Journal of Environmental Radioactivity 102 (2011) 331–335

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

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

Determination of U, Pu and Am isotopes in Irish Sea sediment by a combination of AMS and radiometric methods

M. Srncik [a,b,*], E. Hrnecek [b], P. Steier [c], G. Wallner [a]

[a] Department of Inorganic Chemistry, University of Vienna, Währinger Straße 42, A-1090 Vienna, Austria
[b] European Commission, Joint Research Centre, Institute for Transuranium Elements, P.O. Box 2340, 76125 Karlsruhe, Germany
[c] VERA Laboratory, Faculty of Physics – Isotope Research, University of Vienna, Währinger Straße 17, A-1090 Vienna, Austria

ARTICLE INFO

Article history:
Received 5 November 2010
Received in revised form
13 January 2011
Accepted 21 January 2011
Available online 12 February 2011

Keywords:
Sellafield
Sediment samples
$^{236}U/^{238}U$
$^{240}Pu/^{239}Pu$
$^{242}Pu/^{239}Pu$

ABSTRACT

Samples from a marine sediment core from the Irish Sea (54.416 N, 3.563 W) were analyzed for the isotopic composition of uranium, plutonium and americium by a combination of radiometric methods and AMS. The radiochemical procedure consisted of a Pu separation step by anion exchange, subsequent U separation by extraction chromatography using UTEVA® and finally Am separation with TRU® Resin.

Additionally to radiometric determination of these isotopes by alpha spectrometry, the separated samples were also used for the determination of $^{236}U/^{238}U$ and plutonium isotope ratios by Accelerator Mass Spectrometry (AMS) at the VERA facility.

1. Introduction

Anthropogenic uranium and transuranium elements like plutonium and americium can be found in the nature because of nuclear weapons testing (e.g. Mururoa, Semipalatinsk), nuclear reactor accidents (Chernobyl), discharges of radioactive waste (Sellafield, La Hague, Mayak) and accidents with nuclear devices, e.g. Palomares and Thule (Salbu, 2001). This work will emphasize on sediment samples near the Sellafield reprocessing facility (formerly Windscale) which is located in Cumbria (UK) along the Irish Sea. In the 1950s this facility was used for the production of weapons-grade Pu. Since the 1960s U and Pu are reprocessed from spent fuel in Sellafield (Ketterer and Szechenyi, 2008). Over the past 40 years radionuclides have been discharged from the nuclear fuel reprocessing plant, under the authorization of the government, into the sea and atmosphere (Hu et al., 2010). The magnitude and radionuclide composition of the discharged materials have varied in time depending on factors like the type of the fuel, its burn-up, the nature and duration of storage prior to reprocessing and method of reprocessing (Guéguéniat et al., 1996; Cook et al., 1997).

The discharge history of Pu and Am is well documented by Gray et al. (1995) and shows that an estimated total activity of 609 TBq ^{239}Pu, 22 PBq ^{241}Pu and 542 TBq ^{241}Am were discharged to the Irish Sea during the period 1952–1992. The peak discharge for these transuranium elements occurred in the mid-to-late 1970s. Afterwards the amount of discharged radionuclides decreased 2 to 3 orders of magnitude due to changes mentioned above. At the beginning, the Sellafield operation was focused on the UK weapons program which was associated to low burn-up and therefore a low $^{240}Pu/^{239}Pu$ isotopic ratio. After the late 1960s the major effluent composition was produced from power reactor fuels which had a higher burn-up. According to Kershaw et al. (1995) the isotopic ratio of $^{240}Pu/^{239}Pu$ varied from around 0.05 in the 1960s to 0.25 in 1990s which indicates this effect.

^{236}U is produced in nuclear reactors mainly by thermal neutron capture on ^{235}U. A contribution from alpha-decay of ^{240}Pu is negligible because of its long half-life of 23 Ma and low concentrations in Irish Sea water and sediment (Lee et al., 2008). Due to the reprocessing of spent Magnox fuel rods the BNFL site started at the end of 1950s to discharge tonnes of uranium in an unknown chemical and physical form into the NE Irish Sea. Hamilton and Stevens (1985) have found a ^{236}U activity of 10^{-6} Bq/g (10^9 atoms/g) in sediment samples from the Esk estuary. In 2001, it could be specified by Hamilton (2001) that since 1969 BNFL has discharged about 80 t of reprocessed depleted uranium, coming

---

* Corresponding author. Department of Inorganic Chemistry, University of Vienna, Währinger Straße 42, A-1090 Vienna, Austria. Tel.: +43 1 4277 52624; fax: +43 1 4277 52620.
E-mail address: michaela.srncik@univie.ac.at (M. Srncik).

0265-931X/$ – see front matter © 2011 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jenvrad.2011.01.004

from reprocessing waste in uranium cooling ponds and other sources (e.g. Albright & Wilson plant and the BNFL uranium fabrication plant at Springfield). No enhanced uranium concentration in sediments of the NE Irish Sea and estuaries were found; the values are rather typical for sands and silts. Marsden et al. (2001) analyzed a sediment core from the Esk estuary, West Cumbria (UK), located about 5 km south of the marine effluent discharge from BNFL Sellafield for its $^{236}U/^{238}U$ isotopic ratio via Accelerator Mass Spectrometry (AMS). The obtained values were in the range of 10^{-5} to 10^{-6}, highly above the estimated natural background of 10^{-14} (Zhao et al., 1997).

This work will focus on the determination of ^{236}U, plutonium and americium isotopes in Irish Sea sediment samples. A sediment core of 48 cm length was collected in 1993 from the Irish Sea at 54.416 latitude and −3.563 longitude during a cruise of the research vessel Gauss, Federal Maritime and Hydrographic Agency, Germany, and was divided in 2 cm layers. Samples from the three surface layers down to a depth of 6 cm were taken for analysis and additionally several samples with elevated ^{241}Am concentrations between 10 cm and 40 cm depth as well as one sample from the 46–48 cm layer (Hrnecek et al., 2002). These samples were chosen for the AMS measurement. Furthermore the procedure was validated by using an IAEA standard (IAEA-368, Pacific Ocean Sediment); the respective U, Pu and Am fractions were measured via alpha spectrometry.

2. Materials and methods

2.1. Experimental

All reagents were prepared using deionized Milli-Q (18 MΩ cm) water (Millipore, USA). 65% Suprapure HNO_3 was purified by subboiling distillation. To prevent cross-contamination (with natural U) during the chemical treatment, all laboratory equipment was cleaned with 10% HNO_3 at least for one day and washed three times with Milli-Q water, subsequently dried in a laminar flow bench and stored in clean zip bags. 30% HCl, 30% H_2O_2, 40% HF and 25% NH_3 were of Suprapure grade.

The samples (1 g) were ashed in an electric muffle furnace at 420 °C for 24 h. The weighed ashes were dissolved in 20 ml 8 M HNO_3. Several ash samples were prepared directly for the AMS measurement. The chemical separation of the respective nuclides followed the procedure given by Eichrom Technology Inc. (2005) for U, and for Pu and Am by Hrnecek and Feichtinger (2005) and Moreno et al. (1997). The sample treatment was modified to our needs and is described briefly. The chemical procedure consisted of a Pu separation step from U and Am by anion exchange (Dowex 1 × 8, 100–200 mesh, Dow Chemical Co.) in 8 M HNO_3 after oxidation state adjustment to Pu(IV). The U was separated from Am by using UTEVA® Resin (Eichrom Technologies, LLC) and Am was purified via extraction chromatography using TRU® Resin (Eichrom Technologies, LLC). The U chloride and Pu nitrate separated from the sediment samples were measured in the oxide form (details see Section 2.3) by AMS to get information about the isotope ratios $^{236}U/^{238}U$ and $^{240}Pu/^{239}Pu$, respectively. Furthermore the $^{241}Am/^{243}Am$ ratio was measured in four samples for the comparison between alpha spectrometry and AMS. The yield-monitor-free Pu fractions were controlled for possible ^{242}Pu interference in the sediment samples.

Selected ash samples given in Table 1 and one reference sample (IAEA-368, Pacific Ocean Sediment) were measured after addition of tracers ^{243}Am (SRM 4332D), ^{242}Pu (SRM 4334H) and ^{232}U (SRM 4324B), respectively, in order to check the validity of our results by alpha spectrometry. The radiochemical procedure was the same as described before but for the determination of ^{238}U, ^{238}Pu, $^{239(40)}Pu$ and ^{241}Am by alpha spectrometry thin sources were prepared by microprecipitation with NdF_3 (Hindman, 1983).

Table 1
Comparison of $^{238}Pu/^{239(40)}Pu$ results. The measurement uncertainties are given in ±1σ.

Sample no.	Perna et al. (2005)	This work
	$^{238}Pu/^{239(40)}Pu$	$^{238}Pu/^{239(40)}Pu$
St60, 0–2 cm	0.246 ± 0.011	0.248 ± 0.006
St60, 10–12 cm	0.241 ± 0.009	0.231 ± 0.007
St60, 22–24 cm	0.215 ± 0.009	0.216 ± 0.007
St60, 46–48 cm	0.187 ± 0.005	0.192 ± 0.006

2.2. Alpha spectrometry

Alpha measurements were performed with an OCTÊTETM PC (EG&G Ortec) with ULTRA ion implanted silicon detectors with an active area of 450 mm^2. For efficiency calibrations an ^{241}Am source (GT223, AEA Technology) was used. Measurements were evaluated using MAESTRO for Windows software (EG&G Ortec).

2.3. Accelerator mass spectrometry

After alpha spectrometry, the filters carrying the respective radionuclides were reprocessed for the AMS measurements according to Srncik et al. (2008) with slight modifications. The filters were dissolved in HNO_3/H_2O_2 and evaporated for three times with the same reagents as before. The residue was fumed with conc. HCl (3×), taken up in 2 ml conc. HCl and transferred to a 50 ml vial. The beaker was washed with 7 ml deionized water and this solution was also added to the vial. Finally 1 ml Fe solution containing 1 mg Fe/ml was added. By adding 25% NH_3 the actinides were co-precipitated with $Fe(OH)_3$ at pH 9. As the Pu fractions separated directly for the AMS measurement were in the nitrate form, the samples were dissolved in conc. HCl and fumed three times with the same reagent. The U and Am fraction were already in the chloride form; these were dissolved in 2 ml conc. HCl and then the procedure for U, Pu and Am was as described before. After centrifuging, the $Fe(OH)_3$ precipitate was dried at 100 °C for 2 h and then combusted at 800 °C in small Quartz tubes. The resulting oxide samples were pressed into aluminum sample holders suitable for the ion source of VERA. The VERA setup as used for actinide measurements is described in Vockenhuber et al. (2003) and Steier et al. (2010). Similar to most AMS facilities, it is based on sputtering of negative sample ions UO^- and PuO^-, a first mass separation, and acceleration to 3 MV. Gas stripping to U^{5+} and Pu^{5+} is used to break up molecular isobars (mainly $^{235}UOH^-$ for ^{236}U and $^{238}UOH^-$ for ^{239}Pu), and after a second acceleration to 18 MeV and another mass analysis the ion species of interest is identified in a Time-of-Flight (TOF) and energy detector. By these means, AMS achieves the best abundance sensitivity and the lowest detection limits for long-lived actinides (Fifield, 2008). VERA is especially optimized for this application. The ionization yield in the ion source (ca. 1%), the stripping yield to the 5+ charge state (5%) and the TOF detector efficiency (ca. 20%) combine to a total detection efficiency of 10^{-4}, if only one isotope is measured. The measurement of several isotopes on one sample requires switching, which reduces the efficiency on each single species. Due to variations in the total efficiency, only isotopic ratios can be measured precisely. For uranium, $^{236}U/^{238}U$ is used. The UO^- current is a rough measure for the uranium concentration in the sputter sample. Under typical ion source conditions, 1 μg of U per mg Fe results typically in 20 pA of ion current, but this number may vary by a factor of three between samples. The samples last for several hours under these conditions.

The U^{5+} beam current is measured with a Faraday cup, whereas $^{236}U^{5+}$ is counted in the TOF detector. To correct the detector efficiency, the isotopic ratios are calibrated using our in-house standard “Vienna-KkU” (Steier et al., 2008). No such calibration is necessary at the present level of precision, where all isotopes are measured in the TOF detector. Whereas typical environmental samples yield count rates of a few counts per second or less, the concentration of radionuclides in the Irish Sea sediment samples is much higher. Therefore, we reduced the sputter rate in the ion source strongly to maintain acceptable (a few kHz) detector count rates. Only a small part of the samples was consumed under such conditions. The measurement precision was not limited by counting statistics, but by variations of the total efficiency, which is determined from the reproducibility of repeated measurements on the same sample, and by the variation of the ratios measured on different samples of the standard material Vienna-KkU.

3. Results and discussion

For the IAEA-368 sample the obtained values are (8.5 ± 0.5) Bq/kg ^{238}Pu and (33.4 ± 1.1) Bq/kg $^{239(40)}$Pu. These values are in good agreement with the recommended value of (7.6–8.9) Bq/kg and (29.0–34.0) Bq/kg for ^{238}Pu and $^{239(40)}$Pu, respectively. The activity of ^{241}Am is (1.4 ± 0.1) Bq/kg which is also within the confidence interval of (1.2–1.5) Bq/kg. The obtained values for ^{234}U and ^{238}U are (34 ± 1) Bq/kg and (30 ± 1) Bq/kg, respectively. These are in agreement with the reference values of (21.5–44.8) Bq/kg ^{234}U and (25.0–33.0) Bq/kg ^{238}U. The excellent agreement with this standard indicates the accuracy of our procedure.

The obtained ^{238}Pu/$^{239(40)}$Pu activity ratios from the four selected samples for the alpha spectrometry measurement were compared with a former work published by Perna et al. (2005) (Table 1); ^{238}Pu was decay corrected to 1st December 1993. The samples are in good agreement within the 1σ uncertainty.

The depth profile of the ^{240}Pu/^{239}Pu isotopic ratios for the samples selected for the AMS measurement along the core is shown in Fig. 1. The obtained values for the Pu isotopic ratio are in the range of 0.20–0.33 which indicate a high burn-up of ^{239}Pu. Due to these results the core depth is too short to show the influence of reprocessing of weapons-grade Pu (low burn-up) from the early 1960s. A contribution from global fallout can be excluded due to the measured ^{238}Pu/$^{239(40)}$Pu activity ratios (Table 1). The value for this origin is 0.024 ± 0.004 (Hardy et al., 1973) and the obtained activity ratios are 0.246 at the surface and 0.187 in the deepest investigated layer which is in agreement with the data from Aston et al. (1985) for Irish Sea intertidal sediments.

Table 2
Results of the ^{242}Pu activity [mBq/g] and the isotopic ratio ^{242}Pu/^{239}Pu in sediment samples without tracer. The measurement uncertainties are given in ±1σ.

Sample no.	^{242}Pu [mBq/g]	^{242}Pu/^{239}Pu
St60, 0–2 cm	0.58 ± 0.05	0.0047 ± 0.0003
St60, 10–12 cm	1.05 ± 0.12	0.0064 ± 0.0007
St60, 22–24 cm	2.41 ± 0.13	0.0055 ± 0.0002
St60, 46–48 cm	1.38 ± 0.18	0.0054 ± 0.0007

A correlation between the radionuclide concentration in the sediment layer and the corresponding quantity of the discharged radionuclide per year is generally difficult to assess as cores are often disturbed by redistribution of the adsorbed actinides, e.g. by tidal currents (Kershaw et al., 1990). Also bioturbative mixing of the sediment after deposition complicates the interpretation of, e.g. the Pu transport time or the sedimentation rate (Aston et al., 1985). In the Cumbrian near-shore waters of the Irish Sea Pu is usually bound to suspended particulate material and mixed by tidal activity. Pu remains available for re-suspension in this area for more than six years following discharge (McKay and Walker, 1990). In general one can expect an “average integral ratio” on the discharges in the core.

In the four sediment layers from Table 1 also the activity of ^{242}Pu was determined by investigating analogue samples without adding a tracer (St60: 0–2 cm, 10–12 cm, 22–24 cm, 46–48 cm). By using the activity of the tracer which was determined by alpha spectrometry and the obtained isotopic ratios ^{242}Pu/^{239}Pu by AMS from the samples (with and without tracer), the ^{242}Pu activity for the yield-monitor-free samples could be calculated. In Table 2 the obtained activities of ^{242}Pu and the ^{242}Pu/^{239}Pu isotopic ratios are listed. The typical ^{242}Pu/^{239}Pu isotopic composition for Magnox fuel with 5–6 GWd/tU is 0.0159 (Bairiot, 2008). Our results are 60–70% lower which indicates a shorter irradiation time.

^{238}U is relatively soluble and behaves conservatively in seawater. Additional sources of U in this region can come from discharges from the Sellafield plant and a phosphate plant (Marchon) at Whitehaven (Kershaw et al., 1990). Depleted uranium from BNFL was found in sediment samples from the Esk by Hamilton and Stevens (1985). The extent of the depletion depends on the burn-up of the fuel and is assumed to be around 2000 days. According to Hamilton and Stevens (1985) the Magnox fuel will contain 580 ppm ^{236}U/gU which gives an isotopic ratio ^{236}U/^{238}U of 5.84×10^{-4}. In Table 3 the isotopic ratios of ^{236}U/^{238}U along the core are shown. The values are

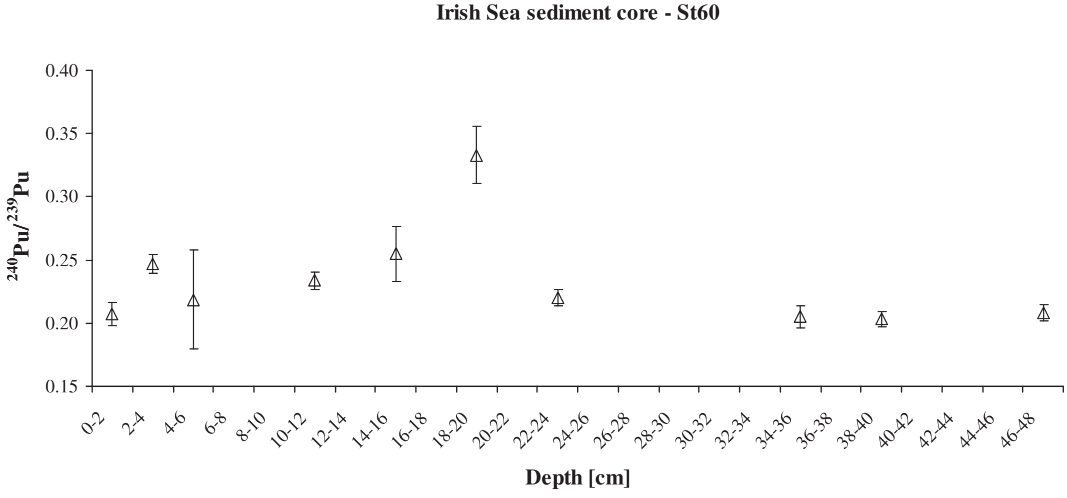

Fig. 1. Vertical distribution of ^{240}Pu/^{239}Pu (Δ). The measurement uncertainties are given in ±1σ.

Table 3
^{236}U/^{238}U results of sediment sample from the Irish Sea. The measurement uncertainties are given in ±1σ.

Sample no.	^{236}U/^{238}U
St60, 0–2 cm	$(2.41 \pm 0.20) \times 10^{-5}$
St60, 2–4 cm	$(1.53 \pm 0.11) \times 10^{-5}$
St60, 4–6 cm	$(1.35 \pm 0.11) \times 10^{-5}$
St60, 10–12 cm	$(2.25 \pm 0.23) \times 10^{-5}$
St60, 14–16 cm	$(4.15 \pm 0.35) \times 10^{-5}$
St60, 18–20 cm	$(4.36 \pm 0.26) \times 10^{-5}$
St60, 22–24 cm	$(3.82 \pm 0.23) \times 10^{-5}$
St60, 34–36 cm	$(3.29 \pm 0.40) \times 10^{-5}$
St60, 38–40 cm	$(3.45 \pm 0.37) \times 10^{-5}$
St60, 46–48 cm	$(2.96 \pm 0.41) \times 10^{-5}$

in the range from $(1.35–4.36) \times 10^{-5}$ and they are clearly above the natural background. An increase of the ratio till 20 cm depth can be noticed which reflects an increase of ^{236}U. The values are similar to the sediment samples from Esk estuary (Marsden et al., 2001). By comparing the distribution of the ^{236}U/^{238}U isotopic ratios it can be noticed that both values show a similar increase down to about 18 cm depth. Afterwards the values decrease rapidly in the Esk estuary samples to a ratio of 9.2×10^{-6}; here the ratio decreases slightly to about 3×10^{-5}. A possible reason for the difference could be a different sediment composition since the samples are not collected from the same location. According to Kershaw and Young (1988) the additional anthropogenic contribution of ^{238}U from Sellafield and Marchon is 8–15%. Assuming that the isotopic ratio of the Magnox fuel is in the range of 10^{-4} (Hamilton and Stevens, 1985), we see a contribution of about 10% along the core.

The four Irish Sea samples prepared for the alpha spectrometry measurement were spiked with ^{243}Am and therefore a first comparison with the AMS measurements (Table 4) was possible. The obtained activity ratios of ^{241}Am/^{243}Am by alpha spectrometry were transferred to the isotopic ratio. The Am samples were combusted in the muffle furnace at 800 °C in small Quartz tubes (height: 2 cm, ø: 1 cm) instead of porcelain crucibles (height: 4 cm, ø top: 3 cm, ø bottom: 2 cm). With the first samples it was noticed that after the combustion the iron oxide was found at the opening and the Quartz lid. As it was impossible to remove the samples from there, the tubes and the lids were washed with hot conc. HCl, the solutions were collected and the iron hydroxide precipitation was repeated. The affected samples were St60 (0–2 cm) and St60 (10–12 cm). In Table 4 the Am results from alpha spectrometry and AMS are presented and they are consistent within 1σ, except the St60 (0–2 cm) sample which is consistent within 2σ. The mean ratio of AMS/Alpha was 1.105 ± 0.014. As ^{241}Am has a relatively short half-life alpha spectrometry is adequate and compared to mass spectrometry techniques the most sensitive method (Vajda and Kim, 2010). We have shown that AMS is in principle applicable for the determination of Am radionuclides but at the moment due to the high cost of an AMS facility not suitable for routine measurements.

Table 4
Comparison of the isotopic ratio ^{241}Am/^{243}Am by alpha spectrometry and AMS. The measurement uncertainties are given in ±1σ.

Sample no.	Alpha spectrometry	AMS
	^{241}Am/^{243}Am	^{241}Am/^{243}Am
St60, 0–2 cm	0.080 ± 0.002	0.110 ± 0.017
St60, 10–12 cm	0.087 ± 0.002	0.078 ± 0.012
St60, 22–24 cm	0.237 ± 0.006	0.249 ± 0.037
St60, 46–48 cm	0.101 ± 0.003	0.111 ± 0.017

4. Conclusion

This work was focused on the determination of ^{236}U/^{238}U and ^{240}Pu/^{239}Pu in layers of an Irish Sea sediment core. The ^{236}U/^{238}U isotopic ratios measured by AMS are far above the natural background, showing the applicability of this ratio to trace sources of irradiated U in the environment. The average ^{240}Pu/^{239}Pu isotopic ratio is 0.23 ± 0.04 which indicates a high burn-up of ^{239}Pu and therefore is not weapons-grade Pu. Due to this result the samples show a time scale from the late 1960s till 1990s. The respective U and Pu isotopic ratios show a maximum at the same layer. Additionally, for the first time the isotopic ratio ^{241}Am/^{243}Am was measured via AMS and compared with alpha spectrometry measurements. The agreement between the two methods was satisfying.

Acknowledgment

The work presented in this paper was partially performed in the frame of a European Commission-Joint Research Centre "In-Service Training" scheme at the JRC-ITU. This financial support is acknowledged. The authors want to thank Dr. H. Nies, IAEA-MEL (present affiliation), and the Federal Maritime and Hydrographic Agency, Hamburg (Germany), for the courtesy of providing the Irish Sea sediment samples and collaboration with the JRC-ITU. Financial support by the FWF Austrian Science Fund (project number P21403-N19) is gratefully acknowledged.

References

Aston, S.R., Assinder, D.J., Kelly, M., 1985. Plutonium in intertidal coastal and estuarine sediments in the Northern Irish Sea. Estuarine, Coastal Shelf Sci. 20, 761–771.
Bairiot, H., 2008. Plutonium management options: liability or resource. Nucl. Eng. Technol. 40, 9–20.
Cook, G.T., MacKenzie, A.B., McDonald, P., Jones, S.R., 1997. Remobilization of Sellafield-derived radionuclides and transport from the north-east Irish Sea. J. Environ. Radioact 35, 227–241.
Eichrom Technologies, Inc., 10th of February 2005. Uranium in Soil (2 grams samples). Analytical Procedures, ACS07, Rev. 1.5.
Fifield, L.K., 2008. Accelerator mass spectrometry of the actinides. Quat. Geochronol. 3, 276–290.
Gray, J., Jones, S.R., Smith, A.D., 1995. Discharges to the environment from the Sellafield site 1951–1992. J. Radiol. Prot. 15, 99–131.
Guéguéniat, P., Germain, P., Métivier, H., 1996. Radioactivity in the Irish Sea: past practices, present status and future perspectives. In: Radionuclides in the Oceans – Inputs and Inventories. Les Editions de Physique, pp. 155–175.
Hamilton, E.I., 2001. Depleted uranium (DU): a holistic consideration of DU and related matters. Sci. Total Environ. 281, 5–21.
Hamilton, E.I., Stevens, H.E., 1985. Some observations on the geochemistry and isotopic composition of uranium in relation to the reprocessing of nuclear fuel. J. Environ. Radioact. 2, 23–40.
Hardy, E.P., Krey, P.W., Volchok, H.L., 1973. Global inventory and distribution of fallout plutonium. Nature 241, 444–445.
Hindman, F.D., 1983. Neodymium fluoride mounting for α spectrometric determination of uranium, plutonium, and americium. Anal. Chem. 55, 2460–2461.
Hrnecek, E., Feichtinger, J., 2005. Assessment of soil contamination in the vicinity of a research reactor during the decommissioning process. J. Radioanal. Nucl. Chem. 264, 377–380.
Hrnecek, E., Aldave de las Heras, L., Betti, M., 2002. Application of microprecipitation on membrane filters for alpha spectrometry, liquid scintillation counting and mass spectrometric determination of plutonium isotopes. Radiochim. Acta. 90, 721–725.
Hu, Q.-H., Weng, J.-Q., Wang, J.-S., 2010. Sources of anthropogenic radionuclides in the environment: a review. J. Environ. Radioact. 101, 426–437.
Kershaw, P., Young, A., 1988. Scavenging of ^{234}Th in the Eastern Irish Sea. J. Environ. Radioact. 6, 1–23.
Kershaw, P.J., Woodhead, D.S., Malcolm, S.J., Allington, D.J., Lovett, M.B., 1990. A sediment history of Sellafield discharges. J. Environ. Radioact. 12, 201–241.
Kershaw, P.J., Sampson, K.E., McCarthy, W., Scott, R.D., 1995. The measurement of the isotopic composition of plutonium in an Irish Sea sediment by mass spectrometry. J. Radioanal. Nucl. Chem. 198, 113–124.
Ketterer, M.E., Szechenyi, S.C., 2008. Determination of plutonium and other transuranic elements by inductively coupled plasma mass spectrometry: a historical

perspective and new frontiers in the environmental sciences. Spectrochim. Acta. Part B. 63, 719–737.

Lee, S.H., Povinec, P.P., Wyse, E., Hotchkis, M.A.C., 2008. Ultra-low-level determination of ^{236}U in IAEA marine reference materials by ICPMS and AMS. Appl. Radiat. Isot. 66, 823–828.

Marsden, O.J., Livens, F.R., Day, J.P., Fifield, L.K., Goodall, P.S., 2001. Determination of U-236 in sediment samples by accelerator mass spectrometry. Analyst 126, 633–636.

McKay, W.A., Walker, M.I., 1990. Plutonium and americium behaviour in Cumbrian near-shore waters. J. Environ. Radioact. 12, 267–283.

Moreno, J., Vajda, N., Danesi, P.R., Larosa, J.J., Zeiller, E., Sinojmeri, M., 1997. Combined procedure for the determination of ^{90}Sr, ^{241}Am and Pu radionuclides in soil samples. J. Radioanal. Nucl. Chem. 226, 279–284.

Perna, L., Jernström, J., Aldave de las Heras, L., Hrnecek, E., de Pablo, J., Betti, M., 2005. Characterization of an Irish Sea radioactively contaminated marine sediment core by radiometric and mass spectrometric techniques. J. Radioanal. Nucl. Chem. 263, 367–373.

Salbu, B., 2001. Actinides associated with particles. In: Kudo, A. (Ed.), Plutonium in the Environment. Elsevier Science Ltd 1, pp. 121–138.

Srncik, M., Hrnecek, E., Steier, P., Wallner, A., Wallner, G., Bossew, P., 2008. Vertical distribution of ^{238}Pu, $^{239(40)}$Pu, ^{241}Am, ^{90}Sr and ^{137}Cs in Austrian soil profiles. Radiochim. Acta. 96, 733–738.

Steier, P., Bichler, M., Fifield, L.K., Golser, R., Kutschera, W., Priller, A., Quinto, F., Richter, S., Srncik, M., Terrasi, P., Wacker, L., Wallner, A., Wallner, G., Wilcken, K.M., Wild, E.M., 2008. Natural and anthropogenic ^{236}U in environmental samples. Nucl. Instrum. Methods Phys. Res. Sect. B. 266, 2246–2250.

Steier, P., Dellinger, F., Forstner, O., Golser, R., Knie, K., Kutschera, W., Priller, A., Quinto, F., Srncik, M., Terrasi, F., Vockenhuber, C., Wallner, A., Wallner, G., Wild, E.M., 2010. Analysis and application of heavy isotopes in the environment. Nucl. Instrum. Methods Phys. Res. Sect. B. 268, 1045–1049.

Vajda, N., Kim, C.-K., 2010. Determination of ^{241}Am isotope: a review of analytical methodology. J. Radioanal. Nucl. Chem. 284, 341–366.

Vockenhuber, C., Ahmad, I., Golser, R., Kutschera, W., Liechtenstein, V., Priller, A., Steier, P., Winkler, S., 2003. Accelerator mass spectrometry of heavy long-lived radionuclides. Int. J. Mass Spectrom. 223–224, 713–732.

Zhao, X.-L., Kilius, L.R., Litherland, A.E., Beasley, T., 1997. AMS measurement of environmental U-236: preliminary results and perspectives. Nucl. Instrum. Methods Phys. Res. Sect. B. 126, 297–300.