



Rapid determination of actinides and ^{90}Sr in river water



A. Habibi^{a,*}, B. Boulet^a, M. Gleizes^b, D. Larivière^c, G. Cote^d

^a IRSN/PRP-ENV/STEME/LMRE, Rue du belvédère, Bâtiment 501, Bois des rames, 91400 Orsay, France

^b IRSN/PRP-ENV/STEME, 31 rue de l'écluse, 78116 Le Vésinet, France

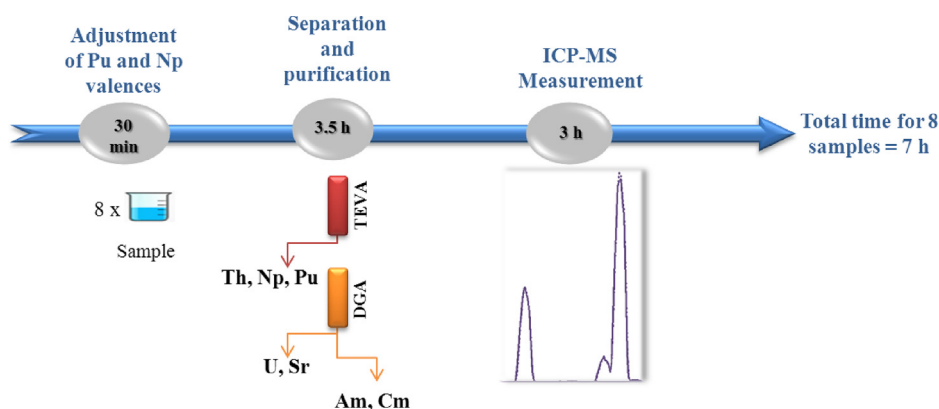
^c Laboratoire de radioécologie, Département de chimie, Université Laval, 1045 Avenue de la médecine, G1V 0A6 Québec, Canada

^d PSL Research University, Chimie ParisTech CNRS, Institut de Recherche de Chimie Paris, 11 rue Pierre et Marie Curie, 75005 Paris, France

HIGHLIGHTS

- A new method to separate six actinides and ^{90}Sr was developed.
- The method was applied successfully to river water samples.
- The separation and the measure take about seven hours.
- The method permits to reach high yields.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 31 December 2014

Received in revised form 3 April 2015

Accepted 12 April 2015

Available online 18 April 2015

Keywords:

Actinides

^{90}Sr

Emergency

River water

Extraction chromatography

Inductively coupled plasma-mass spectrometry

ABSTRACT

Nuclear accidents occurred in latest years highlighted the difficulty to achieve, in a short time, the quantification of alpha and beta emitters. Indeed, most of the existing methods, though displaying excellent performances, can be very long, taking up to several weeks for some radioisotopes, such as ^{90}Sr .

This study focuses on alpha and beta radioisotopes which could be accidentally released from nuclear installations and which could be measured by inductively coupled plasma mass spectrometer (ICP-MS). Indeed, a new and rapid separation method was developed for $^{234,235,236,238}\text{U}$, $^{230,232}\text{Th}$, $^{239,240}\text{Pu}$, ^{237}Np , ^{241}Am and ^{90}Sr . The main objective was to minimize the duration of the separation protocol by the development of a unique radiochemical procedure with elution media compatible with ICP-MS measurements.

Excellent performances were obtained with spiked river water samples. These performances are characterized by total yields exceeding 80% for all monitored radionuclides, as well as good reproducibility ($\text{RSD} \leq 10\%$, $n=12$). The proposed radiochemical separation (including counting time) required less than 7 h for a batch of 8 samples.

© 2015 Elsevier B.V. All rights reserved.

1. Introduction

To limit the exposure of populations to radiological contamination in food and in the environment in accidental and post-accidental scenarios, the development of new and rapid radio-analytical methods oriented toward the quantification of actinides

* Corresponding author. Tel.: +33 169855847.

E-mail address: azza.habibi@irsn.fr (A. Habibi).

Table 1

Overview of existing protocols to rapidly quantify actinides and strontium using radiometric techniques.

Radionuclides analyzed	Matrix	Separation columns used	Detection	Duration time	References
U, Pu, Am, Cm, Th, Np and ^{89/90} Sr	Water and urine	TEVA Resin [®] , TRU Resin [®] , Sr-Resin [®]	Alpha spectrometry Gas proportional counting	4.5–20 h	Maxwell [8]
U, Pu, Am, Th, Pu, Np, Am, Cm, U	Soil	TEVA Resin [®] , TRU Resin [®] , UTEVA Resin [®]	Alpha spectrometry	N.A. ^a	Kim et al. [4]
Pu, Np, Am, Cm, U	Food	TEVA Resin [®] , TRU Resin [®] , DGA Resin [®]	Alpha spectrometry	3.5–19 h	Maxwell et al. [12]
Pu, Np, Am, Cm, U	Urine	TEVA Resin [®] , TRU Resin [®] , DGA Resin [®]	Alpha spectrometry	5–25 h	Maxwell et al. [13]
Pu, Np, Am, Cm, U, Sr	Animal tissue	TEVA Resin [®] , TRU Resin [®] , DGA Resin [®]	Alpha spectrometry Gas proportional counting	1 day	Maxwell and Faison [14]

^a N.A.: not available.

and radiostrontium is essential [1]. These radionuclides have been identified as strategic from an accidental release scenario as they would be responsible of the majority of the alpha and beta contamination of the environment. These radionuclides can be measured using either radiometric or mass spectrometric detectors based on the emission of ionizing radiation following nuclear rearrangement or the mass of radionuclides present, respectively. While their measurements is based on distinct components of the nuclear decay, radiometric and mass spectrometric determination are often complementary. For example, in the case of Pu measurement, ²³⁸Pu can be easily quantified by alpha spectrometry while ²³⁹Pu and ²⁴⁰Pu isotopes cannot be discriminated. In the other hand, measurement by mass spectrometry allows the separate quantification of ²³⁹Pu and of ²⁴⁰Pu, but detection of ²³⁸Pu is challenging without extensive sample preparation due to the isobaric interference caused by ²³⁸U, very abundant in environmental samples [2].

Unfortunately, in emergency response mode, most routine methodologies used for the environmental monitoring of radioactivity, though displaying excellent figures of merits, are labor intensive and have long turn-around time (TAT), making them ill-suited as emergency response radioanalytical procedures. For example, liquid scintillation and proportional counter methods used for the detection of ⁹⁰Sr in environmental samples are time-consuming, especially if secular equilibrium between ⁹⁰Y and ⁹⁰Sr needs to be achieved (ingrowth time up to 2–3 weeks) [3].

Over the last decades, several publications concerning rapid analytical methods for the quantification of alpha and beta emitters in environmental and biological samples have emerged (Tables 1 and 2). These rapid methods frequently use alpha spectrometry and gas proportional counter as detection techniques. These techniques have excellent figure of merits but frequently require sequential isolation of the analytes in order to avoid spectral interferences. In furtherance of this goal, extraction chromatography (EXC) has been applied extensively for the separation and purification of several radionuclides, especially the actinides. Proper EXC resin will provide adequate separation efficiency between the analytes and the rest of the constituents of the matrix [4,5]. Moreover, each EXC resin has specific affinities for several elements of similar chemical families (e.g., actinides, lanthanides, alkaline earth metals) and oxidation states. Resins such as DGA, TRU, TEVA and UTEVA have been used

for the extraction of actinides in environmental matrices [6,7] whereas Sr Spec and DGA have shown useful extraction properties for strontium [8,9]. As EXC can be performed with reduced solvent volume compared to ionic chromatography, which enhance preconcentration while reducing separation time. Mass spectrometric instrumentation such as inductively coupled plasma mass spectrometry (ICP-MS) has shown promises with respect to rapid and accurate determination of long-lived actinides. In addition, analytical procedures designed with ICP-MS as a detection system have shown comparable figures of merit and increase of analytical throughput compared to other radiometric and mass spectrometric approaches. Tables 1 and 2 summarize some of the recent published rapid methods to quantify actinides and radiostrontium, respectively by radiometric techniques and ICP-MS. While mass spectrometry has significant advantages over radiometric detection of numerous actinides, it is plagued by two major issues: its low tolerance to matrix effect, including the composition of the eluents, by, and the presence of interferences (Table 3). Fortunately, many of these drawbacks can be mitigated through the use of EXC, although designing a separation scheme for the critical radionuclides mentioned earlier is complex. Indeed, simultaneous determination of ⁹⁰Sr and actinides by ICP-MS has, to our knowledge, never been reported as difference in chemical behavior on EXC between the actinides and strontium prevent from finding a simple and effective compromise. Determination of low level of ⁹⁰Sr by ICP-MS is also challenging due to the high specific activity of ⁹⁰Sr and the presence of isobaric interferences (e.g., ⁹⁰Y, ⁹⁰Zr) originating from the sample matrix and ⁹⁰Sr decay. These issues remain to be properly addressed via enhancement in instrumental sensitivity and adequate separation before mass spectrometric measurement of this radionuclide can become routine [10,11].

The objective of this study was the development of a new, rapid and unique method for the determination of ^{234,235,236,238}U, ^{230,232}Th, ^{239,240}Pu, ²³⁷Np, ²⁴¹Am and ⁹⁰Sr in river water using a combination of TEVA and DGA resins. The use of eluents compatible with mass spectrometric approach, e.g., concentrations below 10 wt% for acids and below 0.2 wt% for salts, was also investigated in order to couple off-line the separation step to the measurement. This coupling between EXC separation and ICP-MS significantly reduces the turnaround time (TAT) as no previous chemical treatment would be needed before measurement.

Table 2

Overview of existing protocols to rapidly quantify actinides and strontium using ICP-MS measurement.

Radionuclides analyzed	Matrix	Separation columns used	Detection	Duration time	References
U, Pu, Am and Np	Sea water	TEVA Resin [®] , TRU Resin [®] , DGA Resin [®]	Alpha spectrometry ICP-MS	N.A. ^a	Maxwell et al. [9]
U, Pu, Am, Th and Np	Urine	TRU Resin [®]	ICP-MS	N.A. ^a	Hang et al. [15]
Th, Pu, Np, Am, and U	Soil and sediment	TEVA Resin [®] , DGA Resin [®]	Alpha spectrometry ICP-MS	N.A. ^a	Guérin et al. [6]
Th, Pu, Np, and U	Sediment	UTEVA Resin [®]	ICP-MS	9 min/sample	Perna et al. [5]
Pu, Np, Am, and U	Groundwater	TEVA Resin [®] , TRU Resin [®]	ICP-MS	20 min/sample	Henry et al. [16]

^a N.A.: not available.

Download English Version:

<https://daneshyari.com/en/article/1163810>

Download Persian Version:

<https://daneshyari.com/article/1163810>

[Daneshyari.com](https://daneshyari.com)