Applied Radiation and Isotopes 81 (2013) 201–207

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Application of ICP-DRC-MS to screening test of strontium and plutonium in environmental samples at Fukushima

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HIGHLIGHTS

- ICPMS is available for assessing Sr-90 and Pu in Fukushima samples.
- Automated SPE separation device was coupled to ICP-DRC-MS.
- The obtained LOD levels enabled us to use the system as the screening test tool.

ARTICLE INFO

Available online 2 April 2013

Keywords:
^{90}Sr
Pu isotopes
Fukushima Daiichi nuclear power plants (FDNPP)
Inductively coupled plasma mass spectrometry (ICPMS)
Automated solid phase extraction (SPE) equipment

ABSTRACT

An application of sequential automated SPE separation equipment coupled to the quadrupole-based ICPMS instrumentation with a dynamic reaction cell such as a screening test system of ^{90}Sr and Pu isotopes in environmental samples was developed in this work. So far, during the course of a large number of reports as to various specific radioactivities in environmental samples surveyed at radioactive contaminated area around the Fukushima Daiichi Nuclear Power Plants (FDNPP), there is a much smaller number of reports on ^{90}Sr and Pu isotopes than that of ^{134}Cs and ^{137}Cs since the FDNPP accident, and then it would be expected to develop the simple analysis method of these isotopes instead of radiation measurements currently in use. In particular, a screening for ^{90}Sr in environmental samples has been accomplished using an isotopic ratio measurement mode in comparison with the characterization on the Solid Phase Elution (SPE) separation between strontium and zirconium isotopes around the mass-90 fraction. As a result, for a trial analysis of environmental samples of a muddy snow water and a soil which were collected at Fukushima, it was found that the present developed system makes it applicable for achieving up to the specific activity levels of several hundreds Bq/kg (^{90}Sr) and about 1–2 Bq/kg (Pu isotopes) as the screening test system.

1. Introduction

The serious accident of the Fukushima Daiichi Nuclear Power Plants (FDNPP) was caused due to a gigantic tsunami followed by the East Japan Earthquake, where the epicenter was located in the northwest Pacific about 130 km off northeastern Japan, at 14:46 on March 11, 2011. As it turned out, the FDNPP lost the cooling system of nuclear reactors, resulting in hydrogen explosions destroying the external nuclear reactor buildings on March 12 and 14 in Unit 1 and 3 reactors. These explosions of the FDNPP caused massive releases of various radionuclides (^{131}I, ^{137}Cs, ^{134}Cs, ^{136}Cs, and ^{132}Te) into the atmosphere, which are originated when the nuclear power plants have usually operated to generate electricity. These released high volatility radionuclides were carried together with the air parcel like a plume of smoke, and caused accumulation of them on the ground to the northwest direction from the FDNPP. The fact has been borne out by the air dose monitoring data and the accumulation distribution map of radioactivities on the ground reported by the MEXT (MEXT: Japanese Ministry of Education, Culture, Sports, Science and Technology) and the atmosphere

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http://dx.doi.org/10.1016/j.apradiso.2013.03.056

dispersion simulation data by SPEEDI (System for Prediction of Environmental Emergency Dose Information) (MEXT, 2011) in the region of Fukushima and its adjacent prefectures.

So far, having estimated based on observed radiation dose data at survey points around FDNPP, Nuclear and Industrial Safety Agency have reported that total activities of released airborne radioactive cesium isotope (^{137}Cs) would amount to about 15 PBq, and also that total activities of strontium (^{90}Sr) would be two digits smaller than that of ^{137}Cs (METI, 2011). As compared with chemical sample preparations for radiation measurements of β- and α-emitted nuclides of strontium and plutonium, the quantitative analysis for radioactivities of cesium isotopes have been accomplished easily and rapidly due to an already established γ-ray measurement system equipped with an HPGe detector and various specific radioactivities, e.g. agricultural procedure and marine products, have been reported almost everyday by JAEA, Fukushima prefecture, many local governments and so on. However, there is not only a few quantitative analytical reports for the radioactivities of strontium (^{90}Sr) and plutonium (^{239}Pu and ^{240}Pu) using radiation measurements, but also a few administrative services as the screening test for these isotopes, and then there is a need to develop a routine approach and a rapidly simple method which are complementary to radiation measurements currently in use.

Today, an ICPMS analysis plays an important role to measure quantitatively trace and macro-amounts of metal elements in environmental radioactivity, biology, medicine, and materials. It seems that the ICPMS technique in nuclear industry would be expected to reach maturity of a more routine approach required easier sample preparation. The purpose of our work is to develop the application of our quadrupole-based ICPMS system with Dynamic Reaction Cell (ICP-DRC-MS) and automated Solid Phase Elution (SPE) such as an integrated screening test system rapidly and easily analyzing the limit of detection (LOD) levels of ^{90}Sr and Pu isotopes into various environmental samples. For the discrimination of ^{90}Sr, using an isotopic ratio measurement mode installed in the ICPMS system, the LOD levels of ^{90}Sr in environmental samples using the present screening test system have been discussed as compared with the each ion count rate for strontium and zirconium isotopes around the mass-90 fractions isolated by the SPE separation.

2. Experimental

2.1. ICPMS analysis is compared with radiation measurement in nuclear and radiochemistry

Recently, as instrumental analysis of mass spectrometry has continued to gain sensitivity and reliability, an ICPMS analysis, in particular the quadrupole type of ICPMS, has become increasingly useful in the measurement of radionuclides (Jarvis et al., 1992). The optimization of ICPMS is improving our ability to detect the number of radionuclide's atoms, and it allows the near-complete isotopic ratio analysis of any form of sample. As a radioactivity of radionuclide is compared with the number of the atoms, we have known that a radioactive species decays according to an exponential law: $N{=}N_0\text{e}{-}\lambda t$, where N represents the number of atoms, at time t, and N_0 the corresponding quantities when $t{=}0$, and λ is the characteristic decay constant for the species. It has been also known that the radioactivity is proportional to the number of atoms as expressed in the following equation:

$$A=\lambda N=\left(\frac{\ln 2}{T_{1/2}}\right)N \tag{1}$$

where $T_{1/2}$ is the half-life of radionuclide. Fig. 1 shows the relationship (Ohtsuka, 2006) between radioactivity and the number of atoms around the region of 1 mBq. The logarithm of radioactivity is plotted as a function of that of the number of atoms in Fig. 1. The ratio between both the logarithms has the constant slope derived from Eq. (1). That is expressed as the thick solid line, suppose that its line is taken on the condition of 1 mBq radioactivity and 106 atoms. The eight slanting lines represent half-lives of 101 to 107 years with the interval of 10-digit number and they are also derived based on Eq. (1). Total radioactivities of ^{239}Pu and ^{240}Pu including in several soil samples at sites of FDNPP, due to the serious nuclear accident in Japan, had been reported as the detection data of about 10 mBq level per the soil 100 g (~0.1 Bq/kg level) in the rapid communication on 28 March, 2011 by Ref. Tokyo Electric Power Company (2011). From Fig. 1, considering the detection limit of our ICPMS system developed in the present work has approached about 107 atoms, it seems that it will be reasonably sufficient to identify those plutonium isotopes and to determine the specific activities. In addition, we have developed an automatic SPE equipment coupled to the ICP-DRC-MS system to purify and concentrate the elements of interest into matrices using InertSep ME-1 chelate (Sr) and UTEVA (Pu) resins effectively.

2.2. Sample preparation

In this study, two environmental samples (a snow muddy water sample of 10 mL for ^{90}Sr and a soil sample of about 0.3 g for Pu isotopes) were used for verifying the validity of our developed screening test system of ^{90}Sr and plutonium isotopes. For the snow muddy water, its sample was collected at Kawamata-cho in Fukushima on April 25, 2011, where is located at about 40 km northwest direction away from FDNPP. As the sample solution had contained a small amount of slightly soluble slurry materials, the solution was filtrated using a 0.45 μm pore size tetrafluoroethylene polymer (PTFE) membrane filter of 25 mmΦ ID (inside diameter) and obtained as the pre-concentration solution of 10 mL, and then the solution of 5 mL was diluted by 100 mM ammonium acetate up to a total volume of 10 mL. And while, the soil sample was collected at Akauki zone at Namie-cho in Fukushima on May 1, 2012, where it is located at about 20 km northwest direction away from FDNPP. The dried soil sample of 0.344 g was digested by acid combinations of $HN0_3$ (69%, Ultrapur-100, Kanto Chemicals) 5 mL, HCl (36%, Ultrapur-100, Kanto Chemicals) 2 mL, and HF (49%, Ultrapur-100, Kanto Chemicals) 3 mL under microwave radiation. The sample was taken into the exclusive PTFE vessel (XF100, Perkin-Elmer) of the microwave oven (Multiwave3000, Perkin-Elmer), and the microwave program was performed in accordance with a recommended program on the microwave oven system. The acid digested solution was diluted up to a volume of 50 mL using purified water (Easypure RoDi, Barnstead). A residual slightly soluble fraction in the solution was distinguished by a decantation and only the supernatant solution as an analyte was pre-concentrated before an automated SPE separation.

2.3. Sequential automated SPE separation equipment

Sequential automated SPE separation equipment of Gilson's LH (Liquid Handler) GX-271 was set up with the ICPMS instrumentation at the same laboratory room. This automation allows reduction of about 50% in chemical separation time than that of the previous system, and this equipment can often treat a large number of environmental samples. The sample loaded into a sealed 6 mL column can be operated by an automatic air aspiration and dispense, and this probe tip of carbonate syringe can detect a liquid surface sliding down. The LH GX-271 system had incorporated an Eichrom UTEVA™ column (the resin dry weight of 0.6 g) specialized to isolate Pu isotopes from uranium, and also done a GL sciences InertSep ME-1 column (the resin dry weight of 0.25 g) in order to eliminate mainly alkali and alkali

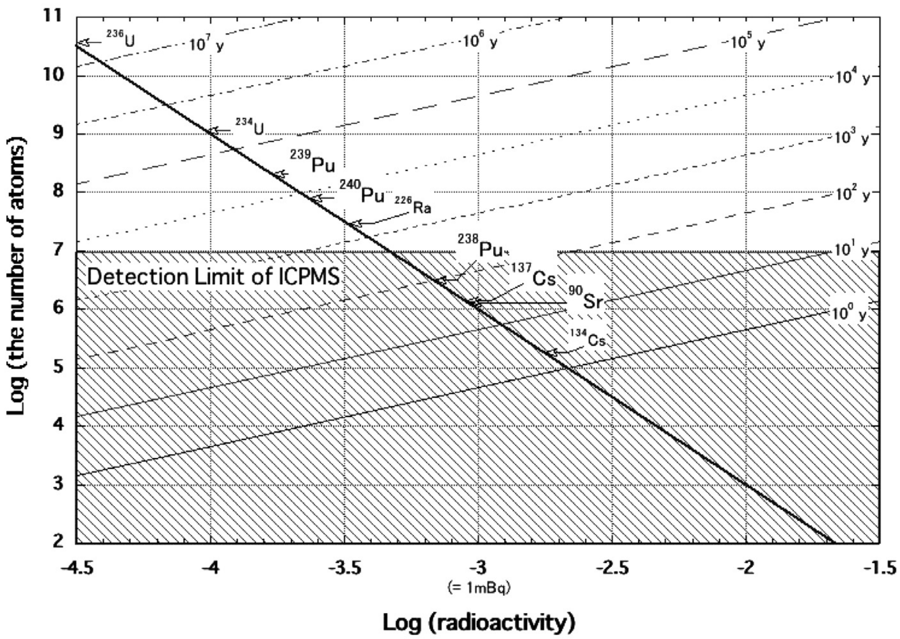

Fig. 1. Relationship between a radioactivity and the number of atoms around the region of 1 mBq. The insert of detection limit of ICPMS is shown as a typical performance of quadrupole ICPMS (Ohtsuka, 2006).

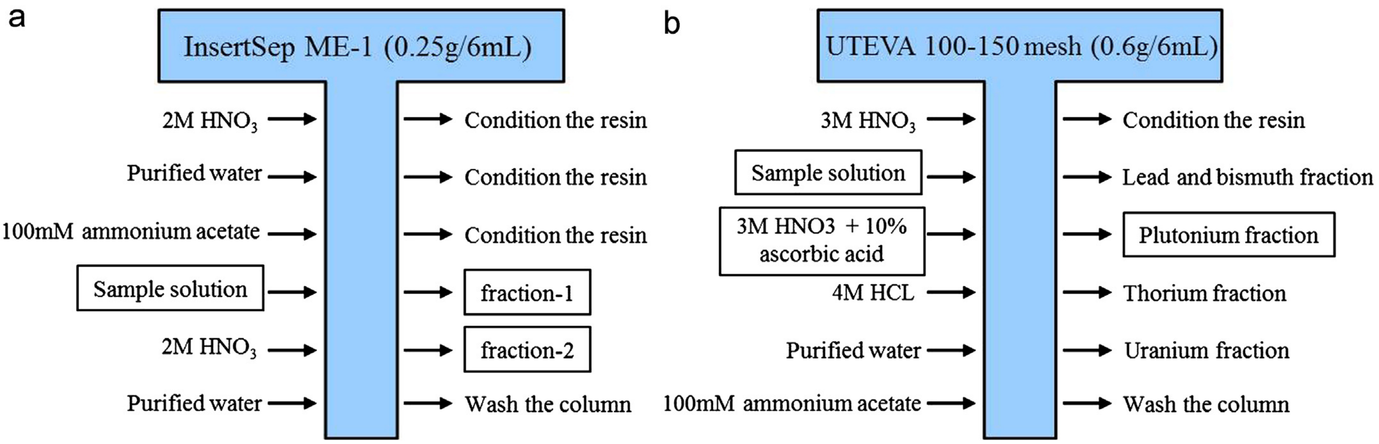

Fig. 2. SPE separation processes of (a) strontium and (b) plutonium elements incorporated in the LH GX-271 system (automated SPE separation system).

earth elements such as ^{90}Sr. Each size of both the exclusive columns was 6 mL and the columns were set up with a disposal elution column port moving automatically on collection tubes in the equipment. Fig. 2(a) and (b) shows SPE separation processes of strontium and plutonium elements incorporated in the LH GX-271 system (automated SPE separation system). Through all the sample preparation and SPE separation processes, a sample blank dealt with the present measurement was controlled with only an estimation of reagent contamination.

2.4. ICPMS instrumentation equipped with Dynamic Reaction Cell (DRC) and detection capability

In the present work, Perkin-Elmer ELAN® DRC II as an ICPMS instrumentation was used for developing the screening test system of low levels of ^{90}Sr and Pu isotopes as well as discriminating these isotopic ratios of interest in environmental samples. The ICPMS instrumentation combines the power of patented DRC technology, performance-enhancing Axial Field™ Technology (AFT) and an high-performance sample introduction system with the ability to run any reaction gas.

The result is the ultimate analytical tool, providing uncompromised sensitivity and performance in all matrices for the almost kind of difficult applications on the base of minimizing background equivalent concentrations. The DRC technology also has contributed to maintaining analyte sensitivity and providing ultra- and trace-level detection limits in a complex any sample due to eliminating poly-atomic interferences. In the present works, several improvements were made for our ICP-DRC-MS system. A low flow rate (about 100 μL min−1) of the PFA-100 micro-flow nebulizer (CETAC Technologies, Inc.) was utilized for the sample introduction in order to improve the LOD of strontium and plutonium elements, which enhanced those ionization efficiencies into a plasma discharge due to a fine-droplet aerosol, resulting in improving the LOD with its about one digit smaller.

For detection capability relative to LOD in the present system, we have referred to the most accurate way in general of assessing instrument detection capability in Ref. Thomas (2007) based on Ref. Currie (1968). Based on the signal-to-background noise observed by the present instrument, and for a 99% confidence level the LOD is typically 3 × standard deviation (SD) of several replicates of the sample blank and is calculated in the following manner:

$$\mathrm{LOD} = \frac{3 \times (\text{Standard Deviation of Background Signal})}{(\text{Analyte Intensity}-\text{Background Signal})} \times (\text{Analyte Concentration}) \tag{2}$$

The LOD values of strontium and plutonium were evaluated with assessing the observed signal intensities of analyte concentrations of 1 μg L^{-1} and backgrounds of strontium and uranium (as quasi-plutonium, because there are no controlled facilities for plutonium in our university.) in the mass spectra around 90 and 239 fractions. And also in Ref. Currie (1968), the critical level of L_c is defined as the following equation:

$$\mathrm{LOD} = k^2 + 2L_c,\ (L_c = k\sigma\ ) \tag{3}$$

where k is abscissa of the standard normal distribution of σ corresponding to probability level, is used for deriving whether or not those signal intensities in the each mass fraction indicate detections. Having defined the limit of detection on the instrumentation as a screening tool, it will be stated that mass biases for strontium and zirconium in the isotopic ratio measurement are corrected by those external standard reference materials (SPEX CertiPrep® Claritas PPT Grade) which are evaluated in advance as each base isotopic ratio. Optimized experimental conditions are summarized in Table 1. Further details related to the ICP-DRC-MS system are described elsewhere (Sakama et al., 2011). About uptaking all sample solutions to the micro-flow nebulizer in the instrumentation, the ADX-500 automated sampler (CETAC Technologies, Inc.) into a clean box coupled to the ICP-DRC-MS was used to avoid some contaminations originated from manual sample injection. The automatic sampler was connected with the PFA-100 micro-flow nebulizer via the PTFE sample probe tube of 0.3 mm ID that was suitable for the low flow rate experimentally. Its system allows a typical uptake time up to the nebulizer entrance to be the optimized period of about 60–70 s by controlling the peristaltic pump set up at the ICP-DRC-MS instrumentation.

3. Results and discussion

3.1. Screening for ^{90}Sr

The limit of detection, accuracy and precision on screening environmental samples for ^{90}Sr discrimination are mainly affected by the occurrence of isobaric atomic and molecular ions at mass-90 fraction (m/z=90). In terms of the problem, in particular, it should be necessary to estimate the most important isobaric interference of ^{90}Zr+ due to the high abundance of environmental soils and slurries relatively. In Table 2, current known stable isotopes of strontium and zirconium elements and their isotopic abundances and isotopic ratios referred to IUPAC recommended values (Bievre and Taylor, 1993) are summarized. Fig. 3(a) and (b) shows the mass spectra of the first elution (described as fraction-1 below) and the second one (fraction-2), respectively, around a mass-90 and the adjacent regions obtained via analyzing the filtrate solution (10 mL) of a snow muddy water sample (that is Kawamata sample) collected at Kawamata-cho in Fukushima by our developed sequential automated SPE separation and ICPMS measurement. In the mass spectrum of fraction-1 in Fig. 3(a), we have found that as those stronger ion count rates of ^{84,86,87,88}Sr, ^{85}Rb and ^{95}Mo isotopes are associated with the group of alkali, alkali earth and molybdenum elements, their elements are hard to absorb at an InertSep ME-1 chelate resin in 100 mM ammonium acetate based solution, and their elements also have eluted immediately in the fraction-1. In contrast, in the mass spectrum of fraction-2 in Fig. 3(b), the other isotopes (zirconium together with those residual strontium and molybdenum) would be eluted from the resin by 10 mL of 2 M HNO_3. This observation indicates that the fundamental characteristic of InertSep ME-1 is attributable to discriminating the elements of interest from alkali and alkali earth ones on the function of chelating interaction by an iminodiacetic acid group selectively (Miura et al., 2000).

In Fig. 3(a) and (b), focusing the signal intensities around the mass-90 region if the contribution of ^{90}Sr has included in the mass fraction or not, and we should have verified the variations of strontium and zirconium isotopic ratios as compared with those isotopic ratios of fraction-1 and fraction-2 respectively. The isotopic ratio analysis mode was used for these fractions with the peak-hopping approach at the present ICP-DRC-MS instrumentation, as the best possible detection limits and accuracies were required for strontium and zirconium elements. Fig. 4 provides a plot of each zirconium isotopic ratio (^{91}Zr/^{90}Zr, ^{94}Zr/^{90}Zr, and ^{96}Zr/^{90}Zr) against fraction-1, fraction-2, and reference respectively. Fig. 5 shows a plot of ^{86}Sr/^{88}Sr against ^{87}Sr/^{88}Sr for strontium. These references of isotopic ratios of zirconium and strontium elements have been calculated by IUPAC recommended isotopic abundances (Bievre and Taylor, 1993). In Figs. 4 and 5, it was found that the isotopic ratios of ^{91}Zr/^{90}Zr, ^{94}Zr/^{90}Zr, ^{86}Sr/^{88}Sr, and ^{87}Sr/^{88}Sr are almost equal to be those corresponding references on the discriminating between alkali earth elements (including Sr) and the others (Zr) using the InertSep ME-1 chelate

Table 1
Optimized experimental conditions of the present ICP-DRC-MS system (Perkin-Elmer ELAN® DRC II).

	^{90}Sr and Pu isotopes measurements
RF power (W)	1500
Solution uptake rate (μL min−1)	100–115
Plasma gas flow rate (L min−1)	15
Auxiliary gas flow rate (L min−1)	1.40
Nebulizer gas flow rate (L min−1)	1.10–1.11
Lens voltage (V)	5.25
Cell rod offset at standard mode (V)	−11
Quardrupole rod offset at standard mode (V)	0
Cell path voltage (V)	−15
RPq	0.25
Axial field voltage (V)	200
Dwell time (ms)	50
Number of sweeps	40
Number of readings	1
Number of replicates per sample	3
Sampler cone	Platinum, 1.1 mms orifice diameter
Skimmer cone	Platinum, 0.9 mm orifice diameter

resin. In contrast, for the ^{96}Zr/^{90}Zr in Fig. 4, we have deduced that the apparent different isotopic ratio from the reference is attributable to an insufficient ion counts of ^{96}Zr statistically at the mass spectra shown in Fig. 3 due to the lower isotopic abundance (the isotopic abundance of ^{96}Zr is 2.8%). Table 3 summarizes that the isotopic ratios of ^{88}Sr/^{90}Zr(^{90}Sr) and ^{91}Zr/^{90}Zr(^{90}Sr) contained in Kawamata sample were experimentally obtained between fraction-1 and fraction-2 by the isotopic ratio analysis mode mentioned above. Comparing the isotopic ratio of ^{91}Zr/^{90}Zr in fraction-1 and its ratio in fraction-2, the difference in isotopic ratios are much smaller and their obtained isotopic ratios are approximately consistent with its reference.

The present result data will be also supported by the nearly consistent decontamination factors of ^{90}Zr and ^{91}Zr in fraction-2 as follows. These factors of ^{90}Zr and ^{91}Zr have been measured to be 59.8% and 59.4%, respectively. In addition, Table 3 shows the isotopic ratio of ^{88}Sr/^{90}Zr in fraction-1 is not less than a factor of 8 over that in fraction-2, and while the decontamination factor of ^{88}Sr in fraction-2 also has been evaluated to be 95.4% due to the effective separation property for an alkali earth element of the present InertSep ME-1 resin. That is, assuming that ^{90}Sr isotope originated from the FDNPP accident is included in environmental samples such as the present Kawamata sample at Fukushima upon the LOD level in this ICP-DRC-MS instrumentation, it can be deduced that the isotopic ratios of ^{91}Zr/^{90}Zr between fraction-1 and fraction-2 are not in agreement clearly, and that the isotopic ratio of ^{88}Sr/^{90}Zr in fraction-1 in not more than a factor of about 0.4 below that in fraction-2. In the present work, we note that the LOD of mass-90 fraction has been evaluated to be 0.6 ng L^{-1} (L_c=0.3 ng L^{-1}) corresponding to several hundreds Bq/kg. As a result, it may be concluded that ^{90}Sr has not been contained in the Kawamata sample at Fukushima with the LOD level or much less at all. Examining the screening test of Sr-90 at the present ICP-DRC-MS system, the isotopic ratio variation in the each fraction eluted from the InertSep ME-1 chelate resin (as a SPE method) is available for estimating the survey of ^{90}Sr originated from the FDNPP accident. It will be possible to reduce chemical separation and analysis time (about 50% reduction) without complicated and difficult experimental treatments and with nearly sufficient accuracy.

Table 2
Stable isotopes of strontium and zirconium elements and their isotopic abundances and isotopic ratios are listed in the IUPAC recommended values (Bievre and Taylor, 1993).

Element	Mass number	Isotopic abundance/%	Isotopic ratio
Sr	84	0.56	0.007
	86	9.86	0.119
	87	7	0.085
	88	82.58	1
	90 (^{90}Sr)	Not needed	Not needed
Zr	90	51.45	1
	91	11.22	0.218
	92	17.15	0.333
	94	17.38	0.337
	96	2.8	0.054

3.2. Screening for plutonium isotopes

For the screening for plutonium isotopes in this work, because of the presence of uranium, thorium, lead and bismuth in an analyte, the accurate determinations of ^{239}Pu and ^{240}Pu using our developed ICP-DRC-MS instrumentation are difficult due to interferences from $^{238}U^{1}H^{+}$ (m/z=239) ion formation and the peak tailing effect from $^{238}U^{+}$ and from unwanted oxide and hydride ions contributed to lead, bismuth and heavy metal elements not only uranium and thorium. These interferences led to an increase in the background signal around the mass-239 and 240 fractions, necessitating the sequential automated separation of plutonium from various elements such as uranium, thorium, lead and bismuth for environmental samples at Fukushima. The present trial results will help the installation of the screening test system for plutonium in so many environmental samples. The separation of Pu was performed by the automated chemical separation equipment set up a SPE column using an UTEVA resin as stated in Section 2.4. Fig. 6(a) and (b) shows the mass spectra of a soil sample collected at Akauki zone at Namie-cho in Fukushima. Fig. 6(a) is the mass spectrum in the mass-220 to 245 region of the sample solution before eluting it to the UTEVA resin, and Fig. 6(b) is also the mass spectrum in the same mass region of the sample elution through the UTEVA resin by 3 M HNO_3 added 10% ascorbic acid. Comparing both the background signals shown in Fig. 6(a) and (b), it can be stated that those interferences are eliminated effectively from the sample elution using the present automated separation system and it allows the more lower limit levels of detection for plutonium isotopes in Fukushima

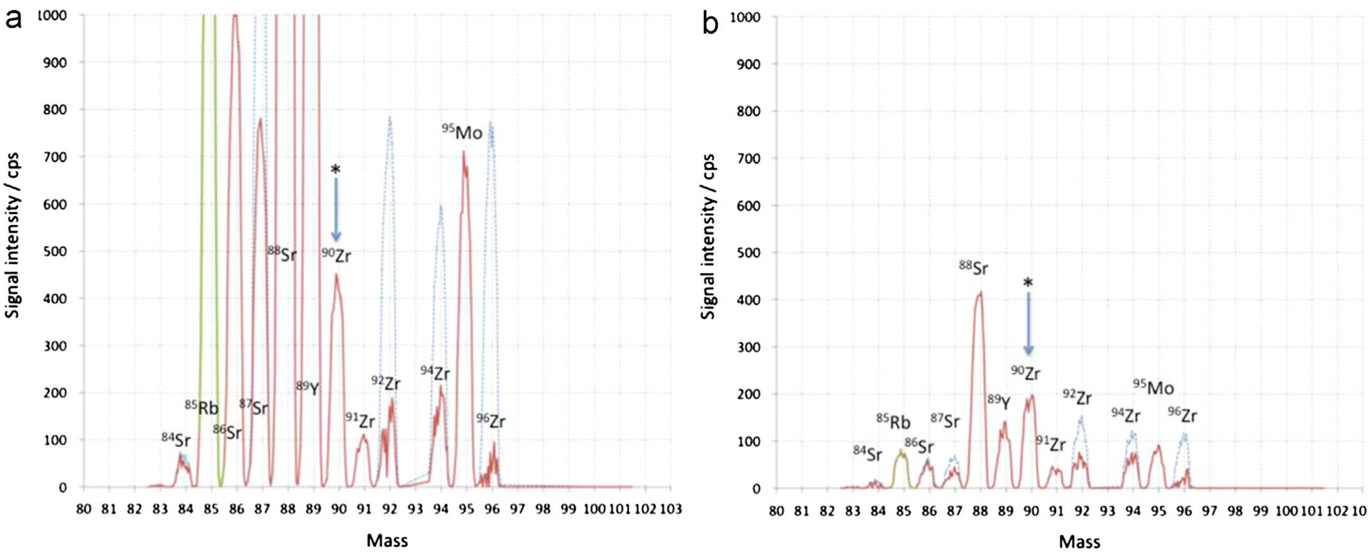

Fig. 3. Mass spectra of (a) the first elution (fraction-1 as described in Section 2.3) and (b) the second elution (fraction-2 as described in Section 2.3).

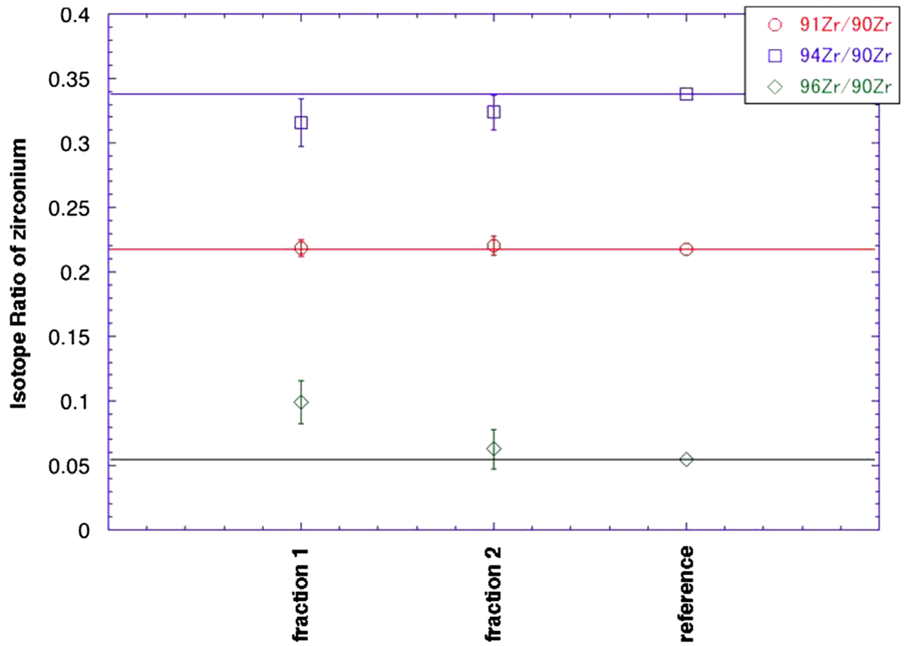

Fig. 4. A plot of each zirconium isotopic ratio ($^{91}Zr/^{90}Zr$, $^{94}Zr/^{90}Zr$, and $^{96}Zr/^{90}Zr$) against fraction-1, fraction-2, and reference.

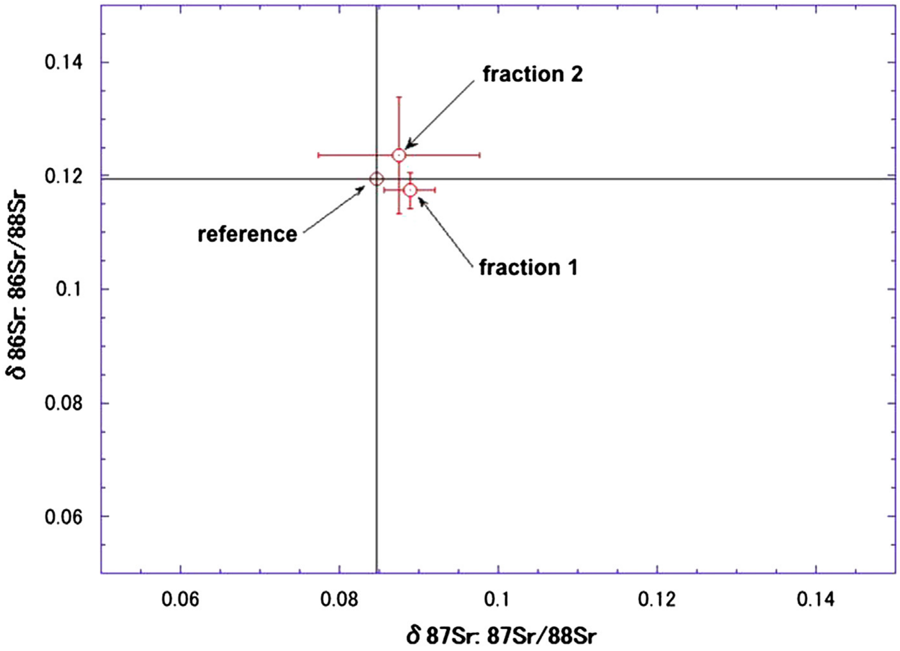

Fig. 5. A plot of $^{86}Sr/^{88}Sr$ against $^{87}Sr/^{88}Sr$ for strontium.

Table 3
Isotopic ratios of $^{88}Sr/^{90}Zr(^{90}Sr)$ and $^{91}Zr/^{90}Zr(^{90}Sr)$ contained in Kawamata sample in comparison with fraction-1, fraction-2 and reference.

Isotopic ratio	Fraction-1	Fraction-2	Reference
^{88}Sr / ^{90}Zr (^{90}Sr)	19.015	2.186	Not needed
^{91}Zr / ^{90}Zr (^{90}Sr)	0.235	0.237	0.218

samples to be discriminated as trail functions as the screening test system. As a result, it may be concluded that decontamination factors of uranium and thorium in this eluted solution of Akauki sample have reached to be not less than 95%, and also that the LOD of plutonium isotopes evaluated from a mass bias of uranium have been deduced to be about 0.02 ng L^{-1} (L_c=0.009 ng L^{-1}) corresponding to about 1–2 Bq/kg in the ICP-DRC-MS instrumentation.

4. Conclusion

We have developed the application of sequential automated SPE separation equipment coupled to the quadrupole-based ICPMS instrumentation with a dynamic reaction cell such as a screening test system of ^{90}Sr and Pu isotopes in environmental samples. The screening system is specifically designed to allow the screening test system based on a simple and rapid analysis method of

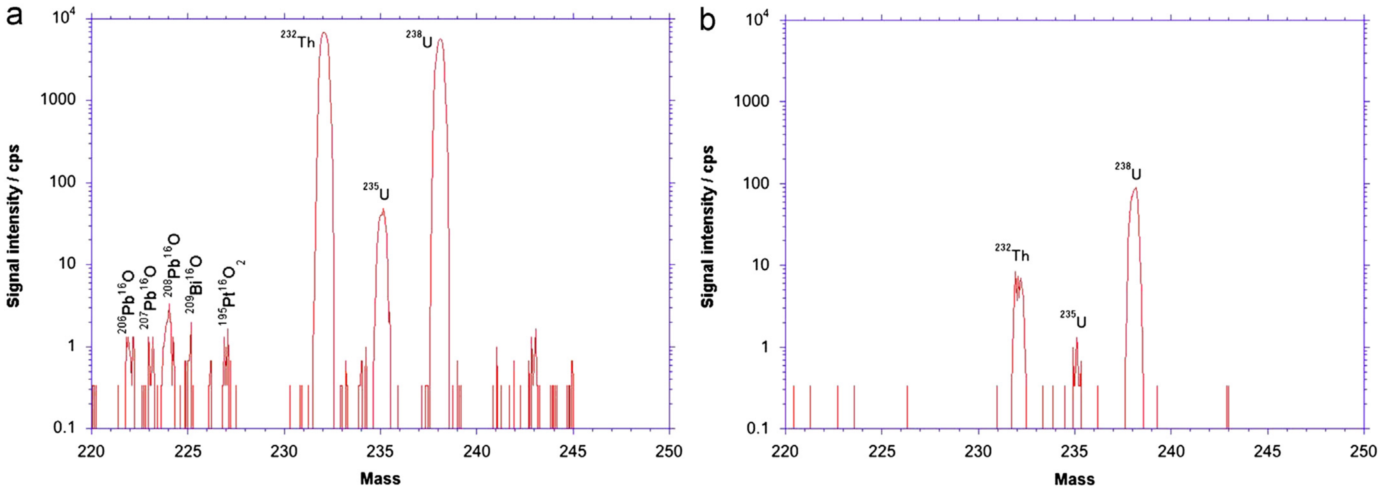

Fig. 6. Mass spectra of a soil sample collected at Akauki zone at Namie-cho in Fukushima. (a) is the mass spectrum in the mass-220 to 245 region of the sample solution before eluting it to the UTEVA resin, and (b) is the mass spectrum in the same mass region of the sample elution through the UTEVA resin by 3 M HNO_3 added 10% ascorbic acid.

these isotopes associated with the FDNPP accident instead of radiation measurements currently in use.

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