. This study showed that the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of mobile water is generally lower than in stagnant water due to a longer time of contact with the sand, source of ^{87}Sr . Khaska et al. performed in 2013 an experiment to simulate the evolution of the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio and concentration of the recharge water in the upper part of a karst aquifer with increasing residence time. The Sr concentration in recharge water increased and the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio approached the value of the limestone (0.707813) after approximately 100 h of contact, thus indicating a quick dissolution of calcite. These works suggests that it could be interesting to test the strontium potential as a tool to support and validate the groundwater residence time at the aquifer scale.

This research focuses on the multilayered coastal aquifer of Bonifacio, located in the south of the Corsica Island (France). This aquifer is made of carbonate sediments lying on and within a fractured granitic depression. The groundwater represents the unique drinking water resource of this area of 3000 perennial inhabitants receiving up to 1,800,000 tourists each year. Groundwater provides nearly all the annual needs for the resident population (approximately 17,000 m^3/year), but during the summer, an additional contribution of surface water from the Figari and Ospedale dams (approximately 750,000 m^3/year), outside the watershed, is required to satisfy the increasing demand generated by the tourism. A previous survey using CFCs and SF_6 determined groundwater residence-times (Santoni et al., 2016). The developed flow pattern concept has highlighted the fundamental role of the granites in the aquifer recharge. The contrasted lithologies in the study area and the knowledge on the groundwater residence time along the flow paths seems consequently promising to track the strontium isotopes behaviour over time within the aquifer. In this way, a detailed description of the mineralization and mixing processes has been conducted here, fundamental prerequisite to validate and refine the flow pattern concept of the aquifer established previously. Strontium isotopes have been successfully employed for examining mixing processes in coastal areas (Anders et al., 2014; Bakari et al., 2013; Khaska et al., 2013; Tillman et al., 2014; Vengosh et al., 1999) or between granites and carbonate rocks (Cary et al., 2015; Petelet et al., 1998) but no study compiled all of these contexts at the same place. This study will also be an opportunity to better constrain the different components of the local water balance, which is a prominent issue for this coastal aquifer that is intensively exploited during the summer period to provide drinking water to the tourists. Indeed, the recharge provided by the neighbouring fractured granites needs to be accurately quantified because constituting an additional natural recharge potentially in favour of the sustainability of groundwater resources despite the semi-arid climatic context usually limiting the aquifers recharge potential (Custodio, 2010; Ghiglieri et al., 2014; Martos-Rosillo et al., 2015; Stigter et al., 2014).

2. Study area

2.1. Location and climate

The Bonifacio plateau is located in the southern part of the Corsica Island (Western Mediterranean, France). It is bounded in equal proportions by granites at the East and the West, and by the Mediterranean Sea at the Northeast and South (Fig. 1). Its triangular shape extends over an area of 25 km^2 with a mean elevation of 80 m asl. The carbonate deposits fill a granitic depression, and are in contact with granitic relief

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