



Determination of low-level Radium isotope activities in fresh waters by gamma spectrometry



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ABSTRACT

A new portable sampling system was developed to extract Radium isotopes from large volumes (up to 300 L) of fresh surface- and ground-waters of low Ra-activities (< 5 mBq/L). Ra is quantitatively adsorbed on a small amount (6.5 g) of MnO₂-coated acrylic fibers, which are then dried and burned at 600 °C in the laboratory. The resulting Mn-oxide powder (about 2 cm³ when compacted) is then analyzed through gamma-ray spectrometry which allows measurement of the whole Ra quartet (²²⁶Ra, ²²⁸Ra, ²²⁴Ra and ²²³Ra) in a single counting of a few days. The usual relative standard combined uncertainties (1σ) are 2–3% for ²²⁶Ra, ²²⁸Ra and ²²⁴Ra; and less than 10% for ²²³Ra. This method was applied to the analysis of Ra in karstic waters of the Lez aquifer, and surface- and ground-waters of the upper and middle Vidourle watershed (South of France). The analyzed waters have relatively low ²²⁶Ra activities (1–4 mBq/L) in both cases, regardless of the contrasted geology (Mesozoic limestone vs crystalline Variscan basement), but clearly distinct (²²⁸Ra/²²⁶Ra) ratios in agreement with the differences in Th/U ratios of the two drained areas. Short-lived Ra isotopes (²²⁴Ra and ²²³Ra) appear to be mainly influenced by near-surface desorption/recoil processes for most of the sampling sites.

1. Introduction

The use of Ra isotopes in water as tracers for hydrological, geochemical and biological processes has received widespread attention for many years. Each isotope gives useful information on the various processes that could be active in different environments, because of their contrasted half-lives and origins (²²⁶Ra, $t_{1/2} = 1600$ y, from the ²³⁸U decay-series; ²²³Ra, $t_{1/2} = 11.4$ d, from the ²³⁵U series; ²²⁸Ra, $t_{1/2} = 5.75$ y and ²²⁴Ra, $t_{1/2} = 3.66$ d, both from the ²³²Th decay-series).

One of the main current applications of Ra isotopes in the hydro-sphere is for tracing submarine groundwater discharge (SGD) in coastal systems (Charette et al., 2001; Gonnee et al., 2008; Hwang et al., 2005; Rama and Moore, 1996; Rodellas et al., 2015; Young et al., 2008). However, fewer studies concern their use in continental fresh surface- and ground-waters, in spite of their interest to distinguish the sources of water masses and the processes and timescales of water-rock interactions (e.g. Chabaux et al., 2003; Osmond and Coward, 2000; Porcelli and Swarzenski, 2003; Szabo et al., 2012; Tomita et al., 2014). Contrary to thermal waters and brines with often high Ra isotope activities, fresh waters have generally much lower activities (typically a

few millibecquerel per liter of ²²⁶Ra, ²²⁸Ra and ²²⁴Ra). This explains in part why very few studies have reported Ra isotope data on continental karstic waters (e. g. Guerrero et al., 2016). As ²²⁶Ra, ²²⁸Ra and ²²⁴Ra have similar activities, they are potentially all measurable through nuclear methods from the same volume of water (e.g. by alpha spectrometry, Eikenberg et al., 2001; Surbeck, 2000). For techniques measuring the number of atoms instead of the activity, such as MC- or SF-ICP-MS, only ²²⁶Ra, with the longest half-life, can be easily measured from relatively small volumes of water (< 2 L, Bourquin et al., 2011; Copia et al., 2015). Note that 1 mBq/L of ²²⁶Ra corresponds to $27.3 \cdot 10^{-15}$ g/L. ²²⁶Ra is also often analyzed through liquid scintillation (Rodríguez et al., 2016; Purkl and Eisenhauer, 2004), or ²²²Rn-emanometry (e. g. Perrier et al., 2016). However, the measurement of ²²³Ra, whose activity is around 20 times lower than that of ²²⁶Ra, requires sampling of much larger volumes of water, regardless of the analytical technique used for the quantification (e.g. RaDDeC, Moore and Arnold, 1996; Moore, 2008, or gamma-spectrometry, Condomines et al., 2010; Van Beek et al., 2010). The interest of gamma-spectrometry is that all four Ra isotopes can be measured in a single analysis, soon after sampling.

Since 1973, Radium concentration on MnO₂-coated fibers (Mn-

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Table 1
Sampling site description.

System	Sampling site	Site ID	UTM coordinates (Zone 31T)		Hydrological context	Geological context
			X (m)	Y (m)		
Lez aquifer	Restinclières	RESO	568440	4840676	Spring	Marly-limestones (Cretaceous)
	Lez	LZSO	568001	4840908	Spring	Limestones (Upper Jurassic) & marly-limestones (Cretaceous)
Vidourle watershed	L'Esclafar	VIHA	564054	4874597	Stream water	Cévennes granites (Carboniferous)
	Figaret-La Borie	FILB	568209	4872143	Stream water	Cévennes granites (Carboniferous) and schists (Cambro-Ordovician)
	Figaret-Résurgence	FIRP	568185	4869702	Outlet	Limestones and dolomites (Middle Jurassic)
	Sauve	SASO	576204	4865746	Spring	Limestones (Upper Jurassic) & dolomites (Middle Jurassic)

fibers) has become one standard technique for measurement of waters with low Radium activity (Moore and Reid, 1973). Indeed, at moderate water flow (below 2 L/min), a few grams of these fibers are able to quantitatively extract Radium from several tens of liters of water, with no chemical reactive consumption. In coastal and marine systems, the most common method consists of collecting several liters (from tens to hundreds) of water in one or many containers, and then, let them flow, by gravity, through a cartridge filled with at least 20 g of Mn-fibers (Dulaiova and Burnett, 2004).

We present in this paper a new portable and optimized sampling system using only 6.5 g of Mn-fibers and adapted to extract, in the field, Ra from up to 300 L of water. After burning of the fibers, the activities of all the four Ra isotopes are then measured through gamma-spectrometry in a single analysis, with counting times from 2 to 7 days (Condomines et al., 2010; Van Beek et al., 2010). Our method was applied to two different and geologically contrasted areas in the South of France: the Lez karstic system developed in Mesozoic limestones, and the Vidourle upper watershed, draining granitic and metamorphic rocks of the Variscan basement as well as Mesozoic dolomites and limestones. The sampling sites are described in Table 1.

2. Measurements and methods

2.1. Sampling and sample preparation

A compact and portable sampling system was built and optimized (Fig. 1). The sampling process is accomplished in two steps:

1) *Sample particle filtering and volume measurement*: between 100 L and 300 L of water are continuously pumped (A in Fig. 1) and filtered

through 25 μm and 5 μm particle filters (B and C), to fill the 300 L ($58 \times 34 \times 186$, in cm) HDPE container (D), adapted with an external water level indicator (1.25 L resolution).

2) *Radium pre-concentration*: The water in the container is pumped (F) through the first acrylic-fiber filter cartridge (G) at ≤ 2 L/min, where most of the finest particles ($< 5 \mu\text{m}$) are retained, and then, through the Mn-fiber cartridge (H), where Ra is quantitatively extracted. Sampling in the field takes between four and five hours for a 300 L water sample. A water level automatic switch (E) has been installed to prevent the pump (F) to get dry.

All fibers and cartridges for the second step were provided by Scientific Computer Instrument[®]. The Mn-fiber cartridge is packed as follows (from bottom to top): half cartridge volume of distilled water, 1 g of acrylic fiber, 6.5 g of Mn-fiber, 2.5 g of acrylic fiber, between 10 and 20 plastic spheres (~ 5 mm diameter), and finally, distilled water until the column is filled. The Mn-fibers were previously rinsed and finely disheveled in distilled water, and then introduced directly into the cartridge, trying to eliminate the air bubbles with a plastic stick. A plastic grid was intercalated between each layer to make easier the extraction of the fiber after sampling. The upper acrylic fiber in the Mn-cartridge was used as an extra filter to minimize the finest particles ($< 5 \mu\text{m}$) adsorption on the Mn-fiber, whereas the lower acrylic fiber was used to avoid possible Mn-fiber floss loss. Both acrylic fibers were also used as a visual indicator of the efficiency of the particle filtration.

In order to optimize the Ra adsorption on the Mn-fiber column, the formation of air bubbles in the column must be avoided, since it can lead to incomplete wetting of the Mn-fibers; and the Mn-fiber must be finely disheveled to maximize the contact surface by separating the fiber floss. The first can be achieved by purging the pipelines all along

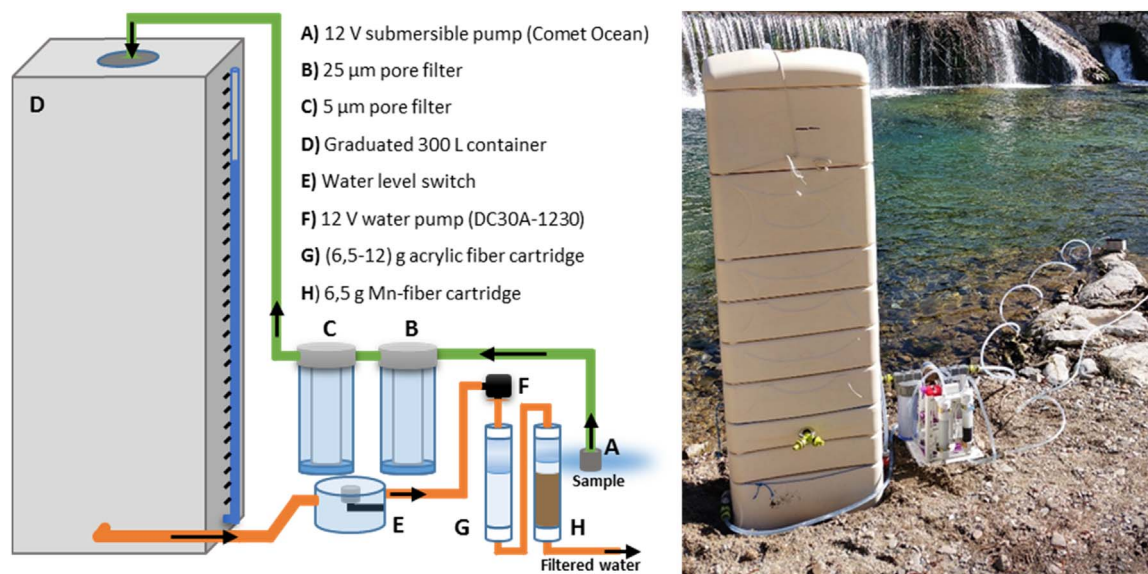


Fig. 1. Radium sampling system. The green line corresponds to the first stage of particle filtration and filling of the container. The orange line indicates the Ra pre-concentration device. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

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