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Isotopic analysis of uranium and plutonium using ICP-MS and estimation of burn-up of spent uranium in contaminated environmental samples†

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Environmental monitoring of actinides and evaluation of the contamination source (nuclear weapons tests, nuclear power plant and nuclear reprocessing plant accidents, *etc.*) requires precise and accurate isotopic analysis of actinides, especially uranium and plutonium. Double-focusing sector-field inductively coupled plasma mass spectrometry (ICP-SFMS) using a low-flow microconcentric nebulizer with membrane desolvation, "Aridus", was applied for isotopic measurements of uranium and plutonium at the ultratrace level. The detection limit (3σ) for ^{236}U and ^{239}Pu after chemical extraction was 0.2 pg l^{-1} in aqueous solution and 0.04 pg g^{-1} in soil, respectively. ^{235}U/^{238}U, ^{236}U/^{238}U and ^{240}Pu/^{239}Pu isotope ratios were measured in soil samples collected within the 30 km zone around the Chernobyl nuclear power plant. The average ^{240}Pu/^{239}Pu isotope ratio in contaminated surface soil was 0.396 ± 0.014. The burn-up grade and the portion of spent uranium in the spent uranium/natural uranium mixture in soil were calculated using the iteration method. A slight variation in the burn-up grade of spent reactor uranium was revealed by analyzing ^{235}U/^{238}U and ^{236}U/^{238}U isotope ratios. A relationship between the ^{240}Pu/^{239}Pu isotope ratio and burn-up of spent uranium was observed.

Introduction

At present, the median concentration of plutonium produced as a result of nuclear weapons tests is about 10^{-13} g g^{-1} in soils of the Northern hemisphere.[1] Considerably higher concentrations of plutonium and other actinides have been found in regions contaminated with accidental fallout from nuclear reprocessing plants and nuclear power plants, *e.g.*, in the Ural region (Russia) and Chernobyl (Ukraine).[1,2] Plutonium is represented in the Chernobyl fallout by five radionuclides; four of them (^{238}Pu, ^{239}Pu, ^{240}Pu and ^{242}Pu) are long-lived alpha emitters and ^{241}Pu is a beta emitter with a half-life of $T_{1/2}$ = 14.4 years.[3] Along with plutonium isotopes, ^{236}U ($T_{1/2}$ = 2.342×10^{7} years) is produced from ^{235}U *via* the (n,γ) reaction $^{235}U + n \rightarrow {}^{236}U + \gamma$, competing with the neutron-induced fission reaction. In spent nuclear fuel the ^{236}U/^{238}U isotope ratio can reach from 2×10^{-3} to 5×10^{-3} depending on the reactor type and fuel burn-up history,[4,5] *i.e.*, higher by several orders of magnitude than the ^{236}U/^{238}U ratio in non-contaminated natural uranium ores (an upper limit of about 6×10^{-10} was found for the ^{236}U/^{238}U isotope ratio in uranium ore samples[6–9]). Thus, knowledge of the isotopic composition of artificial actinides is important for evaluating their origin in the environment. In addition, the isotopic composition of uranium and plutonium allows the burn-up of spent nuclear fuel contaminating the environment to be determined. Burn-up is the most important characteristic of spent nuclear fuel that indicates the degree of utilization of uranium and also the amount of fission products and minor actinides produced during reactor operation.[10] As applied to the fuel of the destroyed fourth block of the Chernobyl nuclear power plant (NPP), this value allows the spread of radionuclides from different core zones to be evaluated, thus revealing the processes in the reactor core during the active stage of the Chernobyl NPP accident.

Sector-field inductively coupled plasma mass spectrometry (ICP-SFMS) is one of the most suitable methods for the isotopic analysis of actinides at ultratrace concentration levels, due to its high sensitivity, good accuracy and precision and the mostly simple sample preparation procedure needed.[11] However, the formation of the uranium hydride ions ($^{235}U^{1}H^{+}$, $^{238}U^{1}H^{+}$) and isobaric interferences caused by molecular ions, *e.g.*, of lead or rare earth elements that can be expected in the actinide mass range (PbO_2^{+}, PbN_2^{+}, *etc.*),[12] are limiting factors for the isotopic measurement of low concentrations of uranium and plutonium. Therefore, matrix separation and the use of nebulizers with desolvators are necessary to reduce molecular ions and improve detection limits of the actinide isotopes mentioned above.[9,12–14]

The aim of this work is to evaluate the application of a rapid and ultra-sensitive isotope analytical procedure based on ICP-SFMS for the determination of the uranium and plutonium isotopic composition and estimation of burn-up of spent uranium in soil samples collected in the vicinity of Chernobyl.

Experimental

Instrumentation

A double-focusing sector-field ICP-MS instrument (ELEMENT, Finnigan MAT, Bremen, Germany) was used for isotopic ratio measurements of uranium and transuranium elements in soil samples after digestion and chemical separation. The ICP torch was shielded with a grounded platinum electrode (GuardElectrode®, Finnigan MAT), which is switched on (ground potential) or off (floating potential) electronically. A low-flow microconcentric nebulizer (MCN)

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with membrane desolvation (Aridus, CETAC Technologies Inc., Omaha, Nebraska, USA) was used for solution introduction into the ICP-SFMS instrument. Aqueous solution was introduced into the Aridus in the continuous flow mode *via* a syringe pump (Harvard Apparatus, Inc., Holliston, MA, USA) or a peristaltic pump (Perimax 12, Spetec GmbH, Erding, Germany). Further details of the ICP-SFMS instrument used can be found elsewhere.[15,16]

Standards and reagents

An isotopic standard solution of uranium (CCLU-500 laboratory standard, Nuclear Research Center, Prague, Czech Republic)[17,18] was used for optimizing isotope ratio measurements of uranium. Uranium isotope ratio values for the CCLU-500 laboratory standard ($^{234}U/^{238}U = 0.011122$, $^{235}U/^{238}U = 0.99991$, $^{236}U/^{238}U = 0.002789$) were established by calibration against the NIST-500 SRM values obtained by thermal ionization mass spectrometry (TIMS).[17] A uranium solution with a natural isotopic composition was prepared as described elsewhere.[9] All reagents were diluted with deionized Milli-Q water (18 MΩ cm,) obtained from a Millipore Milli-Q-Plus water purifier, to the necessary concentrations for determining the isotopic ratios of uranium by ICP-MS. The solutions were acidified to 1% sub-boiled HNO_3.

Samples

Soil samples were taken from the relocation zone of Belarus (4–20 km to the north and west of the Chernobyl NPP) in areas undisturbed by technogenic and anthropogenic activities since the Chernobyl accident. Soil was sampled with a coring device that was specially designed to cut 10 cm thick soil layers down to a depth of 40 cm. The collected soil samples were sieved through a 1.0 mm screen for the removal of stones and fragments of plant roots, and carefully mixed. The samples were then dried to constant weight at 105 ± 5 °C. After homogenization, 250 g of the sample were ashed at 600 ± 50 °C for 1 h. Plant roots and vegetation were incinerated separately at 550 ± 50 °C for 2 h and then the ash was mixed with the sample.

For uranium and plutonium isotopic measurements in upper soil layers, a rapid sample preparation procedure was developed. The 0.5 g soil samples (three soil samples of 0.5 g were taken from every 250 g sample) were digested in closed vessels (XP-1500) in a microwave oven (Microwave Activated Reaction System Mars 5, CEM Corporation, USA) under temperature and pressure control using a mixture consisting of 5 ml concentrated Suprapur nitric acid and 1 ml 40% HF (analytical-reagent grade purity). After digestion the sample solution was evaporated and the residue was dissolved in 4 M HNO_3. 5 ml of an $Al(NO_3)_3$ solution [1000 g $Al(NO_3)_3{\cdot}9H_2O$ and 65 ml ammonia per liter] and 0.1 ml of a 0.1 M $KMnO_4$ solution were added to 1 ml of dissolved sample. After about 5 min, actinides were extracted with 5 ml of a solution of 10% methyl trioctylammonium chloride in isobutyl methyl ketone (IBMK) as described by Küppers.[19] However, such a sample treatment may involve the introduction of a systematic error in determining the plutonium concentration *via* isotope dilution, because the plutonium oxide (PuO_2) formed at high temperatures during the accident is mostly insoluble and may not be completely dissolved, therefore, leading to disequilibrium with the yield tracer. Therefore, for isotopic measurement of Pu, the ashed samples were leached with concentrated HNO_3 and then with concentrated HCl, after the addition of known activities of isotope spikes of ^{233}U and ^{242}Pu, and the residue was treated with warm HF. After solution evaporation, the residue was boiled with concentrated $HClO_4$. After eliminating $HClO_4$ by warming, the residue was dissolved in 8 M HNO_3 and filtered.[20] The solution was then passed through an anion exchange resin, AV-17. Plutonium was eluted by NH_4I solution in HCl.

Table 1 Optimized operating conditions of ICP-SFMS attached to a low-flow microconcentric nebulizer with membrane desolvation (Aridus, CETAC Technologies Inc.)

Rf power/W	1100
Cooling gas flow rate/l min^{-1}	14
Auxiliary gas flow rate/l min^{-1}	1.4
Nebulizer gas flow rate/l min^{-1}	1.1
Sweep gas flow rate/l min^{-1}	3.6
Solution uptake rate/ml min^{-1}	0.1
Spray chamber temperature/°C	70
Membrane temperature/°C	160
Total acquisition time per replicate/min	0.6
Number of replicates	10
Number of measurements per sample	6
Mass window (%)	20
Mass resolution, $m/\Delta m$	300

Measurement procedure

Optimization of the experimental parameters of ICP-SFMS was performed with respect to the maximum ion intensity of $^{238}U^+$ and minimum rate of formation of uranium hydride by using a 1 μg l^{-1} natural uranium solution introduced by the MCN with desolvator Aridus. Optimized experimental parameters of the ICP-SFMS coupled to the Aridus system are summarized in Table 1. The measured uranium isotopic ratio in soil samples was corrected by taking into account the mass discrimination factor (assuming a linear correlation[21,22]) determined experimentally by measuring the CCLU-500 standard solution as well as UH^+/U^+ ratios and the dead time of the ICP-SFMS detector.[23] The combined uncertainty of isotopic ratio measurements was calculated according to the EURACHEM/CITAC Guide[24] taking into account standard deviations of the measured ratios, background (including instrument background and interfering hydride ions) and uncertainties of the mass discrimination factor and the dead time. Figures of merit of the ICP-SFMS coupled to an MCN with desolvator are summarized in Table 2. Application of a membrane desolvator allowed a UH^+/U^+ ratio of 3×10^{-5} to be achieved, compared to the value of 1.5×10^{-4} achieved using the same ICP-SFMS with a nebulizer but without desolvation.[25]

Table 2 Figures of merit of ICP-SFMS coupled to MCN with membrane desolvator (Aridus, CETAC Technologies Inc.)

Parameter	Value
Sensitivity	4.2×10^9 cps per ppm
Instrument background at mass 220 u	0.4 cps
Dead time	24 ns
Uranium hydride ratio formation, UH^+/U^+	3×10^{-5}
Limit of detection (3σ criterion) for $^{236}U^a$ and $^{239}Pu^b$ in aqueous solution	0.2 pg l^{-1}
Limit of detection (3σ criterion) for $^{236}U^a$ and $^{239}Pu^b$ in soil	0.04 pg g^{-1}
Abundance sensitivity (peak tail from ^{238}U at mass 237 u)	9×10^{-6}
Mass discrimination per mass unit (measured using CCLU-500)	0.04%
Precision (RSD) of $^{235}U/^{238}U$ isotope ratio measured in natural uranium	0.026%
Precision (RSD) of $^{235}U/^{238}U$ isotope ratio measured in CCLU-500	0.020%
Precision (RSD) of $^{236}U/^{238}U$ isotope ratio measured in CCLU-500	0.039%

[a] After separation of actinides from matrix. [b] After separation of plutonium from uranium.

Calculation of the portion of spent nuclear fuel and the burn-up

The portion P of nuclear fuel in the spent uranium/natural uranium mixture is determined as a ratio of total density N_i^{f} of all isotopes originating from nuclear fuel (where i refers to isotopes ^{234}U, ^{235}U, ^{236}U and ^{238}U) to the total density N_j^{n} of all isotopes originating from natural uranium (where j refers to isotopes ^{234}U, ^{235}U and ^{238}U). The equation can be simplified as follows when neglecting the less abundant ^{234}U and ^{236}U isotopes:

$$P = \frac{N_{234}^{\mathrm{f}} + N_{235}^{\mathrm{f}} + N_{236}^{\mathrm{f}} + N_{238}^{\mathrm{f}}}{N_{234}^{\mathrm{n}} + N_{235}^{\mathrm{n}} + N_{238}^{\mathrm{n}}} \cong \frac{N_{238}^{\mathrm{f}} \times (1 + I_{235/238}^{\mathrm{f}})}{N_{238}^{\mathrm{n}} \times (1 + I_{235/238}^{\mathrm{n}})} \qquad (1)$$

where $I_{235/238}^{\mathrm{n}}$ and $I_{235/238}^{\mathrm{f}}$ are ^{235}U/^{238}U isotope ratios in natural uranium and nuclear fuel, respectively.

Rigorous calculation of burn-up grade as the loss of uranium and plutonium isotopes owing to fission and radiation capture of neutrons in relation to the weight of fuel loaded into the reactor requires precise information on fuel irradiation history, such as neutron energy spectrum, neutron flux distribution and power density distribution in each fuel assembly. When using a simplified assumption that the only fissile isotope is ^{235}U, the FIFA (fissions per initial fissile atom) formula can be used[26] to calculate burn-up grade, B, as the ratio of burned ^{235}U atoms to initial ^{235}U atom number:

$$B = \frac{N_{235}^{\mathrm{f\ initial}} - N_{235}^{\mathrm{f}}}{N_{235}^{\mathrm{f\ initial}}} \qquad (2)$$

where $N_{235}^{\mathrm{f\ initial}}$ and N_{235}^{f} are the initial and final ^{235}U atom densities in nuclear fuel, respectively.

The ^{235}U/^{238}U isotope ratio measured in the soil sample

$$I_{235/238}^{\mathrm{meas}} = \frac{N_{235}^{\mathrm{n}} + N_{235}^{\mathrm{f}}}{N_{238}^{\mathrm{n}} + N_{238}^{\mathrm{f}}} \qquad (3)$$

can be expressed using eqns. (1) and (2) and neglecting the ^{238}U fission in the reactor spectrum (*i.e.*, assuming $N_{238}^{\mathrm{f\ initial}} = N_{238}^{\mathrm{f}}$) as follows:

$$I_{235/238}^{\mathrm{meas}} = \frac{I_{235/238}^{\mathrm{n}} + I_{235/238}^{\mathrm{f\ initial}} \times (1-B) \times (I_{235/238}^{\mathrm{n}} + P \times (1 + I_{235/238}^{\mathrm{n}}))}{1 + P \times (1 + I_{235/238}^{\mathrm{n}}) + I_{235/238}^{\mathrm{f\ initial}} \times (1-B)} \qquad (4)$$

where $I_{235/238}^{\mathrm{f\ initial}}$ is ^{235}U/^{238}U isotope ratio in "fresh" not irradiated nuclear fuel of the Chernobyl reactor.

The ^{236}U/^{238}U isotope ratio measured in soil sample can be expressed as follows:

$$I_{236/238}^{\mathrm{meas}} = \frac{N_{236}^{\mathrm{f}}}{N_{238}^{\mathrm{n}} + N_{238}^{\mathrm{f}}} = \mathrm{I}_{236/238}^{\mathrm{f}} \times \frac{1}{1 + \frac{1 + I_{235/238}^{\mathrm{f}}}{P \times (1 + I_{235/238}^{\mathrm{n}})}} \qquad (5)$$

With an increase in burn-up, the ^{235}U/^{238}U ratio becomes lower due to fission of ^{235}U (^{235}U+n → fission fragments) and neutron capture with γ emission (^{235}U + n → ^{236}U + γ). The latter reaction leads to the ^{236}U production causing an increase in the ^{236}U/^{238}U ratio in irradiated nuclear fuel. Therefore, eqn. (5) can be rewritten as follows:

$$I_{236/238}^{\mathrm{meas}} = \frac{\sigma_{\mathrm{n},\gamma}}{\sigma_{\mathrm{n,f}} + \sigma_{\mathrm{n},\gamma}} \times \frac{B \times I_{235/238}^{\mathrm{f\ initial}} + P \times (1 + I_{235/238}^{\mathrm{n}})}{1 + P \times (1 + I_{235/238}^{\mathrm{n}}) + I_{235/238}^{\mathrm{f\ initial}} \times (1-B)} \qquad (6)$$

where $\sigma_{\mathrm{n,f}}$ and $\sigma_{\mathrm{n},\gamma}$ are the cross sections of ^{235}U (n,f) and ^{235}U (n,γ) reactions in the neutron spectrum of the Chernobyl reactor. Rigorous calculation of cross sections requires knowledge of the neutron density and neutron energy spectrum in the reactor core. A simplified estimation of $\sigma_{\mathrm{n,f}}$ and $\sigma_{\mathrm{n},\gamma}$ can be made for the thermal neutron spectrum. Thus, in the thermal neutron spectrum, about 14 atoms of ^{236}U should be produced per 100 burned ^{235}U atoms (without taking any account of ^{236}U production from the alpha decay of ^{240}Pu and burning ^{236}U). Approximately the same value was calculated by Kirchner and Noack[4] for the Chernobyl reactor core. It should be noted that the calculation results of the isotopic composition of Chernobyl nuclear fuel obtained by Kirchner and Noack[4] do not coincide with the calculations made by Begichev *et al.*[3] nor with previous experimental measurements of ours.[25] Despite these differences they are accepted here as the first approximation for estimating the spent uranium portion and spent uranium isotopic composition in soil samples, because of the lack of reliable data on ^{236}U abundance in the literature.[3] A more accurate analysis would require direct measurement of the isotopic composition of spent uranium and this will be the subject of separate work.

By using eqns. (4) and (6), the spent uranium portion, P, and burn-up of spent uranium, B, were calculated using the iteration method from the measured uranium isotopic ratios of the soil samples (represented by a mixture of spent uranium and natural uranium). For calculation of the parameters mentioned above, a computer code was written using Borland Pascal. As initial conditions, ^{235}U/^{238}U isotope ratios of 0.00725 and 0.02067 (initial enrichment was 2% weight[3]) were accepted for natural uranium and fresh nuclear fuel, respectively.

Results and discussion

Isotopic ratios of uranium and plutonium

Table 3 shows the results of uranium isotopic measurement in soil samples collected in the vicinity of Chernobyl, after a simplified sample preparation procedure (microwave-induced digestion and actinide extraction with IBMK[19]). The data in

Table 3 Uranium and plutonium isotope ratios measured by ICP-SFMS in soil samples from the vicinity of Chernobyl in comparison to calculated data (ref. 3) and data obtained by ICP-MS (ref. 27) and RIMS (refs. 28 and 29)

Sample	^{235}U/^{238}U	^{236}U/^{238}U	^{240}Pu/^{239}Pu
I	(9.68 ± 0.09) × 10^{-3}	(1.04 ± 0.02) × 10^{-3}	0.404 ± 0.006
II	(9.46 ± 0.09) × 10^{-3}	(8.04 ± 0.15) × 10^{-4}	0.398 ± 0.005
III	(8.72 ± 0.08) × 10^{-3}	(5.58 ± 0.11) × 10^{-4}	0.400 ± 0.006
IV	(8.60 ± 0.08) × 10^{-3}	(4.08 ± 0.08) × 10^{-4}	0.378 ± 0.006
V	(8.25 ± 0.07) × 10^{-3}	(4.08 ± 0.08) × 10^{-4}	0.402 ± 0.004
VI	(8.07 ± 0.09) × 10^{-3}	(3.23 ± 0.07) × 10^{-4}	0.410 ± 0.007
VII	(8.04 ± 0.09) × 10^{-3}	(3.23 ± 0.07) × 10^{-4}	0.403 ± 0.004
VIII	(7.64 ± 0.06) × 10^{-3}	(1.10 ± 0.03) × 10^{-4}	0.369 ± 0.006
Average			0.396 ± 0.014
Calculated (ref. 3)			0.42
Measured using ICP-MS (ref. 27)			0.408
Measured using RIMS (refs. 28 and 29)			0.38

the table are sorted according to the distance of the sampling points from the Chernobyl NPP. The isotopic composition of all the investigated Chernobyl soil samples differs from those of natural uranium; *i.e.*, the ^{235}U/^{238}U and ^{236}U/^{238}U isotope ratios range from 7.64×10^{-3} to 9.68×10^{-3} and from 1.10×10^{-4} to 1.03×10^{-3}, respectively, as natural uranium is mixed with spent reactor uranium in the contaminated soil samples. After separation of uranium and plutonium, the ^{240}Pu/^{239}Pu isotope ratios were measured (see Table 3). The ^{240}Pu/^{239}Pu isotope ratios obtained for eight samples are in the relatively narrow range of 0.369 to 0.410, which corresponds to that found by Muramatsu *et al.*[27] (0.386–0.412). The average value of 0.396 ± 0.014 is lower than the ^{240}Pu/^{239}Pu isotope ratio calculated for Chernobyl reactor fuel[3] (0.42), but it is closer to the average ratios measured by ICP-MS[27] and resonance ionization mass spectrometry (RIMS)[28,29] in contaminated soil samples from the vicinity of Chernobyl (0.408 and 0.38, respectively).

Isotopic composition and burn-up grade of spent uranium

As natural uranium is mixed with spent reactor uranium in contaminated soil samples, the observed uranium isotope ratios (Table 3) are in a relatively wide range. Analyzing ^{235}U/^{238}U and ^{236}U/^{238}U isotopic ratios reveals the portion of nuclear fuel in the spent uranium/ natural uranium mixture and provides information about the burn-up grade of spent reactor uranium. Table 4 shows the results of calculating the spent reactor portion in soil samples and the ^{235}U/^{238}U and ^{236}U/^{238}U isotopic ratios in spent uranium. In general, the portion of spent uranium decreases with increasing distance from the Chernobyl NPP. Calculated ^{235}U/^{238}U isotope ratios in spent reactor uranium are slightly higher than average ratio in the reactor core ^{235}U/^{238}U ratio calculated by Begichev *et al.*[3] The calculated burn-up grade of nuclear fuel is compared with ^{240}Pu/^{239}Pu isotope ratios in Fig. 1. The data in Fig. 1 are sorted according to the burn-up grade of uranium. With increasing burn-up grade, the ^{240}Pu/^{239}Pu isotope ratio increases due to the higher production of ^{240}Pu *via* the ^{239}Pu (n,γ) ^{240}Pu reaction. These results point to a slight variation in the burn-up grade of spent reactor uranium over the contaminated zone.

Thus, the measurement of ^{235}U/^{238}U and ^{236}U/^{238}U isotope ratios provides direct information on burn-up of spent uranium, contrary to calculation methods based on isotope ratios of fission products (*e.g.*, ^{134}Cs/^{137}Cs isotope ratio). However, the knowledge of neutron spectra would be advantageous for a more accurate estimation of ^{236}U yield and the contribution of fission of ^{238}U (with fast neutrons) and also ^{239}Pu and ^{241}Pu formed from ^{238}U by the successive reactions of neutron capture and β-decay.

Table 4 Results of calculation of spent uranium portion (%) in spent/ natural uranium mixture in soil samples and ^{235}U/^{238}U and ^{236}U/^{238}U isotope ratios in spent uranium

Sample	Spent uranium portion (%)	^{235}U/^{238}U isotope ratio	^{236}U/^{238}U isotope ratio
I	77.1	0.0104	0.00134
II	62.4	0.0108	0.00129
III	42.8	0.0107	0.0013
IV	33.5	0.0113	0.00122
V	30.7	0.0105	0.00133
VI	24.5	0.0106	0.00132
VII	24.3	0.0105	0.00133
VIII	9.3	0.0115	0.00119
Average		0.0108	0.00129
Calculated (ref. 3)		0.01004	

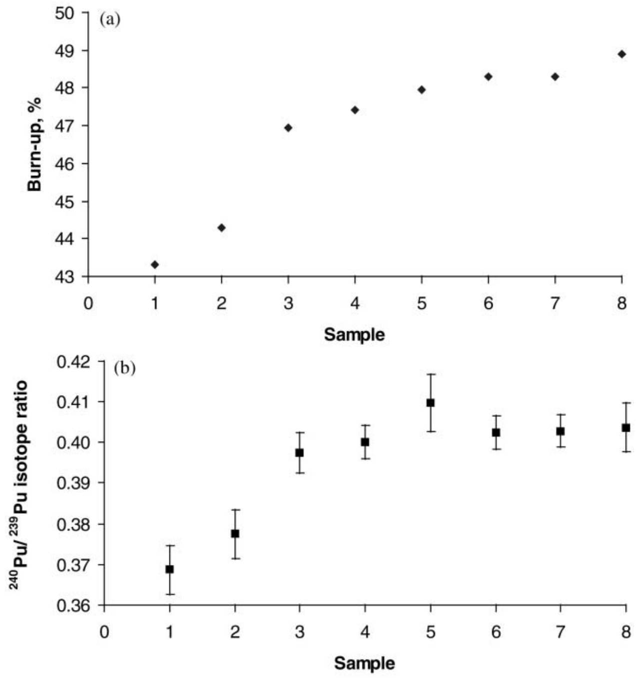

Fig. 1 Burn-up of spent uranium (a) and ^{240}Pu/^{239}Pu isotope ratios (b) measured in soil samples collected in the vicinity of Chernobyl.

Conclusions

ICP-SFMS offers high sensitivity for uranium and plutonium determination when combined with an MCN with a desolvator, Aridus, for solution introduction. The desolvation of the aerosol reduces the rate of formation of uranium hydride ions UH$^+$/U$^+$ down to 3×10^{-5}, giving a detection limit for ^{236}U and ^{239}Pu in soil after chemical separation as low as 0.04 pg g^{-1}.

Determination of ^{235}U/^{238}U and ^{236}U/^{238}U isotope ratios in soil samples allows the simultaneous calculation of the portion of spent uranium in the spent/ natural uranium mixture and burn-up of spent uranium. As expected, the ^{240}Pu/^{239}Pu isotope ratio in contaminated soil samples correlates with the burn-up of spent uranium within experimental error. A slight variation of burn-up grade of spent uranium observed by analyzing ^{235}U/^{238}U, ^{236}U/^{238}U and ^{240}Pu/^{239}Pu ratios requires closer study of burn-up grade over the contaminated zone in the future. In a future study more precise calculation of burn-up will be performed taking into account the neutron spectrum in the reactor core and the contribution of fission by other fissile nuclides.

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