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Low-level ^{226}Ra determination in groundwater by SF-ICP-MS: optimization of separation and pre-concentration methods

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Abstract

Background: Inductively coupled plasma mass spectrometry (ICP-MS) techniques have been widely used for analysis of long-lived environmental radionuclides. In this paper, we present an optimization of the sector field (SF)-ICP-MS technique for the analysis of ^{226}Ra in groundwater samples using a method of pre-concentration of radium in water samples.

Methods: The separation protocol and a sequential application of ion exchange and extraction chromatography have been optimized, and related polyatomic interferences and matrix effects affecting the ^{226}Ra signal were investigated.

Results: Analyzing 12 replicates (water spiking at 22 fg g^{-1} of ^{226}Ra), the ^{226}Ra recovery efficiency close to 100 % has been obtained. The instrumental ^{226}Ra detection limit of 0.09 fg g^{-1} (3σ criterion) and the absolute detection limit of 0.05 fg in a 25-mL groundwater sample have been reached.

Conclusions: An optimization of the radium separation method and a pre-concentration of radium in groundwater samples led to high radium recoveries, almost up to 100 %. The same could be said with respect to the separation of the interfering elements, important for the quantitative ^{226}Ra analysis by SF-ICP-MS. The improvements in the separation and pre-concentration techniques also helped to improve the ^{226}Ra detection limit down to $0.05\text{ fg}/25\text{ mL}$ of groundwater sample.

Keywords: Groundwater; SF-ICP-MS; ^{226}Ra ; Extraction chromatography; Polyatomic interferences

Background

The radium-226, ^{226}Ra ($T_{1/2} = 1622$ years), is a naturally occurring radioisotope belonging to the ^{238}U decay chain, which has widely been used as a tracer in groundwater and seawater transport and mixing (Smith et al. 2012), as well as for studying water-rock interactions in groundwater reservoirs (Reynolds et al. 2003). Several analytical radiometric and mass spectrometry techniques have been developed for the analysis of ^{226}Ra in environmental samples: radon emanation (Kim et al. 2001), liquid scintillation spectrometry (Repinc and Benedik 2002), alpha-ray spectrometry (Morvan et al. 2001), gamma-ray spectrometry (Johnston and Martin 1997), thermal ionization mass spectrometry (TIMS) (Cohen

and O'Nions 1991), and inductively coupled plasma mass spectrometry (ICP-MS) (Chabaux et al. 1994; Park et al. 1999; Lariviere et al. 2003; Lariviere et al. 2005; Zheng and Yamada 2006; Varga 2008; Cozzella et al. 2011; Hou and Roos 2008; Butler et al. 2015). With the exception of ICP-MS, these methods are time consuming, often expensive, and therefore not convenient when large number of samples are to be analyzed. However, the ICP-MS method has also some drawbacks, first of all possible interferences, which could make precise ICP-MS analysis of long-lived radionuclides in the environment difficult (Wyse et al. 2001; Lee et al. 2008; Lehto and Hou 2010; Lujanienė et al. 2013).

The aim of the present work has been to develop a robust technique for the ^{226}Ra analysis of groundwater samples collected from the Gran Sasso (GS) aquifer (central Italy) (Plastino et al. 2013). Within the framework of the Environmental Radioactivity Monitoring for

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Earth Sciences (ERMES) project carried out at the Istituto Nazionale di Fisica Nucleare (INFN) Gran Sasso National Laboratory, 1-L groundwater samples have been collected weekly since 2008 at ten different sites located in the underground laboratory (Plastino et al. 2010; Plastino et al. 2011). In order to analyze with high precision such large number of samples necessary for the development of ^{226}Ra time series, it has been proposed to develop a new analytical technology, which would be based on ICP-MS.

To accomplish the above requirements, a method described by Lariviere et al. (2005) has been chosen as a suitable base for further developments. This method consists of the sequential application of a cation exchange resin (50W-X8) and an extraction chromatographic resin (Sr resin). The first method has been widely used to separate ^{226}Ra in liquid matrices (Lariviere et al. 2005; Varga 2008; Cozzella et al. 2011; Hou and Roos 2008; Butler et al. 2015), while the latter retains Sr and partially also Ba, the main polyatomic interferences in the analysis of ^{226}Ra in groundwater samples (Chabaux et al. 1994; Horwitz et al. 1992). Due to high-precision data requirements for the development of the ^{226}Ra groundwater time series, the original Lariviere et al. (2005) method has been modified by adding a pre-concentration part, effects of which were thoroughly investigated. This work focuses therefore on the optimization of the separation protocol, on the pre-concentration of samples, and on the evaluation of particularly related polyatomic interferences and matrix effects that could affect the ^{226}Ra peak window.

Methods

Reagents and standards

Deionized water (Milli-Q water system, Millipore, Bedford, USA) and hyperpure hydrochloric and nitric acids (Panreac, Barcelona, Spain) were used during this study. Mono-elemental standard of Ca, Ba, Mg, Sr, Ce, La, and Nd (NIST, Gaithersburg, USA) in stock solutions were diluted to obtain matrix-matched standards and spikes. Radium standard and spikes were obtained by dilution from a 100.5 Bq g^{-1} STD (PTB, Braunschweig, Germany). Pre-concentration and separation of ^{226}Ra were performed using two commercially available resins: a strong cation exchange resin (AG-50W-X8) and a crown ether-based resin (Sr resin) (Triskem, Bruz, France).

Instrumentation

In order to determine ^{226}Ra in groundwater samples, a sector field (SF)-ICP-MS (Finnigan Element 2, Thermo Scientific), equipped with an Apex Q high-sensitivity introduction system (Element Scientific Inc.), was used. The Apex Q system was tested with three different micro-nebulizers: 64, 100, and 200 $\mu\text{L}/\text{min}$, using a 100-ppt Thermo Tuning solution and monitoring of ^{238}U as

a descriptive of heavy elements. The 100 $\mu\text{L}/\text{min}$ one showed the best performance. A spray chamber operational temperature of 100 $^{\circ}\text{C}$ and a Peltier-cell condenser operational temperature of 2 $^{\circ}\text{C}$, optimal for aqueous samples, were chosen. The N_2 flow pressure was not crucial for the gain of the system, in the sense that above a certain value, flux variations did not modify the counting rate in the uranium window. The instrument has been optimized daily with respect to the torch position and to the sample and the auxiliary gas flow, in order to reach a maximum counting rate and a stable signal. The acquisition parameters were chosen with an isotopic ratio measurement approach (Hamester et al. 1999), with a narrow mass window in order to gain in sensitivity, and with a long acquisition time to improve counting statistics. The optimized parameters are reported in Table 1.

Sample preparation

The optimization of the procedures for the ICP-MS analysis of ^{226}Ra in groundwater samples was carried out with the aim to accomplish separation of radium from matrix constituents and from spectral interfering elements. One gram of (AG-50W-X8) resin was packed into a plastic cartridge. The column was pre-washed and conditioned using 10 mL of 4-M HNO_3 , 5 mL of H_2O Milli-Q, and 10 mL of 1.7-M HCl. Then, 25 mL of acidified (pH 1 HCl)

Table 1 Optimized values of Finnigan Element 2 with Apex Q introduction system

Instrumental parameters	
Gas flow (L min^{-1})	
Sample	0.875
Auxiliary	0.2–0.3
Cool	16
RF power (W)	1250
Torch position	Optimized daily
Interface cones	Nikel
Acquisition parameters	
Resolution	300
Number of run	3
Number of passes	3
Samples per peak	50
Mass window (%)	40
Integration window (%)	100
Sample time (s)	0.5
Acquisition time (s)	10
Apex Q parameters	
Spray chamber temperature ($^{\circ}\text{C}$)	100
Peltier-cell condenser temperature ($^{\circ}\text{C}$)	2
N_2	On

sample was loaded. After that, 15 mL of 2.5-M HCl were eluted to wash away Ca, Mg, and the other matrix constituents.

At this point, the AG-50W-X8 and Sr resin cartridges were connected in series, and 10 mL of 4-M HNO₃ were eluted through to desorb Ra from AG-50W-X8 and separate it from Sr and Ba in the Sr resin. Then, 10 mL of 3-M HNO₃ were eluted in order to completely recover Ra. At the end of this procedure, the solution containing Ra was collected, evaporated, and re-dissolved in 0.5 mL of 3-M HNO₃.

Results and discussion

Starting from the a priori assumption that ²³⁸U decay chain is in secular equilibrium in the investigated groundwater samples, and having a ²³⁸U concentration between 1 and 2 ng g⁻¹, a ²²⁶Ra concentration around 0.3–0.7 fg g⁻¹ was expected. At such a low concentration, radium was barely detectable by available instrumentation, so a pre-concentration was needed. A pre-concentration factor of 50 was chosen because, on one side, it guarantees a measurable signal and a good counting statistics; on the other hand, a small sample amount of 25 mL of water is sufficient for the analysis. The ²²⁶Ra ICP-MS measurements suffer of both spectral and nonspectral interferences; therefore, it is crucial to understand how these elements are separated in the procedure, how big are their levels in the final solution, and which are their effects on the measurement.

Polyatomic interferences studies

As reported in the literature (Epov et al. 2003), ²²⁶Ra measurements by ICP-MS may suffer several polyatomic interferences:

- ⁸⁸Sr¹³⁸Ba
- ⁴⁰Ar⁴⁰Ar¹⁴⁶Nd
- ⁸⁷Sr¹³⁹La
- ⁸⁶Sr¹⁴⁰Ce

The aim of this part of the work has been to characterize in Gran Sasso groundwater samples the matrix concentrations of the elements Ba, Sr, Ce, La, and Nd, which are responsible for interferences, and to evaluate if, after a pre-concentration, these interferences could really be observed, and what would be their impacts on the Ra peak. First of all, each interfering element signal in the groundwater sample and in the solution after the elution procedure was measured. Then, the signal values after evaporation were calculated (Table 2). Considering the values in the fifth column of this table as representatives of the signals of interfering elements after evaporation and re-dissolution, we mimicked these concentrations using single-element standard solutions and then evaluated these effects in the ²²⁶Ra peak window.

Table 2 The blank (BLK) and Gran Sasso (GS) groundwater signal values (columns 2 and 3)

Isotope	BLK (s ⁻¹)	GS sample (s ⁻¹)	Sr resin (s ⁻¹)	EVAPC (s ⁻¹)	Std (s ⁻¹)
¹³⁸ Ba	149 · 10 ³	97 · 10 ⁶	3 · 10 ⁶	120 · 10 ⁶	–
⁸⁸ Sr	44 · 10 ³	51 · 10 ⁶	490 · 10 ³	19.7 · 10 ⁶	63.9 · 10 ⁶
¹³⁹ La	175	1.8 · 10 ³	–	90.5 · 10 ³	960 · 10 ³
¹⁴⁰ Ce	475	830	–	41.5 · 10 ³	910 · 10 ³
¹⁴⁶ Nd	308	1.4 · 10 ³	–	68 · 10 ³	200 · 10 ³
²²⁶ Ra	0.6	–	–	–	0.6

The fourth and fifth columns report signal values after Sr elution and total pre-concentration procedure, respectively. The sixth column reports signal values of a Ce, La, Nd, and Sr standard (50 pg g⁻¹ of Ce, La, and Nd and 10 ng g⁻¹ of Sr) for interference evaluation

In order to evaluate ⁸⁸Sr¹³⁸Ba effects, an interference calibration curve using 500 ng g⁻¹ of Ba, and increasing the levels of Sr (0, 10, 20, 30, 40, and 50 ng g⁻¹), was obtained and measured. Concentration values were selected by comparing the expected signals to Sr and Ba calibration curves. When the concentrations of Ba and Sr were under 500 and 20 ng g⁻¹, respectively, the counts in the Ra window could be considered as a fluctuation of the background, while above these values, they were signs of an interference (Fig. 1).

Possible interferences from Ce, La, and Nd were evaluated measuring a solution containing 50 pg g⁻¹ of Ce, La, and Nd and 10 ng g⁻¹ of Sr. These concentrations were around ten times higher than actual values in the final step of the procedure. As shown in Table 2, no sign of interferences were observed.

Matrix interferences

A presence of Ca and Mg in groundwater can affect the sensitivity of the ICP-MS system, both in a positive and a negative way. Assuming a pre-concentration factor of 50, the matrix element concentration values are reported in Table 3. Starting from these values (column 3 (x)), and maintaining the ratio between element concentrations, we constructed a matrix calibration curve for four levels (8x/3, x, x/2, x/6), with a fixed level of Ra (33 fg g⁻¹). There was a nonlinear decrease in the element signal, likely due to a loss of ionizing efficiency, together with a quick and progressive obstruction of the skimmer cone.

These effects preclude a priori the application of chromatographic extraction methods without the separation of matrix elements, as for example in the Lariviere et al. (2003) method, or in the mere application of a Sr resin in order to eliminate polyatomic interferences. After application of the elution procedure, the matrix elements were separated with efficiency higher than 99 %. Then, the sample was concentrated by a factor 40, obtained evaporating to dryness the 20 mL of elution products

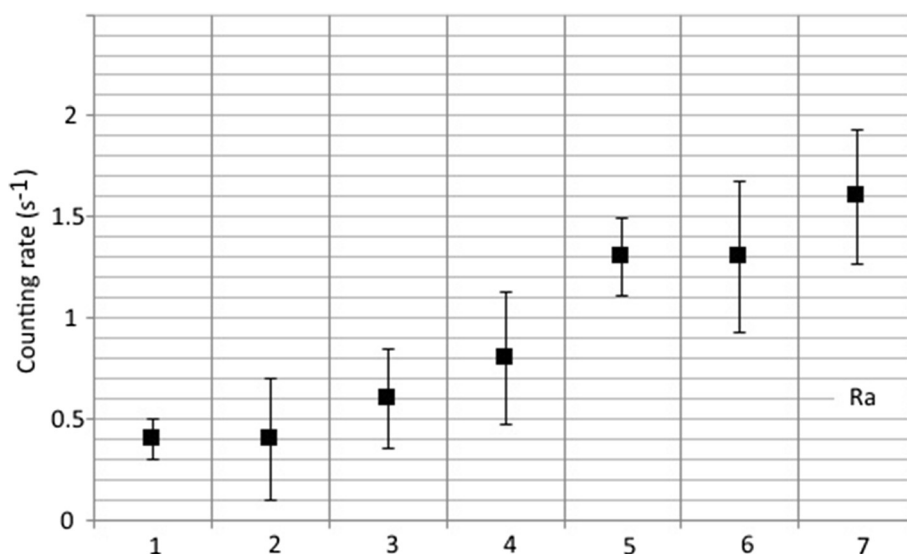


Fig. 1 Counting rates in the Ra peak in solutions. The fixed concentration of Ba was 500 ng g⁻¹. The Sr concentrations varied (X-axis): 1 – blank; 2 – 0; 3 – 10; 4 – 20; 5 – 30; 6 – 40; 7 – 50 ng g⁻¹

and re-dissolving in 0.5 mL of 3-M HNO₃. Signal values and relevant efficiencies are reported in Table 3. The matrix effect was then evaluated for concentrations around those previously obtained, using five matrix-matched ²²⁶Ra standards, with increasing concentrations of Ca (1, 2, 5, 10, and 20 μg g⁻¹) and Mg (0.25, 0.5, 1.25, 2.5, and 5 μg g⁻¹), and comparing them with a standard without Ca and Mg, at the same ²²⁶Ra concentration (27.5 fg g⁻¹). The obtained results are also reported in Table 3. Results showed that the presence of the matrix has a positive effect on the signal, remaining quite

constant over the concentration range. This can be taken into account in measurement sessions performing a calibration curve acquisition with standards matching the Ca and Mg proposed concentrations.

Optimization of separation procedures and recovery studies

The utilization of a strong cation exchange resin (AG-50W-X8) in separating Ra from water matrix constituents has been well documented in the literature (Lariviere et al. 2003, 2005; Varga 2008; Cozzella et al. 2011; Park et al. 1999). The procedure can be divided into four steps:

- (i) pre-washing and conditioning
- (ii) sample loading
- (iii) washing away Ca, Mg, and other matrix constituents using HCl
- (iv) recovering of Ra using HNO₃

These methods differ mainly in HCl molarity in step 3. In order to evaluate the separation efficiencies and the

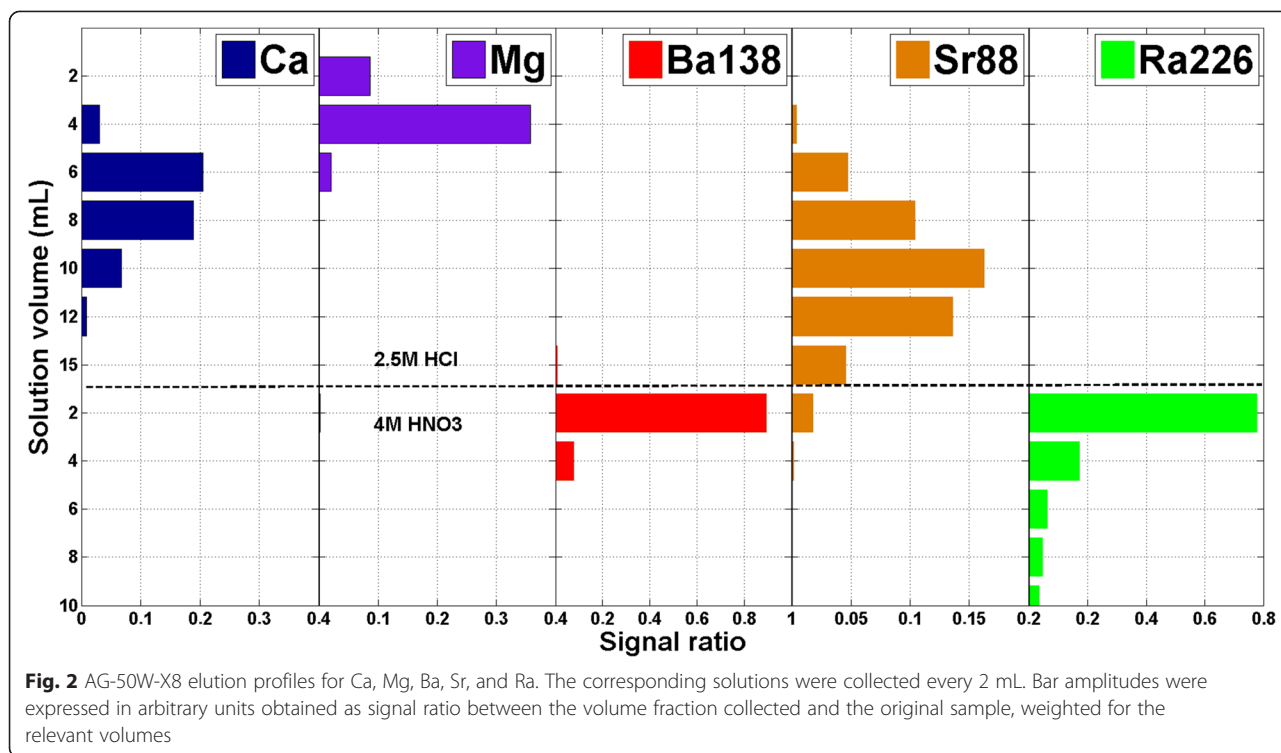
Table 3 Typical Ca, Mg, K, and Na concentration values in a sample of Gran Sasso (GS) groundwater (column 2), theoretical values after pre-concentration without separation (column 3), Ca and Mg signals in GS groundwater (column 4), after elution procedure (column 5), after evaporation (column 7), and the relevant separation efficiencies (column 6)

Isotope	GS (μg g ⁻¹)	GSAPCWS(x) (μg g ⁻¹)	GS (s ⁻¹)	After proc. (s ⁻¹)	Sep. eff. (%)	After evap. (s ⁻¹)
Ca	18	1000	46 · 10 ⁶	0.26 · 10 ⁶	99.4	10.4 · 10 ⁶
Mg	9.4	500	860 · 10 ⁶	1.63 · 10 ⁶	99.8	65.2 · 10 ⁶
Na	0.9	50				
K	0.2	10				
Isotope	S1	S2	S3	S4	S5	S6
Ca (μg g ⁻¹)	1	2	5	10	20	0
Mg (μg g ⁻¹)	0.25	0.5	1.25	2.5	5	0
Ra (s ⁻¹)	93(2)	93(1)	96(3)	97(1)	95(1)	85(2)

In the latter part of the Ra signals with respect to increasing values of Ca and Mg, each sample was spiked at ²²⁶Ra concentration of 27.5 fg g⁻¹

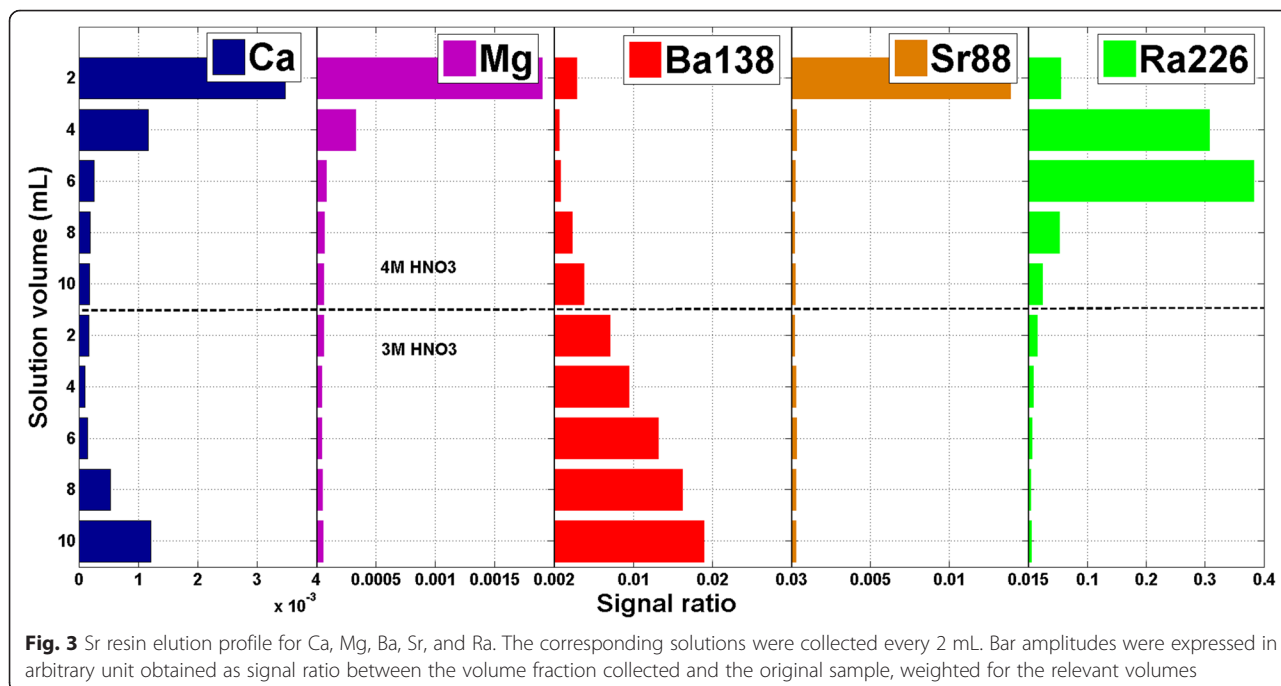
Table 4 The Ra recovery efficiencies and the Ba, Sr, Ca, and Mg separation efficiencies

Step 3	Recovery eff. (%)	Separation efficiency (%)				
HCl M	²²⁶ Ra	⁴³ Ca	²⁵ Mg	⁸⁸ Sr	¹³⁸ Ba	
1.7	86.9	68	98.2	19.8	23.4	
2.5	100	99.7	99.9	96.4	12.1	
4	64.2	99.8	99.9	99.7	96.2	
6	9.1	99.8	99.9	99.6	76.4	



radium recovery for each method, four replicas with different HCl molarity in phase 3 were done, using a GS sample spiked at 22 fg g⁻¹ of ²²⁶Ra. The obtained results are reported in Table 4. Considering recovery and separation efficiencies, a 2.5-M HCl wash seemed the best choice, as suggested by Lariviere et al. (2005).

The elution profiles for most important elements are reported in Figs. 2 and 3. They graphically show in which step of the procedure each single element was eliminated or recovered. The first one shows a very good separation efficiency for Ca and Mg and also a very good recovery efficiency for Ra. Ba and Sr were not retained properly by



this resin, so using Sr resin was required to avoid formation of the $^{88}\text{Sr}^{138}\text{Ba}$ interference.

After washing with HCl (step 3), the two cartridges were connected in series and the rest of the procedure was performed. The series connection allows to avoid an intermediate evaporation step in the Lariviere et al. (2005) procedure, with a significant reduction of its duration. Loading a sample in the Sr resin at 4-M HNO_3 did not affect the column capacity factor k' , as previously described (Plastino et al. 2010). After the elution in Sr resin, as shown in Fig. 3, the separation efficiencies of Ba and Sr were 98 and 99.3 %, respectively, and the resulting values of Ba and Sr were below the level of interference formation (Fig. 1). The amount of 3-M nitric acid rinse was then reduced to 5 mL.

Analyzing 12 replicates (water spiking at 22 fg g^{-1} of ^{226}Ra), the ^{226}Ra recovery efficiency close to 100 % has been obtained thanks to the optimization of the radium separation method and a pre-concentration of radium in groundwater samples. The same could be said with respect to the separation of the interfering elements, important for the quantitative ^{226}Ra analysis by SF-ICP-MS. The improvements in the separation and pre-concentration techniques also helped to improve the ^{226}Ra detection limit down to $0.05 \text{ fg}/25 \text{ mL}$ of groundwater sample. The absolute detection limit for a water sample equivalent of $1.95 \cdot 10^{-18} \text{ g g}^{-1}$ has been obtained.

Conclusions

We presented an optimization of the SF-ICP-MS technique for the analysis of ^{226}Ra in groundwater samples using a method of pre-concentration of radium in water samples. The separation protocol and a sequential application of ion exchange and extraction chromatography have been optimized, and related polyatomic interferences and matrix effects affecting the ^{226}Ra signal were investigated. The ^{226}Ra recovery efficiency close to 100 % has been obtained thanks to the optimization of the radium separation method and a pre-concentration of radium in groundwater samples. The same could be said with respect to the separation of the interfering elements, important for the quantitative ^{226}Ra analysis by SF-ICP-MS.

The improvements in the separation and pre-concentration techniques helped to improve the ^{226}Ra detection limit down to $0.05 \text{ fg}/25 \text{ mL}$ of groundwater sample. The absolute detection limit for a water sample is equivalent to $1.95 \cdot 10^{-18} \text{ g g}^{-1}$. A hydrological paper describing the measured ^{226}Ra concentrations and discussing its behavior in the groundwater system of the Gran Sasso massif is under preparation.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

LC was responsible for the method optimization, sample preparation, and data evaluation. SN was responsible for the SF-ICP-MS measurements and method optimization. WP was responsible for the coordination of the project, organization of groundwater sampling campaigns in the Gran Sasso National Laboratory, data evaluation, and drafting of the manuscript. MC was responsible for the data evaluation. PPP was responsible for the data evaluation and for the finalization of the manuscript. All authors read and approved the final manuscript.

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