



Ultra-trace determination of ^{90}Sr , ^{137}Cs , ^{238}Pu , ^{239}Pu , and ^{240}Pu by triple quadruple collision/reaction cell-ICP-MS/MS: Establishing a baseline for global fallout in Qatar soil and sediments



Mohamed A. Amr ^{a, b, *}, Abdul-Fattah I. Helal ^b, Athab T. Al-Kinani ^c,
Perumal Balakrishnan ^a

^a Central Laboratories Unit, Qatar University, P.O. Box 2713, Doha, Qatar

^b Nuclear Physics Department, NRC, Atomic Energy Authority, Cairo, 13759, Egypt

^c Radiation Protection Section, Ministry of Environment, Doha, Qatar

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ABSTRACT

The development of practical, fast, and reliable methods for the ultra-trace determination of anthropogenic radionuclides ^{90}Sr , ^{137}Cs , ^{238}Pu , ^{239}Pu , and ^{240}Pu by triple quadruple collision/reaction cell inductively coupled plasma mass spectrometry (CRC-ICP-MS/MS) were investigated in term of its accuracy and precision for producing reliable results. The radionuclides were extracted from 1 kg of the environmental soil samples by concentrated nitric and hydrochloric acids. The leachate solutions were measured directly by triple quadrupole CRC-ICP-MS/MS. For quality assurance, a chemical separation of the concerned radionuclides was conducted and then measured by single quadrupole-ICP-MS. The developed methods were next applied to measure the anthropogenic radionuclides ^{90}Sr , ^{137}Cs , ^{238}Pu , ^{239}Pu , and ^{240}Pu in soil samples collected throughout the State of Qatar. The average concentrations of ^{90}Sr , ^{137}Cs , ^{238}Pu , ^{239}Pu , and ^{240}Pu were 0.606 fg/g (3.364 Bq/kg), 0.619 fg/g (2.038 Bq/kg), 0.034 fg/g (0.0195 Bq/kg), 65.59 fg/g (0.150 Bq/kg), and 12.06 fg/g (0.103 Bq/kg), respectively.

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1. Introduction

Radioactive fallout from past accidental releases of radionuclides during ground nuclear tests has contaminated the globe with radioactive materials. Most of these radio-contaminants are fission products of uranium and plutonium. Among these, ^{90}Sr , ^{137}Cs , ^{235}U , ^{238}U , ^{238}Pu , ^{239}Pu , and ^{240}Pu have been identified in some soils around the world. The half-lives of these radioisotopes are very long, reaching up to thousands of years. Therefore, radionuclides represent long-term health and environmental problems. Knowledge of the concentrations of these radioisotopes and their isotopic compositions in soil provide valuable information concerning nuclear activities in the affected regions. Several countries in the Middle East have started ambitious nuclear energy programs, and other countries in the area have declared their intention to generate electricity using nuclear power reactors in the near future. These

programs require routine assessments on the fallout activities in the area. To assess any future fallout contamination resulting from the increase in these nuclear power activities, a baseline should be established.

The most widely used analytical techniques for the determination of ^{90}Sr , ^{137}Cs , ^{238}Pu , ^{239}Pu , ^{240}Pu are radio-analytical techniques (Marzo, 2014; Amr and Abdel-Lateef, 2011). There is an increasing need to develop faster analytical methods for emergency response, including the analysis of environmental samples. The recent accident at the Fukushima Nuclear Power Plant in March, 2011 heightens the need to have fast analyses for radionuclides in environmental samples in the event of a nuclear accident (Maxwell et al., 2013). Inductively coupled plasma mass spectrometry equipped with collision/reaction cell (ICP-CRC-MS) has become a preferred method for the rapid and accurate determination of some radioisotopes in several matrices due to its low sample consumption, high sensitivity, excellent precision, and potential for multi-elemental analysis (Marzo, 2014). All the radiometric methods require previous chemical separation and pre-concentration in order to avoid interference from other radionuclides and problems

* Corresponding author. Central Laboratories Unit, Qatar University, P.O. Box 2713, Doha, Qatar.

E-mail address: amrewis@hotmail.com (M.A. Amr).

with self-absorption due to the presence of matrix elements (Marzo, 2014). ^{90}Sr is a by-product of nuclear fission found in nuclear fallout and presents a health problem since it substitutes for calcium (Ca) in bone, inhibiting removal from the body (Amr and Abdel-Lateef, 2011). There are a number of reported analytical methods that use ion exchange/extraction chromatography to determine ^{90}Sr by radio-analytical techniques. Vajda and Kim (2010) provide a very good overview of recent ^{90}Sr separation and analytical measurement techniques. Bojanowski and Skib (1990) have reported a method using fuming nitric precipitation, although handling fuming nitric acid is very tedious and time-consuming. Wang et al. (2004) reported a sequential method to determine actinides and ^{90}Sr in soil samples, and Ageyev et al. (2005) also described a sequential method for soil samples. After ashing the samples at 550 °C, the samples were leached with 8 M nitric acid, resulting in chemical yields for Sr of 50–70%. The method is, however, relatively complex and would not be considered a rapid method. Tavčar et al. (2005) leached Sr from large soil samples of up to 10 g using concentrated nitric acid. The leached solution was filtrated and evaporated, and the residue was re-dissolved in 1 M HNO_3 before being loaded on Sr-resin. Then, 100 mL of water was used to elute Sr from the Sr-resin, resulting in an average chemical yield of 67%. Recently, Maxwell et al. (2013) developed a new method for the determination of ^{90}Sr in large soil samples collected from Fukushima, Japan. The method allows rapid pre-concentration and separation of Sr in large soil samples for the measurement of Sr isotopes by gas flow proportional counting. Betti et al. (1996) measured the concentration of ^{90}Sr in soil, sediment, and grass (1 ppt) by glow discharge mass spectrometry (GDMS) and Wendt et al. (1997) measured the concentrations of ^{89}Sr and ^{90}Sr in soil, grass, milk, and urine by resonance ionization mass spectrometry (RIMS) after chemical separation. Solatie et al. (2002) measured the concentration of ^{90}Sr in soil samples near a nuclear facility by low-level liquid scintillation counting and reported its activity as being in the range of 6.2–96.5 Bq/kg. Maxwell et al. (2013) reported that the concentration of ^{90}Sr in large soil samples collected from the Fukushima Nuclear Power Plant is 1.35 Bq/kg. Ageyev et al. (2005) measured the concentration of ^{90}Sr , ^{238}Pu , $^{239,240}\text{Pu}$, ^{241}Am and ^{244}Cm in a set of layers of undisturbed sod-podzol soil samples collected in the 30-km zone around Chernobyl NPP, Ukraine. The determined concentrations were in the range of 420–135,000 Bq/kg. Desideri et al. (2003) measured the concentration of ^{90}Sr , ^{137}Cs , ^{238}Pu , $^{239+240}\text{Pu}$ and ^{241}Am levels in terrestrial and marine ecosystems around the Italian base in Antarctica. Asgharizadeh et al. (2009) measured the activity concentration of ^{90}Sr in soil and sediment samples from the southern shores of Iran. The reported ranges of the activity concentration of ^{90}Sr were 0.64–3.01 Bq/kg and 0.40–1.60 Bq/kg for soil and sediment samples, respectively. The ^{90}Sr concentration in soil samples from the southern shores of Iran is not uniform but varies from sample to sample and location to location. This non-uniformity may be attributed to their uneven and irregular distribution in the Earth's crust and also to various topographical and agricultural activities (Asgharizadeh et al., 2009). The average ^{90}Sr concentration in soil samples collected in Islamabad, Pakistan was reported as 4.20–4.37 Bq/kg (Jabbar et al., 2003). Al-Kheliewi et al. (2008) reported the concentration of ^{90}Sr , ^{137}Cs , ^{238}Pu , ^{239}Pu , and ^{240}Pu in surface marine sediments from the Al-Khafji, Mneefa, and Al-Jobail coastal areas in Saudi Arabia. The radioactivity levels of ^{90}Sr , ^{137}Cs , ^{238}Pu , ^{239}Pu , and ^{240}Pu indicated global fallout contamination due to nuclear weapons testing, as well as some contribution from the Chernobyl accident. Additionally, Al-Kheliewi et al. (2008) recommend that more surface marine sediments from the coastal areas along the Arabian Gulf be collected, measured, and compared with other values worldwide.

Furthermore, Al-Kheliewi and Shabana (2007) measured the activity concentration of some anthropogenic radionuclides in the surface marine sediments near the Ras Abu-Gamees area, east of Saudi Arabia and close to the southern border of Qatar. The reported concentration of radionuclides in that region are reasonably lower than or comparable to the levels in marine sediments collected from Ghazaouet Bay and from the Barcelona and Tarra-gona areas in the Mediterranean, respectively (Al-Kheliewi and Shabana, 2007). Al-Hamarneh et al. (2003) measured the activity concentration of ^{40}K , ^{134}Cs , ^{137}Cs , ^{90}Sr , ^{241}Am , ^{238}Pu and $^{239+240}\text{Pu}$ in surface and core soil samples collected from different regions of Jordan. Helal et al. (2004) reported that the $^{90}\text{Sr}/^{86}\text{Sr}$ and $^{240}\text{Pu}/^{239}\text{Pu}$ isotope ratios measured in soil samples collected near nuclear facilities confirm that the source of ^{90}Sr and Pu is global fallout. Al-Meer et al. (2013) reported a baseline for ^{90}Sr in Qatari soils.

Mass spectrometry techniques ICP-MS, AMS, RIMS, TIMS, and SIMS have been used for the determination of ^{135}Cs and ^{137}Cs (Taylor et al., 2008; Eliades et al., 2013; Hölggye et al., 2004; Ohno and Muramatsu, 2014; Chao and Tseng, 1996; Zheng et al., 2014a,b; Russell et al., 2014; Suseno and Prihatiningsih, 2014; Hancock et al., 2014; Pibida et al., 2001, 2004; Moreno et al., 1999; Lee et al., 1993; Edgington et al., 1991; Thakur et al., 2013). ^{137}Cs is a short-lived radionuclide (30.07 years). ^{137}Cs was released into the environment during nearly all nuclear weapon tests and some nuclear accidents, most notably the Goiânia accident, the Chernobyl disaster and the Fukushima Daiichi disaster (Marzo, 2014; Maxwell et al., 2013; Zheng et al., 2014a; Misumi et al., 2014). The interest in determining Pu isotopes in environmental samples results from dosimetric reasons in the case of accidents or releases, interest in its biogeochemical behavior in the environment, and tracing the source of the plutonium using the isotopic composition as a fingerprint (Steinhauser et al., 2014; Bu et al., 2014; Kaixuan et al., 2013; Srencik et al., 2014; Shozugawa et al., 2012). The Pu isotopes of greatest interest in environmental samples are ^{238}Pu , ^{239}Pu , ^{240}Pu , and ^{241}Pu (Warneke et al., 2002; Rodushkin et al., 1999; Muramatsu et al., 2001; Kershaw et al., 1995; Koide et al., 1985; Dai et al., 2002; Buesseler, 1997; Aldridge et al., 2003). Alpha spectrometry is used for the determination of ^{238}Pu and $^{239+240}\text{Pu}$. Due to the similar energies of alpha particles from ^{239}Pu (5.16 MeV) and ^{240}Pu (5.17 MeV), α -spectrometry can only measure the sum activity of ^{239}Pu and ^{240}Pu . Spectrometric interference in the α -spectrometric determination of ^{238}Pu and $^{239+240}\text{Pu}$ are mainly from ^{214}Am , ^{210}Po , ^{224}Ra , ^{229}Th , ^{231}Pa , ^{232}U , and ^{243}Am . In addition, the matrix elements must also be removed to improve the resolution (Armstrong et al., 2014; Testa et al., 1998). Therefore, Pu must be separated from the matrix and other interfering radionuclides before measurement. Co-precipitation, solvent extraction, ion exchange chromatography and extraction chromatography are often used for the separation of Pu (Hamilton et al., 1994; Lee et al., 1998; Sam et al., 1998; Heldal et al., 2002; Hernandez et al., 1998; Matishov et al., 1999).

1.1. Objectives of this study

The artificial radionuclides can be released into the environment through a variety of different processes. The most relevant sources of anthropogenic radioisotopes are global fallout from atmospheric weapons tests, local fallout accidentally released from nuclear power plants and spent-fuel reprocessing plants, and short-lived nuclides from the production of radioisotopes for various applications. It should be noted that no investigations have been carried out regarding environmental radioactivity in Qatar. Consequently, it is essential to have accurate, reliable, and precise analytical methods to measure their concentration and isotopic ratios. To

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