MC-ICP-MS analysis of non-natural U isotope ratios using a ^{229}Th/^{232}Th external mass bias correction

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Civil and military uses of uranium lead to the possibility that U exists in the environment with non-natural isotope ratios. Therefore, characterisation of environmental U cannot rely on prior knowledge of any particular isotope ratio. For example, it is common to use the natural ^{235}U/^{238}U ratio to quantify mass bias effects in ICP-MS measurements and adjust ^{234}U/^{238}U accordingly. Such an approach cannot be applied to environmental samples whose ^{235}U/^{238}U is unknown. In this paper we demonstrate accurate and precise measurement of ^{235}U/^{238}U, ^{234}U/^{238}U and ^{236}U/^{238}U by multiple collector ICP-MS using a ^{229}Th–^{232}Th double spike to correct mass bias. This method has a major advantage over external correction of mass bias using bracketing standards because it can successfully correct small matrix-dependent changes in mass bias between standards and real environmental samples. However, care must be taken to avoid contamination of the analyte with Th from the sample which will differ in isotopic composition from the ^{229}Th–^{232}Th spike. Data are presented on non-natural U standards with a variety of ^{235}U enrichment and depletion. The efficacy of the method for environmental samples is demonstrated with two analyses of soil samples taken from a military site where depleted uranium (DU) munitions have been tested. While the presence of DU in these soils is clearly indicated, detailed source apportionment is not possible because DU appears to be highly variable in isotopic composition.

Introduction

^{235}U-enrichment to support civil and military uses of nuclear energy raises the possibility of environmental dispersal of U whose isotopic composition differs from natural abundances (*i.e.*, ^{235}U/^{238}U = 0.007 25). Both accidental and deliberate emissions from nuclear facilities could discharge enriched U into the environment. The complementary by-product of nuclear weapons production, so-called depleted uranium (DU), is widely used in applications such as armour-piercing munitions and aircraft ballast which carry an inherent risk of environmental dispersal. Accurate and precise isotopic analysis of U is critical to the evaluation of anthropogenic impact on radionuclide distribution. Moreover, the availability of sites contaminated with DU offers the possibility of studying U mobility in the environment by tracing the behaviour of isotopically anomalous U.

Until recently, the preferred method for accurate and precise isotopic analysis of U has been thermal ionization mass spectrometry (TIMS).[1] This method is used extensively in the nuclear industry for monitoring ^{235}U/^{238}U during fuel production and in geochronology for determining the U concentrations of minerals by isotope dilution mass spectrometry, typically with a ^{235}U tracer. Ionization efficiency (*i.e.*, the ratio of charged ions generated to neutral atoms evaporated) in TIMS is inversely proportional to the first ionization potential of the analyte, which for U is quite high (6.14 eV). While various schemes have been demonstrated that increase ionization efficiency by increasing the work function of the ion source filament or ionizing U as a molecular ion (*e.g.* UO$^+$), typical ionization efficiencies for U are low (*ca.* 0.01%) and show a tendency to decrease with larger sample sizes.[1]

Another disadvantage of TIMS is instrumental mass fractionation. As a sample is progressively evaporated and ionized from the mass spectrometer filament ion source, light isotopes ionize preferentially. Thus, during an analysis, which would typically consist of tens of individual ^{235}U/^{238}U determinations over an hour or so, the measured ^{235}U/^{238}U will decrease with time as the sample is consumed. The precision of the measurement is limited by this mass fractionation and its accuracy depends on the ability to correct for the effect. In many TIMS applications, mass fractionation can be corrected by internal normalization to an invariant (*i.e.* non-radiogenic) isotope ratio. For example, in Sr isotope measurements, ^{87}Sr/^{86}Sr is corrected for mass fractionation by monitoring ^{86}Sr/^{88}Sr which is assumed, by convention,[2] to be 0.1194 but can range from >0.120 to <0.118 during an analysis.

In ^{235}U/^{238}U analysis, such internal normalization is not possible because there is no invariant isotope ratio. Fortunately, TIMS mass fractionation is small (of the order of 1‰) and a first-order correction can be applied by carefully matching unknowns to U standards to ensure similar fractionation

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behaviour. A more robust approach would be to "double spike" unknowns with a mixed tracer containing non-natural U isotopes, *e.g.*, ^{233}U and ^{236}U, and normalise to the pre-determined ^{233}U/^{236}U of the double spike. However, the ^{233}U–^{236}U double spike will contain traces of natural U isotopes but in non-natural relative abundances. Successful correction for spike addition requires careful characterization of the double spike isotopic composition which would be hampered by (1) the low abundance of minor isotopes and (2) the absence of any known isotope ratio to correct instrumental mass bias effects during spike characterization. Moreover, environmental samples may contain anthropogenic ^{233}U and ^{236}U; quantification of these minor isotope abundances would require spiked (^{235}U/^{238}U) and unspiked (minor isotopes) analyses to characterise thoroughly the U isotope composition of an unknown sample.

Multiple collector inductively coupled plasma mass spectrometers (MC-ICP-MS) combine the high precision magnetic sector mass spectrometers used in TIMS with the high energy plasma ion source used extensively in quadrupole ICP-MS instruments. The potential of such instruments in U analysis has been demonstrated previously and there are some tangible advantages compared with TIMS.[3,4] A major difference between ICP-MS and TIMS is the nature of mass discrimination. The ICP interface induces a substantial mass bias (approximately 1% for U) which favours transmission of heavy isotopes into the mass spectrometer. However, this effect is relatively stable through an analysis because the sample is continually aspirated into the plasma ion source rather than progressively consumed as in TIMS. Provided the instrument is stable, the precision of an analysis is little affected by mass bias, although a large inaccuracy needs to be corrected. For natural samples, such as corals and speleothems, accurate ^{234}U/^{238}U measurements, for ^{234}U–^{230}Th geochronology, have been demonstrated by correcting mass bias using natural ^{235}U/^{238}U and an exponential mass bias correction.[5]

In addition to its temporal stability, the mass bias characteristics of the ICP interface are similar for different elements of similar mass. This has been extensively demonstrated for Pb isotope measurements using natural ^{203}Tl/^{205}Tl to effect an external mass bias correction in the absence of an invariant Pb isotope ratio.[6–8] In detail, the external correction appears to break down due to differences in the mass bias behaviour of Pb and Tl.[9] However, the Tl-doping external correction is sufficiently robust to facilitate Pb isotope measurements that are accurate to better than 0.1%. Such precision would be sufficient to detect the presence of small quantities of DU in environmental samples. Thus, we set out to test the feasibility of using MC-ICP-MS with external mass bias correction for the determination of non-natural U isotope abundances. The method developed combines the double-spike and external correction methods by doping samples with a Th tracer that is enriched in the short-lived ($t_{1/2}$ = 7340 a) ^{229}Th isotope. While the utility of this method is likely to be limited by differences in the mass bias behaviour of U and Th, its advantages are the relatively simple procedure required to characterise the ^{229}Th–^{232}Th spike and the ability to measure the minor U isotopes in a single spiked sample.

Experimental

MC-ICP-MS

The MC-ICP-MS used in this study is an upgraded Micromass (now GV Instruments) IsoProbe (Serial No. HC024) equipped with 9 Faraday collectors and an ion-counting Daly-photomultiplier detector located behind a wide-access retarding potential (WARP) filter. Seven additional ion-counting channeltron multipliers are fitted but were not used in this study. The WARP filter is an important component in the context of this study because it greatly improves the abundance sensitivity of the instrument (*i.e.*, the ability to separate a minor peak from the tail of an adjacent major peak) from >20 ppm at -1 amu for the Faraday detectors to <200 ppb at the Daly-Photomultiplier detector. The enhanced abundance sensitivity allows us accurately to measure the minor U isotopes without requiring a further correction for peak tails.

The sample introduction system is an Elemental Scientific Inc. Apex-Q device equipped with an ACM desolvating membrane. An Elemental Scientific Inc. PFA-ST nebuliser operated at a flow rate of 50 μl min^{-1} connects directly to the sample probe of a Cetac Technologies ASX-500 autosampler controlled by the instrument software. Additional operating conditions for the instrument are given in Table 1.

^{229}Th–^{232}Th double spike—spike equilibration

^{229}Th was obtained from the Oak Ridge National Laboratory and mixed with natural Th taken from a 1000 μg ml^{-1} ICP-MS calibration solution (Baker Analyzed®) to yield a mixture with ^{229}Th/^{232}Th close to unity. In the first attempt to prepare a ^{229}Th–^{232}Th spike, we mixed ^{229}Th stored in a mixture of 1.5 M HNO_3–3 M HF with natural Th stored in 1.5 M HNO_3. Repeated measurements of separate aliquots of this solution (see below for exact procedure) gave a wide range of ^{229}Th/^{232}Th greatly outside the precision of each individual measurement. Approximately 20 ml of mixed spike was then repeatedly dried down in 16 M HNO_3 and stored in 1.5 M HNO_3 for two weeks. This solution yielded much more reproducible ^{229}Th/^{232}Th measurements for separate aliquots. We conclude that mixing between ^{229}Th as fluoride and ^{232}Th as nitrate was kinetically inhibited such that successive aliquots of the mixed spike had different ^{229}Th/^{232}Th ratios. This

Table 1 Operating conditions for the MC-ICP-MS

Accelerating voltage	*ca.* 6000 V
Rf generator	Serren IPS 12000
Rf power	1350 W
Ar gas flow rates	
Cool gas	13.5 l min^{-1}
Intermediate	1 l min^{-1}
Collision gas	1.8 ml min^{-1}
Sample introduction system	Elemental Scientific Inc. Apex-Q with ACM membrane desolvation unit
Nebuliser	PFA 50 μl min^{-1}
Heating stage	100 °C
Peltier cooling stage	2 °C
Nebuliser 1 Ar	0.85 l min^{-1}
Nebuliser 2 Ar	0.05 l min^{-1}
Sweep gas	Argon
Carrier gas	Nitrogen

experience cautions against storage of Th spikes in HNO_3–HF mixtures as this may result in a failure to achieve equilibration between spike and sample. Subsequently, a new ^{229}Th–^{232}Th spike was prepared and stored in 1.5 M HNO_3. Calibration of this spike and U isotope ratio analyses using the spike were done exclusively in HNO_3 media.

^{229}Th–^{232}Th double spike—spike isotope ratio calibration

The ^{229}Th–^{232}Th double spike in 1.5 M HNO_3 containing approximately 5 ng g^{-1} of each isotope was mixed with a 50 ng g^{-1} natural U 1.5 M HNO_3 solution prepared from the certified reference material (CRM) NBL112-A (formerly NBS SRM 960) obtained from the US Department of Energy, New Brunswick Laboratory. ^{229}Th, ^{230}Th, ^{235}U and ^{238}U were measured simultaneously using Faraday detectors with ion currents in the 1–10 pA range. Ion beam intensities were corrected for zeroes measured at the peak-centred magnet setting used to collect data for the previous sample in a sequence of analyses. Zeroes were measured for 60 s in a blank solution of the same 1.5 M HNO_3 used to dilute the samples immediately prior to the analysis; throughout we refer to this procedure as the "on-peak zero". The ^{229}Th/^{232}Th of the double spike was measured and corrected for mass bias using an exponential correction factor determined from the measured ^{235}U/^{238}U (NBL112-A has a natural ^{235}U/^{238}U of 0.007 253). The mean ^{229}Th/^{232}Th of the double spike was 0.910 87 ± 0.001 16 (2 sd, $n = 17$).

U isotope measurements

The U isotopic compositions of three U solutions prepared from certified reference materials, whose isotopic compositions are known to differ, were measured. In addition, we measured four samples of unknown isotopic composition, two DU solutions and two environmental soil samples from a military site that has been used for test firing of DU munitions. It was considered important to evaluate the method on real samples as well as standard U solutions because real samples will contain ^{232}Th but no ^{229}Th. Thus, to maintain the integrity of the ^{229}Th/^{232}Th mass bias correction, it is critical that sample Th does not contaminate the U fraction separated for analysis.

U from the unknown soil sample was separated on Eichrom Industries Inc. U/TEVA.spec resin using a method similar to the first column stage of the procedure described by Yokoyama *et al.*[10] Matrix elements were eluted with 4 M HNO_3: Th was eluted with 5 M HCl and U with 0.1 M HNO_3. The second column stage was omitted here because it is largely designed to remove Zr from the Th fraction which was not of interest in this study. In order to optimise the U–Th separation, it was necessary to increase the volume of 5 M HCl eluted to prevent carry over of Th into the U fraction.

U solutions and separated samples were spiked with the ^{229}Th–^{232}Th tracer to achieve ion beams of approximately 0.2 V for ^{229}Th and ^{232}Th and 1–2 V for ^{238}U. These allow ^{229}Th/^{232}Th and ^{235}U/^{238}U to be determined using Faraday detectors in static multi-collection mode. Simultaneously, ^{234}U was measured on the Daly photomultiplier detector with a beam intensity of about 4000 cps. All ion beams were corrected for background using on-peak zeroes. The typical on-peak zero at mass 234 is <50 cps; this is high compared with TIMS instruments (<1 cps) but it is sufficiently stable to facilitate a successful subtraction. Moreover, MC-ICP-MS measurements are made with fairly intense ^{234}U/^{238}U signals over short periods and are therefore less vulnerable to background than relatively low intensity TIMS measurements with longer acquisition times would be.

^{229}Th/^{232}Th was used to assess mass bias by comparison with the value determined for the double spike and an exponential mass bias law. The resultant mass bias factor (β_{Th}) was then used to correct ^{235}U/^{238}U and ^{234}U/^{238}U. The efficiency of the Daly photomultiplier detector is about 97% of that of a Faraday and this "Daly–Faraday gain" shows some temporal drift (typically $<0.1\%$ per hour). ^{234}U/^{238}U was corrected for this bias by bracketing analyses of unknowns between measurements of the NBL112-A CRM. After mass bias correction using ^{235}U/^{238}U to give a mass bias factor (β_U), inaccuracy of ^{234}U/^{238}U for NBL112-A was attributed to the Daly–Faraday gain. The mean of the two bracketing NBL112-A standards was used to correct for this gain factor in the ^{234}U/^{238}U determined as an unknown. Such an approach has worked successfully for natural U samples.[11] Thus we are able to measure both ^{235}U/^{238}U, corrected for mass bias using the ^{229}Th–^{232}Th double spike, and ^{234}U/^{238}U corrected for mass bias with the Th spike and for Daly–Faraday gain using bracketing standards. Data for both unknowns and bracketing standards were collected as 5 blocks of 20 × 5 s cycles. Each analysis took approximately 10 min (including a delay to allow sample to aspirate from the auto-sampler tube to the plasma) and consumed about 25 ng of total U. Analyses were run automatically under computer control and typically allowed to proceed unattended.

Having measured the ^{235}U/^{238}U of each sample, a second mass spectrometric measurement was made on the same sample solution to determine ^{236}U/^{238}U. This time, ^{235}U and ^{238}U are measured on Faraday collectors and used to determine the mass bias factor. ^{236}U was measured on the Daly photomultiplier detector whose relative gain was determined by bracketing standards. For ^{236}U we used U010 as the bracketing standard because it contains measurable ^{236}U whereas NBL112-A, being natural U, is effectively free of ^{236}U. Data for both unknowns and bracketing standards were collected as 5 blocks of 20 × 5 s cycles. Backgrounds were corrected using on-peak zeroes. The ^{236}U background is *ca.* 200 cps and the high background probably reflects the fact that ^{236}U has been introduced to the instrument in other applications at levels that gave 10 mV ^{236}U signals. The measurement was identical in time and amount of analyte to the ^{234,235}U/^{238}U measurement. Measured ^{236}U/^{238}U was corrected off-line for mass bias and Daly gain using the ^{235}U/^{238}U from the earlier analysis and the mean mass bias corrected ^{236}U/^{238}U of the bracketing standards. In principle, the two analyses could be combined into a two cycle dynamic routine but that would be slightly less efficient on sample consumption due to the increased duty cycle associated with peak-switching the magnet.

Table 2 U isotope data

U solutions	N	^{235}U/^{238}U	sd (%)	^{234}U/^{238}U	sd (%)	^{236}U/^{238}U	se (%)[a]
NBL112-A							
This study	7	0.007252	0.099	0.00005283[c]	0.158	—	—
Recommended[b]	21	**0.007253**		**0.00005286**	**0.024**	—	—
CRM U010							
This study	10	0.010132	0.065	0.0000543	0.181	0.0000678[c]	0.1060
Certified		**0.010140**	**0.050**	**0.0000547**	**0.462**	**0.0000688**	**0.516**
CRM U030-A							
This study	16	0.031354	0.043	0.0002874	0.106	0.00000613	0.3122
						0.00000614	0.3127
						0.00000625	0.3664
Certified		**0.031367**	**0.027**	**0.0002866**	**0.108**	**0.0000618**	**0.417**
JM (25 ng)	9	0.003424	0.168	0.00002133	0.182	0.0001480	0.0761
JM (50 ng)	7	0.003425	0.057	0.00002131	0.123		
			se (%)		se (%)[d]		se (%)[d]
DU-1	1	0.003875	0.0489	0.00002387	0.1277	0.00010804	0.1207
	1	0.003868	0.0525	0.00002374	0.1298	0.00010823	0.1152
CHARM-3 DU[e]	1	0.001998		0.00001059		0.00000302	
Environmental samples							
Soil-A	1	0.003636	0.0213	0.00002095	0.0794	0.00001227	0.1438
Soil-B	1	0.002903	0.0117	0.00001452	0.0520	0.00001252	0.1073

[a] Errors expressed as percentages are standard errors (se) for individual analyses and standard deviations (sd) for replicate analyses and certified values. [b] Ref. 13. [c] NBL112-A ^{234}U/^{238}U and U010, ^{236}U/^{238}U corrected for Daly–Faraday gain using separate aliquots of NBL112-A and U010, respectively, as bracketing standards. [d] Errors on bracketing standards used to correct Daly–Faraday gain are propagated to final ^{234}U/^{238}U and ^{236}U/^{238}U ratios. [e] Calculated from specific activities, ref. 12.

Results

U isotope standard results

The efficacy of the ^{229}Th/^{232}Th double spike method was demonstrated using NBL112-A analysed as an unknown and two additional reference materials supplied by the New Brunswick Laboratory. U010 is nominally 1% ^{235}U and U030-A nominally 3% ^{235}U and their ^{234}U/^{238}U ratios differ by about a factor of 5. Our mean results for these reference materials (Table 2, Fig. 1) are accurate within the precision of the certified values for both ^{235}U/^{238}U and ^{234}U/^{238}U. A single ^{236}U/^{238}U determination of U010 bracketed by a second (unspiked) U010 solution is slightly beyond the combined errors of the analysis (standard error) and the certified value (standard deviation) but three ^{236}U/^{238}U analyses of U030-A are all accurate. ^{236}U/^{238}U analyses of NBL112-A were highly variable but always $<7 \times 10^{-7}$. Since NBL112-A should be ^{236}U-free, we consider that the signal measured at mass 236 (corrected for on-peak zero) is likely to result from ^{235}UH^{+}. This represents <0.02% of the ^{235}U peak and is trivial compared with the precision attainable on ^{236}U/^{238}U.

In addition, we analysed the isotopic composition of two internally-prepared DU solutions. JM was prepared by diluting a Johnson-Matthey Specpure$^{®\ 1000\ \mu g\ ml^{-1}}$ ICP-MS calibration solution to give approximately 50 ng g^{-1}. In common with other commercially available concentration calibration solutions, JM is prepared from DU. DU-1 was prepared from a sample of DU supplied by British Nuclear Fuels plc (BNFL) in 1993, originally for use as a reducing agent in the preparation of samples for H isotope analysis. The DU was dissolved in 16 M HNO_3 and diluted to give approximately 100 ng g^{-1} in 1.5 M HNO_3.

Although these solutions are not certified for isotopic composition, the analysis of pure DU is informative because the relatively low levels of ^{234}U and ^{235}U will limit analytical performance. Thus, our results for these solutions (Table 2) are likely to illustrate the minimum analytical performance that would be expected for environmental samples. The low ^{235}U/^{238}U of DU makes it critical to ensure that the ^{235}U ion beam is sufficiently intense to be measured reliably on a Faraday detector. For natural U, a 50 ng g^{-1} U solution yields approximately 1 V ^{238}U and 7 mV ^{235}U. For a 50 ng g^{-1} solution of DU-1, the ^{235}U intensity is reduced to *ca.* 3 mV and the low intensity results in relatively poor analytical precision (Table 2). Increasing the DU-1 concentration to 100 ng g^{-1} gives 7 mV ^{235}U and substantially improved external precision (Table 2).

The DU measurements are of interest because they display higher ^{234,235,236}U/^{238}U than a previously published analysis of a CHARM-3 DU penetrator.[12] It appears that DU supplied by BNFL in 1993 and used in the Johnson Matthey standard is less depleted than that used in the DU weapon which was supplied to BAe Systems (Royal Ordnance) by Starmet Corporation of the USA.[12] Moreover, the three DU samples and natural U are co-linear in a plot of ^{234}U/^{238}U *versus* ^{235}U/^{238}U (Fig. 2) suggesting that the U-enrichment process concentrates both ^{235}U and ^{234}U in constant proportion. The DU-1 and JM samples are also enriched in ^{236}U compared to the CHARM-3 weapon. However, a similar correlation is not found in

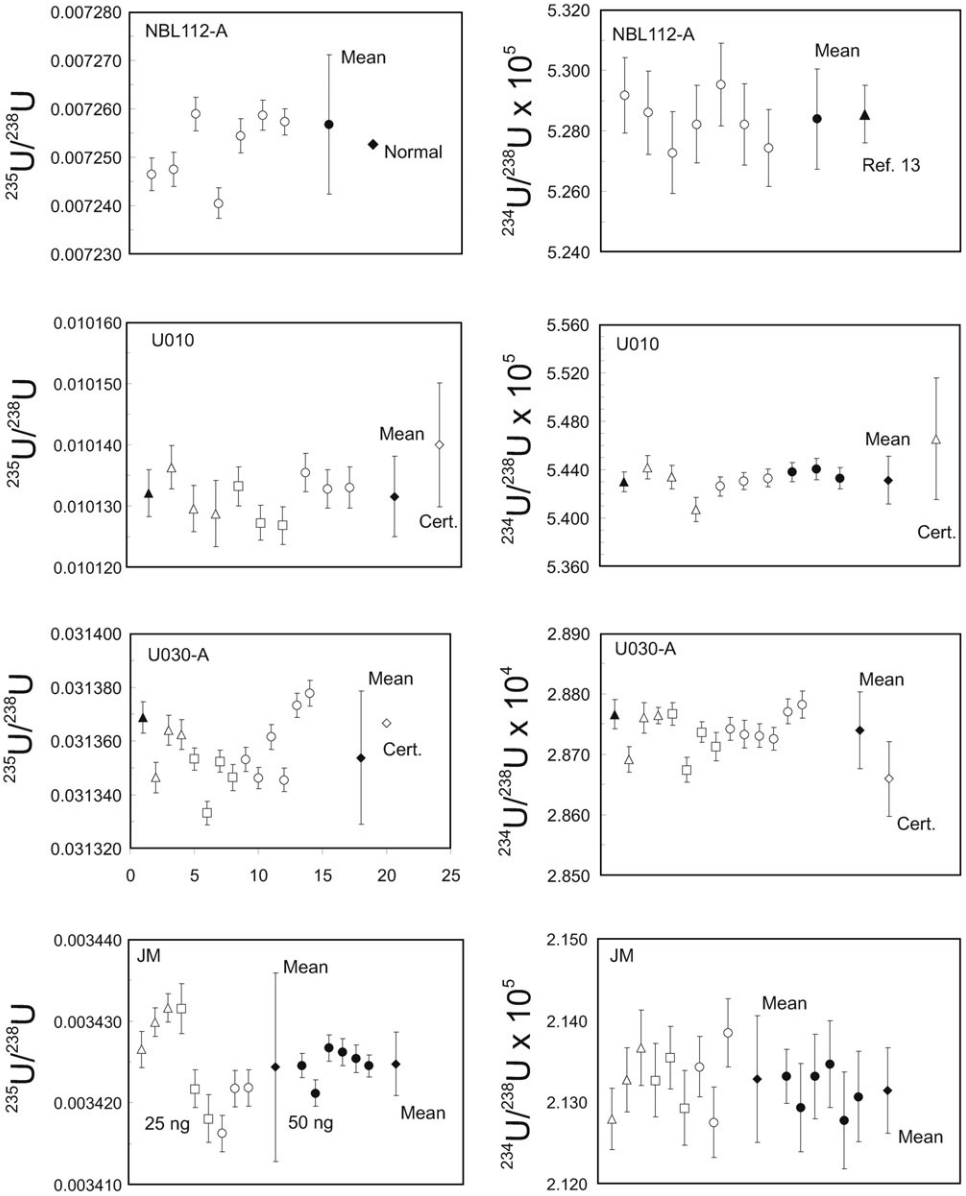

Fig. 1 $^{235}U/^{238}U$ and $^{234}U/^{238}U$ for the NBL112-A (analysed as unknown), U010 and U030-A Certified Reference Materials and the JM internal laboratory reference solution. Symbols represent separate analytical sessions. Error bars (omitted when smaller than symbol) are 2 standard errors for individual analyses and 2 standard deviations for the mean and certified or recommended (cert.) value.

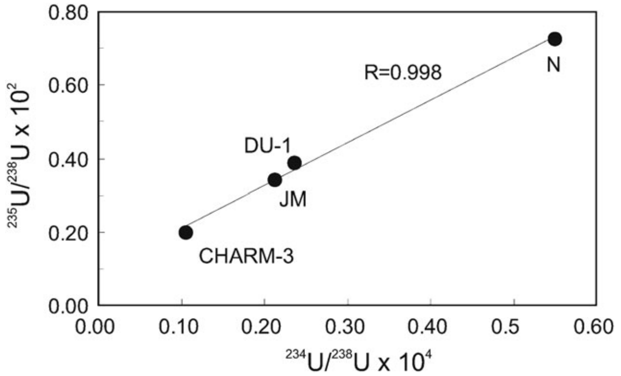

Fig. 2 U isotope composition of natural U (N) and (in order of decreasing $^{235}U/^{238}U$) DU-1 and JM laboratory reference solutions and a CHARM-3 penetrator.[12]

$^{236}U/^{238}U$ *versus* $^{235}U/^{238}U$, presumably reflecting different starting $^{236}U/^{238}U$ compositions which depend on the proportion of (^{236}U-enriched) reprocessed nuclear fuel in the different DU sources.

Soil sample results

The ability of our sample preparation method to eliminate sample Th from the U fraction can be assessed by comparison of the exponential correction factor of the sample (β_{Th}) with those of the bracketing standards (β_U). Breakthrough of sample ^{232}Th into the U fraction would manifest itself as an apparent change in mass bias between the sample and the bracketing standards because β_{Th} is calculated from the measured $^{229}Th/^{232}Th$ which would be reduced by incorporation of natural ^{232}Th. Minor changes in mass bias are to be expected because this is a matrix-dependent phenomenon and the sample matrix, even after U pre-concentration, is likely to differ from the pure U of the bracketing NBL112-A standard. Indeed, it is these small changes in mass bias that preclude the accurate correction of mass bias effects using standard–sample bracketing and call for the double-spike approach. Because the sample Th has a ^{229}Th/^{232}Th ratio approaching zero, any breakthrough of sample Th will result in a major change in ^{229}Th/^{232}Th which will yield a β_{Th} factor that is demonstrably erroneous. For example, the analyses reported here had $\beta_{\mathrm{Th}} = -1.72$ and -1.76, compared with average $\beta_{\mathrm{U}} = -1.82$ and -1.77 for the bracketing standards. A previous measurement on a similar sample before the U–Th separation had been optimised gave $\beta_{\mathrm{Th}} = -13.7$ compared with $\beta_{\mathrm{U}} = -1.81$ and -1.80 for the standards, clearly indicating the presence of sample ^{232}Th.

The analysed soil samples (Table 2) are demonstrably contaminated with DU, having lower ^{235}U/^{238}U and ^{234}U/^{238}U than natural and higher ^{236}U/^{238}U. Interestingly, the soil samples fall between the DU sample prepared for this study (DU1) and the CHARM-3 weapon.[12] Thus, it is not possible to offer more detailed source apportionment between natural and anthropogenic sources of U without further information on the variation in isotopic composition of the DU dispersed into the soil at this site.

Conclusions

Accurate and precise analysis of U isotope ratios in samples with non-natural U isotopic composition by MC-ICP-MS has been demonstrated. The method uses a ^{229}Th–^{232}Th double spike to correct mass bias, allowing simultaneous determination of ^{235}U/^{238}U and ^{234}U/^{238}U. ^{236}U/^{238}U can be determined in a second measurement using the measured ^{235}U/^{238}U to correct mass bias. Careful sample preparation is required to prevent breakthrough of sample ^{232}Th into the separated U fraction, which would compromise the mass bias correction by altering ^{229}Th/^{232}Th. The method provides a reliable analytical protocol for the monitoring of anthropogenic U inputs to the natural environment. Sample size requirements are small (*ca.* 50 ng total U), which allows the detection of anthropogenic U at levels far below those considered to be of radiological concern. Such measurements will allow assessment of the environmental dispersal of anthropogenic U. Moreover, the sensitivity of the technique provides the opportunity to detect ultra-trace amounts of depleted uranium and thereby study the detailed behaviour and associations of U at contaminated sites.

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