



## Speciation analysis of $^{129}\text{I}$ , $^{137}\text{Cs}$ , $^{232}\text{Th}$ , $^{238}\text{U}$ , $^{239}\text{Pu}$ and $^{240}\text{Pu}$ in environmental soil and sediment

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# Speciation Analysis of $^{129}\text{I}$ , $^{137}\text{Cs}$ , $^{232}\text{Th}$ , $^{238}\text{U}$ , $^{239}\text{Pu}$ and $^{240}\text{Pu}$ in Environmental Soil and Sediment

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## Abstract

The environmental mobility and bioavailability of radionuclides is related to their physicochemical forms, namely species. We here present a speciation analysis of important radionuclides including  $^{129}\text{I}$  (also  $^{127}\text{I}$ ),  $^{137}\text{Cs}$ ,  $^{232}\text{Th}$ ,  $^{238}\text{U}$  and plutonium isotopes ( $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ ) in soil (IAEA-375) and sediment (NIST-4354) standard reference materials and two fresh sediment samples from Øvre Heimdalsvatnet Lake, Norway. A modified sequential extraction protocol was used for the speciation analysis of these samples to obtain fractionation information of target radionuclides. Analytical results reveal that the partitioning behaviour, and thus the potential mobility and bioavailability, is exclusively featured for the individual radionuclide. Iodine is relatively mobile and more readily binds to organic matter, while plutonium is mainly related to both organic matter and nitric acid leachable fractions. Thorium is predominated in nitric acid leachable fractions and cesium is mainly observed in nitric acid and aqua regia leachable fraction and residue. Our analytical results reveal that around 50% of uranium might still remain in the residue which could not be extracted with aggressive acid, namely *aqua regia*.

**Key words:** Speciation analysis, Fractionation, Iodine, Plutonium, Uranium, Thorium, Cesium, Environment, Soil, Sediment, Partitioning, Potential mobility and bioavailability.

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## 1. Introduction

Radionuclides are present in the environment as results of natural process and/or nuclear activities of humans. Amongst a number of radionuclides,  $^{129}\text{I}$ ,  $^{137}\text{Cs}$ ,  $^{232}\text{Th}$ ,  $^{238}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  are ones of the most important radionuclides in relation to environmental radioactivity.  $^{129}\text{I}$ , a beta-emitting radioisotope of iodine, with a half-life of  $1.57 \times 10^7$  years, was mainly released from the spent nuclear fuel reprocessing plants.  $^{137}\text{Cs}$ , with a half-life of 30.17 years, is currently the largest source of environmental radioactivity generated and released from the nuclear weapon tests and Chernobyl accident, and is also one of the two principal medium-lived fission products, along with  $^{90}\text{Sr}$ .  $^{232}\text{Th}$  and  $^{238}\text{U}$ , as important natural radionuclides, have very long half-lives of  $1.405 \times 10^{10}$  and  $4.468 \times 10^9$  years, respectively. Both  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  are alpha emitters with half-lives of 24065 and 6537 years, respectively, and were mainly released through nuclear weapon tests, nuclear accidents and discharges from nuclear facilities. Due to the long-term persistence of these radionuclides in the environment and their highly radiological hazards to the environment and human health, it is imperative to understand their partitioning behaviour and to estimate their potential mobility and bioavailability through speciation analysis, which is critical for environmental risk assessment and disposal of nuclear wastes.

Sequential extraction has been proved to be a useful and practical technique for speciation analysis of various radionuclides in solid environmental samples including soils and sediments (Desideri, et al., 2002; Desideri, et al., 2001; Lucey, et al., 2007; Oughton, et al., 1992; Skipperud, et al., 2005; Riise, et al., 1990; Tessier, et al., 1979). This technique is particularly valuable when used to investigate differences in speciation as a function of source or environmental characteristics, or to compare the behaviour of different radionuclides in the same soil or sediment. However, most speciation analysis reported in literature are mainly focused on the investigation of one or two radionuclides individually, scarce data are available for the comprehensive investigation and comparison of the partitioning behaviour of several different radionuclides at a time (Blanco, et al., 2004; Napoles, et al., 2004; Lucey, et al., 2004).

In this study we performed a speciation analysis of  $^{129}\text{I}$ ,  $^{137}\text{Cs}$ ,  $^{232}\text{Th}$ ,  $^{238}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  in environmental solid samples (viz. soil and sediment) to investigate and compare their partitioning behaviour, and thus the mobility and bioavailability in the environment.

## 2. Experimental

## 2.1. Reagents and samples

$^{129}\text{I}$  standard solution (NIST-SRM-4949C, National Institute of Standard Technology, MD, USA), carrier free  $^{125}\text{I}$  (Amersham Pharmacia Biothech, Little Chalfont, Buckinghamshire, UK),  $^{127}\text{I}$  carrier (Woodward company, MICAL Specialty Chemical, New Jersey),  $^{242}\text{Pu}$  standard solution (NBL-CRM 130, New Burnswick Laboratory, Argonne, IL, USA), in-house  $^{239}\text{Pu}$  solution (Risø-DTU, Denmark) and standard solutions of U and Th (National Institute of Standard Technology, Gaithersburg, MD, USA) were used throughout the experiments. The reagents used for sequential extraction and sample preparation were of analytical grade and all solutions were prepared using de-ionized water (18 M $\Omega$ /cm) from a Milli-Q system.

In this work, four samples were selected for the speciation analysis and partitioning behaviour study including two standard reference materials, IAEA-375 and NIST-4354 as representatives of contaminated dry soil and sediment, respectively, and two sediments collected from Øvre Heimdalsvatnet Lake, Norway (61°25.10'N, 8°53.49'E) without any pre-treatment as representatives of fresh environmental sediment. IAEA-375 soil (0-20 cm) was obtained in July 1990 from the “Staryi Viskov” collective farm in Novozybkov, Brjansk, Russia, where was heavily contaminated by Chernobyl accident fallout. The IAEA-375 standard reference material, is distributed by International Atomic Energy Agency (Vienna, Austria). The accident at Chernobyl nuclear power plant took place on 26<sup>th</sup> of April, 1986 and after the initial explosion the graphite in the reactor caught fire and burned for many days. The accident resulted in a substantial release of radionuclides (e.g.,  $^{137}\text{Cs}$ , U, Pu) presumably regarded in particulate form due to the fires. NIST-4354 is freshwater lake sediment collected from the Perch Lake in the Chalk River laboratories' waste management area, Atomic Energy of Canada Limited (AECL), Canada. The sediment material is highly anoxic and contains ca. 50% organic matter by weight (Inn, 1987). The NIST-4354 sediment reference material, the material was distributed by National Institute of Standards and Technology (Gaithersburg, MD, USA) (Inn, 1987). Radionuclides contained in this sediment is primarily derived from the discharge of nuclear facility and waste treatment in the Chalk River Laboratories and radioecology studies in the lake in the 1970s. . Both reference materials have been dried, milled, sieved and sterilized before the distribution and the pH values of their solutions are both 7 measured in our work. This sediment therefore represent the materials contaminated by liquid discharges from nuclear facility. Two fresh sediments from Øvre

Heimdalsvatnet Lake were directly collected in a plastic vial in 2009 and stored at  $-4^{\circ}\text{C}$  without any treatment before analysis. There is not any direct releases/discharges of radioactivity to this lake, radionuclides in both sediments are presumably regarded as the results of atmospheric fallout during the past decades. The pH of the two Øvre Heimdalsvatnet Lake sediments solutions were both 5 as investigated in our work.

## 2.2. Sequential extraction procedure

The detailed sequential extraction procedure is listed in Table 1. 10 g of samples (two aliquots of each) were processed for sequential extraction experiment. A solution/sample ratio of 10 (v/w) was used for extraction in each step. After extraction the leachate was separated from the solid residue by centrifugation at 3000 rpm for 20 min. Before next step, the residue was rinsed with deionised water (18.2 M $\Omega$ -cm) with a water/sample ratio of 5 (v/w) under stirring for 10 min., then the washing water was combined with leachate after centrifugation. The combined solution was filtered through a membrane with a pore size of 0.45  $\mu\text{m}$  (Startorius AG, Gottingen, Germany) to remove the small particles. The remaining solid on the membrane was combined with the residue for the leaching of next step. The final solution (except the one for iodine determination) was acidified with concentrated HNO<sub>3</sub> to pH 2 immediately after filtering to prevent the adsorption of radionuclides onto the wall of the sample container.

**F1.** Deionised water was added to the solid sample and the sample was shaken for 1 hour at room temperature. This fraction is defined to represent water soluble species.

**F2.** 1 M NH<sub>4</sub>Ac-HAc (pH of soil or sediment solution) was added to the residue from F1 and the sample was shaken for 2 hours at room temperature. This fraction should reflect species associated to solids via physical sorption mechanisms, i.e. mainly exchangeable species.

**F3.** 1 M NH<sub>4</sub>Ac-HAc (pH 5) was added to the residue from F2 and the sample was stirred for 2 hours at room temperature. This fraction should reflect carbonate bound species.

It should be noted that since the pH values of the two Øvre Heimdalsvatnet Lake fresh sediment solutions are 5, extraction step for F3 was skipped for these two samples. Accordingly, F2 was regarded to represent the combination of F2 and F3, and should reflect species associated with exchangeable and carbonate components.

**F4.** 0.04 M  $\text{NH}_2\text{OH}\cdot\text{HCl}$  in 25 % (v/v) HAc (pH 2) was added to the residue from F3 and stirred in hot water bath at 80 °C with stirring for 6 hours. This fraction should reflect species associated to solids via chemical sorption mechanisms which can be released into the extraction solution with a weak reducing agent, i.e. mainly include species bound to Fe/Mn oxides.

After F4 each solid residue was divided into two sub-samples, A and B, approximately 20 % and 80% by weight, respectively, taking the approximate liquid content of the sample into account. Sub-sample A was used for the following fractionation of iodine, while sub-sample B was used for the other radionuclides.

**F5A.** 3 M NaOH was added to the residue A and the sample was stirred for 3 hours at 85 °C. The leachate was separated from the solid residue by centrifugation. 5% NaOCl was added to the residue and the sample was stirred for 2 hours at 85 °C. After separation of the leachate by centrifuge, the two leachates by NaOH and NaClO solution were combined. This fraction should reflect species associated with organic matter.

**F5B.** 30 %  $\text{H}_2\text{O}_2$  was gradually added to the residue B, in which  $\text{HNO}_3$  has already been added to final pH 2, and the sample was agitated for 5.5 hours at 80 °C with stirring. After separation of the leachate by centrifuge, 3.2 M  $\text{NH}_4\text{Ac}$  in 20 %  $\text{HNO}_3$  (v/v) was added and the extraction was continued for 30 minutes at room temperature. Similar to F5A, this fraction should reflect species associated to solids which can be released into the extraction solution with an oxidizing agent, i.e. mainly consists of organic matter related species.

**F6A.** Residues from F5A were dried in the oven at 65 °C in the original centrifuge tubes. This fraction should define the  $^{127}\text{I}$  and  $^{129}\text{I}$  species remained in the residue from F5A step.

**F6B.** 7 M  $\text{HNO}_3$  was added to the residue from F5B and heated at 80 °C while stirring for 6 hours. This fraction should reflect species associated to accessible minerals.

**F7B.** Residue from F6B was transferred to a beaker and *aqua regia* was added, and the sample was heated on a hotplate under reflux (100 °C) for approximately 4 hours. The leachate was separated by filtration through a glass fiber filter. The residue was washed with 0.2 M HCl. This fraction should reflect species associated to resistant minerals (most likely clays).

**F8B.** This fraction defines the species remaining in the residue from F7B, and was only used for  $^{137}\text{Cs}$  measurement.

### 2.3. Determination of radionuclides

The final solutions from F1 to F4 were divided into two aliquots, 20% and 80% by weight for the determination of iodine isotopes and the other radionuclides, respectively. Solution from F5A was only analyzed for iodine and solutions from F5B, F6B and F7B were analyzed for the other radionuclides except iodine isotopes.

For the determination of  $^{127}\text{I}$ , 1 mL of leachate was first taken to a vial from the collected solution and directly diluted in 20 mL of 1%  $\text{NH}_3\cdot\text{H}_2\text{O}$  solution, and measured with ICP-MS after addition of  $\text{Cs}^+$  as internal standard. To the remaining solution,  $^{125}\text{I}$  (as NaI) was added as chemical yield tracer and 2.0 mg of stable  $^{127}\text{I}$  (Woodward iodine) as a carrier. For the 20% solution from F1 to F4 and all solution from F5A, iodine was separated by solvent extraction using  $\text{CCl}_4$  after converting iodine to molecular iodine. The remaining residue (F6A) was combusted at 800 °C under oxygen flow (Hou, et al., 2009c) after addition of  $^{125}\text{I}$  tracer. The released iodine, as  $\text{I}_2$ , was trapped in alkali solution (0.4 M NaOH, 0.025 M  $\text{K}_2\text{S}_2\text{O}_5$ ). 1.0 mL solution was taken to a vial and diluted to 20 mL by deionized water for  $^{127}\text{I}$  measurement. The remained solution was used for separation of  $^{129}\text{I}$  by  $\text{CCl}_4$  extraction after 2.0 mg of  $^{127}\text{I}$  carrier was added. Iodine was separated from each fraction using a procedure reported (Englund, et al., 2007) for target preparation. The prepared AgI targets were used for  $^{129}\text{I}$  measurement by AMS using 3 MV Tandem accelerator in Uppsala University. In this work, the concentrations of total iodine isotopes ( $^{129}\text{I}$  and  $^{127}\text{I}$ ) were determined using the same procedure as for the residue (F6A) by directly analyzing original samples.

$^{137}\text{Cs}$  in the original sample as well as in each fraction was directly measured using 50 or 100 mL of aliquots taken from the separated leachates with gamma spectrometry. The samples were counted approximately 24 hours to gain enough counts in the  $^{137}\text{Cs}$  peaks.

Due to low concentration and high interference,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  in the fractions were first separated from the matrix and other interfering radionuclides before the measurement with ICP-MS. The radiochemical procedure used in this work for the separation of Pu has been reported elsewhere (Qiao, et al., 2009). TEVA<sup>®</sup> resin was used for the chemical separation and  $^{242}\text{Pu}$  was used as chemical yield

tracer. For the measurement of  $^{232}\text{Th}$  and  $^{238}\text{U}$ , about 2 mL of the each fraction was diluted in 20 mL of 0.5M  $\text{HNO}_3$  solution, and directly measured by ICP-MS after addition of indium ( $\text{InCl}_3$ ) as internal standard. The operational condition of ICP-MS and the detailed methods for the measurement of  $^{232}\text{Th}$ ,  $^{238}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  have been described elsewhere (Qiao, et al., 2009).

### 3. Results

#### 3.1. $^{129}\text{I}$ and $^{127}\text{I}$

The analytical results of  $^{127}\text{I}$  and  $^{129}\text{I}$  are shown in Table 2. The concentrations of  $^{127}\text{I}$  and  $^{129}\text{I}$  in the six fractions are in range of 0.028 - 0.840  $\mu\text{g/g}$  ( $^{127}\text{I}$ ) and 0.064 - 0.476  $\text{mBq/kg}$  ( $^{129}\text{I}$ ) for IAEA-375 soil, and 0.060 - 5.351  $\mu\text{g/g}$  ( $^{127}\text{I}$ ) and 1.014 - 23.021  $\text{mBq/kg}$  ( $^{129}\text{I}$ ) for NIST-SRM-4354 sediment, respectively. For the sediment from Øvre Heimdalsvatnet Lake, similar concentration ranges of  $^{127}\text{I}$  and  $^{129}\text{I}$  in the fractions are obtained, with the values of 0.003 - 3.439  $\mu\text{g/g}$  in sediment-1 and 0.004 - 2.341  $\mu\text{g/g}$  in sediment-2 for  $^{127}\text{I}$ , and 0.031 - 0.519  $\text{mBq/kg}$  in sediment-1 and 0.032 - 0.406  $\text{mBq/kg}$  in sediment-2 for  $^{129}\text{I}$ , respectively.

$^{129}\text{I}/^{127}\text{I}$  atomic ratios (Table 2) vary among the six fractions showing a discrepancy among samples. The  $^{129}\text{I}/^{127}\text{I}$  atomic ratios in the fractions vary from  $8 \times 10^{-8}$  to  $35 \times 10^{-8}$  for IAEA-375 soil,  $60 \times 10^{-8}$  to  $305 \times 10^{-8}$  for NIST-4354 sediment and  $2 \times 10^{-8}$  to  $140 \times 10^{-8}$  for the two Øvre Heimdalsvatnet Lake sediments, respectively. For IAEA-375 soil and NIST-4354 sediment, higher  $^{129}\text{I}/^{127}\text{I}$  atomic ratios were observed in fractions F1-F4 amongst the six fractions obtained, wherein the highest values were encountered in F2 and F4 for IAEA-375 and NIST-4354, respectively. In the two sediments from Øvre Heimdalsvatnet Lake, the  $^{129}\text{I}/^{127}\text{I}$  atomic ratios are much higher in F1 and F2+3 than in other three fractions, whereupon F1 always obtain the highest values for both sediments.

The graphic distributions of  $^{127}\text{I}$  and  $^{129}\text{I}$  fractions are shown in Fig. 1. For the entire samples, both  $^{127}\text{I}$  and  $^{129}\text{I}$  are mainly distributed in the last three fractions, i.e. F4, F5A and F6A, with the majority being found in organic matter related fraction with the proportions of  $55 \pm 7\%$  ( $^{127}\text{I}$ ) and  $38 \pm 5\%$  ( $^{129}\text{I}$ ) for IAEA-375 soil,  $76 \pm 6\%$  ( $^{127}\text{I}$ ) and  $53 \pm 9\%$  ( $^{129}\text{I}$ ) for NIST-4354 sediment,  $73 \pm 6\%$  ( $^{127}\text{I}$ ) and  $57 \pm 23\%$  ( $^{129}\text{I}$ ) for Øvre Heimdalsvatnet Lake sediment-1 and  $69 \pm 14\%$  ( $^{127}\text{I}$ ) and  $51 \pm 5\%$  ( $^{129}\text{I}$ ) for Øvre Heimdalsvatnet Lake sediment-2.

#### 3.2. $^{137}\text{Cs}$



The analytical results of  $^{137}\text{Cs}$  fractionation in IAEA-375 soil and both sediments from Øvre Heimdalsvatnet Lake are presented in Table 3 and Fig. 2.  $^{137}\text{Cs}$  concentrations were lower than the detection limit in most fractions from NIST-4354 sediment under the measurement condition; therefore no result of  $^{137}\text{Cs}$  was reported for this sample. It can be seen from Table 3 that the concentrations of  $^{137}\text{Cs}$  in the overall eight fractions vary over two to three orders of magnitude, for example, from 0.003 to 1.325 Bq/g in IAEA-375 soil, 0.010 to 0.529 Bq/g in Øvre Heimdalsvatnet Lake sediment-1 and 0.011 to 1.130 Bq/g in Øvre Heimdalsvatnet Lake sediment-2, respectively.

For IAEA-375 soil, the highest  $^{137}\text{Cs}$  proportion is observed in the fraction (F6B) treated with 7M  $\text{HNO}_3$  ( $43 \pm 9\%$ ), followed by the fraction (F7B) leached with *aqua regia* ( $37 \pm 10\%$ ), and the concentrations of  $^{137}\text{Cs}$  in other fractions are relative low, especially in water soluble fraction ( $\sim 0.1\%$ ). For the sediments from Øvre Heimdalsvatnet Lake, the majority of  $^{137}\text{Cs}$  occur in the residue ( $\sim 35\%$ ), followed by 7 M  $\text{HNO}_3$  leachable fraction F6B (30-35%). Similarly, small amount of  $^{137}\text{Cs}$  is observed in the other fractions, especially, in water soluble fraction ( $<1\%$ ).

### 3.3. $^{232}\text{Th}$ and $^{238}\text{U}$

The fractionation results for  $^{232}\text{Th}$  are shown in Table 4 and Fig. 3. The variation of  $^{232}\text{Th}$  concentrations among the investigated fractions in IAEA-375 soil (0.015– 14.750 Bq/kg) is slight larger than that in NIST-4354 sediment (0.039– 15.984 Bq/kg), while sum of  $^{232}\text{Th}$  concentration in the seven fraction are close to total concentration measured by *aqua regia* leaching.  $^{232}\text{Th}$  concentrations in all fractions are generally comparable between the two Øvre Heimdalsvatnet Lake sediments, which are both in the range of 0.0005-4.5 Bq/kg. The distribution of  $^{232}\text{Th}$  in different fractions indicates that more than 80% of  $^{232}\text{Th}$  is released by leaching with 7M  $\text{HNO}_3$  for all samples.

The concentrations and distribution of  $^{238}\text{U}$  in the fractions are summarized in Table 5 and Fig. 4. For IAEA-375 soil,  $^{238}\text{U}$  is mainly associated with 7 M  $\text{HNO}_3$  leachable fraction (F6B,  $38 \pm 10\%$ ) with the concentration of  $4.284 \pm 1.132$  Bq/kg and organic matter fraction (F5B,  $23 \pm 8\%$ ) with the concentration of  $2.612 \pm 0.905$  Bq/kg, the lowest proportion of  $^{238}\text{U}$  is obtained in water soluble fraction (F1,  $0.6 \pm 0.1\%$ ) with the concentration of  $0.068 \pm 0.007$  Bq/kg. For NIST-4354 sediment, majority  $^{238}\text{U}$  is found in carbonate fraction (F3,  $37 \pm 4\%$ ) and Fe/Mn oxides fraction (F4,  $31 \pm 12\%$ ). For the two sediment samples from Øvre Heimdalsvatnet Lake, half of  $^{238}\text{U}$  is bound to organic matter,

and 20% of  $^{238}\text{U}$  is leached with 7 M  $\text{HNO}_3$ . It should also be noticed that around 15-20% of  $^{238}\text{U}$  in the two sediments is bound to exchangeable and carbonate fractions (F2+F3).

### 3.4. $^{239}\text{Pu}$ and $^{240}\text{Pu}$

The analytical results of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  fractionation are summarized in Table 6 and Fig.5. The lowest  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  concentrations are observed in IAEA-375 soil within the ranges of 0.005– 0.062 Bq/kg ( $^{239}\text{Pu}$ ) and 0.005– 0.064 Bq/kg ( $^{239}\text{Pu}$ ), while the highest are found in NIST-4354 sediment within the ranges of 0.056– 0.813 Bq/kg ( $^{239}\text{Pu}$ ) and 0.032– 0.698 Bq/kg ( $^{239}\text{Pu}$ ).

IAEA-375 soil and one sediment sample from Lake Hemidalen have similar distributions of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  among the seven fractions, that is, about 50% of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  are bound to organic matter fraction (F5B), and about 20% of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  are released with 7 M  $\text{HNO}_3$  leaching (F6B). Differently, for the NIST-4354 and the second sediment samples from Øvre Heimdalsvatnet Lake, 50-60% of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  are associated with 7 M  $\text{HNO}_3$  leachable fraction (F6B) and about 20% with organic matter fraction (F5B).

## 4. Discussion

### 4.1. Origin and concentration of radionuclides in different samples

In this work, the concentrations of total  $^{127}\text{I}$  and  $^{129}\text{I}$  were only determined for IAEA-375 soil and the total concentration of  $^{129}\text{I}$  agrees well with the reference value (see Table 2). However, the sums of  $^{127}\text{I}$  and  $^{129}\text{I}$  concentrations in all the six fractions are slightly lower compared with the corresponding total concentrations. This might be attributed to the loss of iodine isotopes during the analytical procedure since iodine is readily volatile.

As can be seen from Table 3, within the analytical uncertainty, the concentrations of total  $^{137}\text{Cs}$  are in good agreement with the sum of  $^{137}\text{Cs}$  concentration in the overall fractions for all samples. While, big difference between the two sediments from Øvre Heimdalsvatnet Lake is observed with respect to  $^{137}\text{Cs}$  concentration in each fraction and original sample. It reveals that the  $^{137}\text{Cs}$  concentrations vary significantly with the sampling locations even though they are from the same lake. Differently, this phenomenon was not observed for other radionuclides including  $^{129}\text{I}$ ,  $^{232}\text{Th}$ ,  $^{238}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  in this work. Since there is no direct radioactive source in the local area of this lake,  $^{137}\text{Cs}$  in the lake sediment is mainly originated from the fallout of both weapons testing major in 1960's and Chernobyl accident

in 1986, where a slightly high deposition of Chernobyl fallout was observed. The input of  $^{137}\text{Cs}$ , especially  $^{137}\text{Cs}$  carried suspended particulates; from the surround area through moving by river water is probably one of the major sources of  $^{137}\text{Cs}$  in the sediment, causing an inhomogeneous distribution of  $^{137}\text{Cs}$  and its speciation. The high concentration of  $^{137}\text{Cs}$  in the IAEA soil is attributed to the Chernobyl accident. Because the IAEA-375 soil standard reference was prepared using the surface soil collected from the heavily Chernobyl contamination area with a dilution using less contamination soil. The total concentration of  $^{137}\text{Cs}$  in the two lake sediment is comparable with that in the IAEA-375 soil, confirming the high Chernobyl fallout in the Lake area in Norway. The certified value of total  $^{137}\text{Cs}$  in the NIST-4354 sediment is only 0.0334Bq/g (decay correction to 2009), this is a typical value for atmospheric fallout from weapons testing, indicating limited discharges of  $^{137}\text{Cs}$  from nuclear facilities of Chalk River laboratory. This value is about 100 times lower than the IAEA-375 soil and Øvre Heimdalsvatnet Lake sediment, confirmed a high contamination of the IAEA-375 soil and sediment of lake Øvre Heimdalsvatnet. The fractionation results of  $^{232}\text{Th}$  and  $^{238}\text{U}$  (see Tables 4 and 5) for the two reference materials (IAEA-375 and NIST-4354) demonstrate that the sums of concentrations in the seven fractions are lower compared with the reference values of total concentrations. This might be attributed to that U and Th existing in the crystal of minerals or form inert/refractory compounds in the sample cannot be extracted out even with *aqua regia*.

For IAEA-375 soil, the sum of  $^{239+240}\text{Pu}$  concentrations in the seven fractions are comparable with the reference values, although a slightly lower value is obtained for the total concentration of  $^{239+240}\text{Pu}$  measured in this work after leaching with *aqua regia* (see Table 6). The total concentration of  $^{239+240}\text{Pu}$  in NIST-4354 sediment determined in this work agrees with the reference value within the analytical uncertainty. However, the sums of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  concentrations in all 7 fractions for NIST-4354 sediment are lower than their corresponding total concentrations recommended. It reveals part of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  might still remain in the final residue of NIST-4354 sediment.

The differences of total concentrations of radionuclides among different samples are obviously notable, which is attributed to the different sources of radionuclide in these samples. NIST-4354 sediment has relatively higher concentration of  $^{129}\text{I}$  and isotopes of plutonium, but lower  $^{137}\text{Cs}$  concentration. The concentrations of uranium are similar in the four investigated samples, and the concentration of thorium in NIST-4354 sediment is similar to that in the IAEA-375 soil, but higher

than that in the two fresh lake sediment samples. NIST-4354 was collected from the Perch Lake in the Chalk River Laboratories' waste management area, AECL. The high level of  $^{129}\text{I}$  and radioisotopes of plutonium in this sediment might be attributed to the direct contamination of this sediment by the discharges from nuclear facility and waste treatment in the Chalk River Laboratories and radioecology studies in the lake in the 1970s. The  $^{129}\text{I}/^{127}\text{I}$  ratio ( $(6\sim 34)\times 10^{-7}$ ) is 3-4 orders magnitude higher than the global fallout level, and even comparable with level in marine sediment in the North Sea and Irish Sea which were heavily contaminated by European reprocessing plants in Sellafield (UK) and La Hague (France). The IAEA-375 soil was prepared by dilution of Chernobyl accident contaminated soil with some less contaminated soil, therefore has moderate radionuclide concentrations. The Lake Øvre Heimdalsvatnet sediments are mainly the result of atmospheric fallout from decades past. The less difference in the concentration of uranium and thorium in the samples might reflect the natural source of these two elements in the investigated samples.

## **4.2. Partitioning behaviour of nuclides in different samples**

Different radionuclides behave differently regarding to the fractionation due to different physicochemical behaviour of radionuclides. A number of environmental variables such as redox potentials, carbonates, inorganic and organic ligands, sorption of environmental surfaces, residence time, and pH also affect the partitioning behaviour of these radionuclides. Full appreciation of the behaviour of radionuclide in environment requires knowledge of the initial character of the contaminating events as well.

### **4.2.1. $^{127}\text{I}$ and $^{129}\text{I}$**

For all the samples analysed in this work, the largest proportions of  $^{129}\text{I}$  and  $^{127}\text{I}$ , are associated with organic matter fractions (see Fig. 1). Our results are consistent with the observation showing that in some soils/sediments iodine becomes to a large extent bound to organic substances by microbial reactions involving enzymatic oxidation and reduction (Behrens, 1982).

For IAEA-375 soil,  $^{129}\text{I}/^{127}\text{I}$  atomic ratios ( $2 \times 10^{-7}$ ) in exchangeable fraction F1 are two orders of magnitude higher than the ones related to global fallout ( $10^{-9}$ ), which indicate the Chernobyl signature for  $^{129}\text{I}$ . It is known that ca. 6 kg of  $^{129}\text{I}$  has been released from the Chernobyl accident and IAEA-375 soil was collected from the heavily Chernobyl-contaminated area and diluted with less contaminated

soil. In addition,  $^{129}\text{I}/^{127}\text{I}$  atomic ratios in F1 from IAEA-375 soil are also comparable with the values from terrestrial area in Western Europe, e.g. Denmark ( $7.45 \times 10^{-8}$  in soil and  $(1.37\text{-}2.77) \times 10^{-7}$  in grass) (Hou, et al., 1999). In NIST-4354 sediment,  $^{129}\text{I}/^{127}\text{I}$  atomic ratios in F1 reach to  $2 \times 10^{-6}$ , as high as the values observed in North Sea (Hou, et al., 2007) which is related to the discharge from La Hogue (France) and Sellafield (U.K.) reprocessing plants. Since NIST-4354 sediment was collected from the Perch Lake in the Chalk River Laboratories' waste management area, the high  $^{129}\text{I}/^{127}\text{I}$  value in this sediment is clearly attributed to the contamination of the nuclear activities in Chalk River Laboratories to this lake. For the two sediment samples from Øvre Heimdalsvatnet Lake,  $^{129}\text{I}/^{127}\text{I}$  atomic ratios ( $(1\sim 140) \times 10^{-8}$ ) are comparable with the values obtained for precipitation in Denmark ( $(5\sim 76) \times 10^{-8}$ ) (Hou, et al., 2009b) which is attributed to the  $^{129}\text{I}$  releases from La Hogue and Sellafield reprocessing plants both direct atmospheric release and re-emission from the North Sea, Irish Sea and English Channel.

It is noticed that for all the samples analyzed in this work, the relatively higher  $^{129}\text{I}/^{127}\text{I}$  atomic ratios are dominated in the first three or four fractions (F1-F4 for IAEA-375 and NIST-4354, F1-F3 for the two fresh lake sediments), from F4 or F5 until the last fraction,  $^{129}\text{I}/^{127}\text{I}$  atomic ratios are 3-50 times lower depending on the sample. This might be attributed to the slow isotope exchange rate of  $^{129}\text{I}$ . With the decrease of the accessibility of the geochemical phase, the abundance of  $^{129}\text{I}$  decreases. The different  $^{129}\text{I}/^{127}\text{I}$  atomic ratio distribution patterns between the reference materials (namely, IAEA-375 soil and NIST4354 sediment) and the two fresh lake sediments might be attributed to the property of samples including sample treatment process, since the two reference materials have been air dried, milled and sterilized before the distribution, while the two lake sediments were wet and fresh samples, which were directly delivered to the laboratory for the speciation analysis after sampling.

The distribution of  $^{129}\text{I}/^{127}\text{I}$  atomic ratio varies with fractions as well as with samples, suggesting the different partitioning behaviour between  $^{127}\text{I}$  and  $^{129}\text{I}$ . As a natural isotope of iodine,  $^{127}\text{I}$  is originated in the environment from the bedrock, plants as well as the atmospheric precipitation.  $^{129}\text{I}$  detected in the environment is mainly anthropogenic source through human nuclear activities since 1940's. The released  $^{129}\text{I}$  was gradually incorporated into environmental components with time and weathering, but the distribution of  $^{129}\text{I}$  in the environment has not reached to physicochemical equilibrium status with  $^{127}\text{I}$ . The reported natural atomic ratio of  $^{129}\text{I}/^{127}\text{I}$  is about  $10^{-12}$  and has being increased to  $10^{-10}$  to  $10^{-4}$

in terrestrial environment due to releases from human nuclear activities (Hou, et al., 2009c). The  $^{129}\text{I}/^{127}\text{I}$  atomic ratios for all samples analyzed in this work are in the range of  $10^{-6}$  to  $10^{-8}$ , confirming the sources of  $^{129}\text{I}$  in these samples are from the external fallout or discharge.

#### 4.2.2. $^{137}\text{Cs}$

Different from iodine,  $^{137}\text{Cs}$  is more readily associated to mineral crystals (see Fig. 2) for all samples analysed in this work. The results demonstrates a  $^{137}\text{Cs}$  in soil and sediment is less leachable once it is integrated into the sample matrix, thus revealing lower potential mobility and availability of  $^{137}\text{Cs}$  in the environment, which is consistent with the results reported by many researchers (Desideri, et al., 2001; Lujanienė, et al., 1999; Hou, et al., 2003).  $^{137}\text{Cs}$  remained in the residue (F8B) might be the similar deposits as natural caesium in the environment, i.e. pegmatite. The highest release of  $^{137}\text{Cs}$  from the two fresh lake sediments were observed in 7M  $\text{HNO}_3$  leaching fraction and less in the aqua regia fraction, which means these sediments are probably contain more assessable minerals than resistant minerals (most likely clays) or the binding of  $^{137}\text{Cs}$  to resistant mineral is relatively slower than to accessible mineral. By contrast, the IAEA-375 shows the highest  $^{137}\text{Cs}$  release in the 7 M  $\text{HNO}_3$  leachate and nearly as much as from aqua regia fraction, perhaps indicating a different clay mix compared with the two fresh lake sediments.

#### 4.2.3. $^{232}\text{Th}$ and $^{238}\text{U}$

U and Th are naturally occurring radioactive elements which have persisted in the environmental since the formation of the Earth. Thorium has solely one stable oxidation state, IV, and occurs in several minerals including thorite ( $\text{ThSiO}_4$ ), thorianite ( $\text{ThO}_2 + \text{UO}_2$ ) and monazite (Lehto and Hou, 2010). Uranium has stable oxidation states of IV and VI, and the typical occurrence of uranium in soil and sediment is probably the same form as it is found in ores, i.e. pitchblende (a mixture of  $\text{UO}_2$  and  $\text{UO}_3$ ) and carnotite ( $\text{K}_2(\text{UO}_2)_2(\text{VO}_4)_2 \cdot 3\text{H}_2\text{O}$ ) (Lehto and Hou, 2010). Environmental conditions, such as pH and Eh, will control the distribution of oxidation states thus influence the physicochemical associations of radionuclides, and uranium is a classic example. Both Th and U in the tetravalent state are extremely resistant to leaching (Hanson, 1980). However, the oxidation of U(IV) to U(VI) results in much higher uranium motilities in the environment due to the formation of stronger uranyl carbonate complexes.

Our results on the fractionation of thorium reflect natural signatures (see Fig. 3), as it can be seen that 80% of Th is released by 7 M HNO<sub>3</sub> extraction for all samples suggesting Th is moderately matrix bound. This is coherent with the former conclusion that Th behaves immobile in soil or sediment (Valkovic, 2000). From Fig. 4, the highest proportion (ca. 50%) of U is observed in Fe/Mn oxides for the two fresh lake sediments indicating uranium is dominantly in uranyl cation form which is chemically bound to Fe/Mn oxides. For IAEA-375 soil, the highest release (ca. 40%) of uranium is from nitric acid leachable fraction, 25% of uranium is bound to organic and 20% of uranium forms complexes with carbonate. Since the partitioning pattern of uranium in IAEA-375 soil is so different from the two fresh lake sediments, it is presumed that the high release of uranium from 7 M HNO<sub>3</sub> extraction is tapping the Chernobyl signature in this fraction. In the Chernobyl accident, after the initial explosion, the graphite in the reactor caught fire and burnt for many days, uranium might form refractory oxides before settling to the soil. This is confirmed by relative higher <sup>235</sup>U (1.2 Bq/kg) in IAEA-375 corresponding to <sup>235</sup>U/<sup>238</sup>U atomic ratio of  $8.36 \times 10^{-3}$  comparing with the natural abundance of <sup>235</sup>U of 0.72% (Strachnov et al. 1996). By contrast, NIST-4354 sediment is anoxic sediment and has very high organic content. Uranium partitioning pattern from NIST-4354 is different from IAEA-375 and shows high association to the carbonate and Fe/Mn oxides, which is consistent with the type of the sediment that is holding the extractable uranium.

In addition, the two fresh sediments from Øvre Heimdalsvatnet Lake had not been dried but used as wet. This difference may have an effect on the behaviour of uranium: in dried sediments stored in aerobic conditions may have larger proportions of oxidized uranium than those used as wet. This might be a clue for the possible remaining of uranium in the final residue from IAEA-375 and NIST-4354, as we observed that large differences existed between the sums of U concentrations in F1-F7B compared with the reference values.

#### 4.2.4. <sup>239</sup>Pu and <sup>240</sup>Pu

Distribution of Pu fractions indicates ca. 50% of Pu (see Fig. 5) is distributed in organic matter fraction and ca. 20% in 7 M HNO<sub>3</sub> leachate for both IAEA-375 soil and the fresh lake sediment-1. Large variations were observed in the measurement of <sup>239</sup>Pu, <sup>240</sup>Pu concentrations in 7 M HNO<sub>3</sub> fraction for fresh lake sediment-2, which might be caused by a sort of contamination and consequently bring a different plutonium distribution pattern in fresh lake sediment-2. On the contrary, ca. 50% of Pu

is released by 7 M HNO<sub>3</sub> leaching and moderate amount (20%) of Pu is bound to organic matter for NIST-4354 sediment. Plutonium have multiple oxidation states including Pu(III), Pu(IV), PuO<sub>2</sub><sup>+</sup>, PuO<sub>2</sub><sup>2+</sup> in natural environment and characterization studies suggested that pentavalent plutonium and organically complexed Pu(IV) are important mobile forms under aerobic conditions . Therefore, it presumably suggests the binding of plutonium with organic content in NIST-4354 sediment should be somehow prohibited by the anoxic environment.

The <sup>240</sup>Pu/<sup>239</sup>Pu atomic ratios are also given in Table 6. Both two fresh lake sediments have the lowest <sup>240</sup>Pu/<sup>239</sup>Pu atomic ratios in the Fe/Mn oxides fraction, and the highest ratios in 7 M HNO<sub>3</sub> fraction The different values of <sup>240</sup>Pu/<sup>239</sup>Pu atomic ratios observed between the two fresh lake sediment might be attributed to the low concentrations of <sup>239</sup>Pu and <sup>240</sup>Pu in most fractions and the relatively high uncertainties. By contrast, for IAEA-375 soil and NIST-4354 sediment, the highest <sup>240</sup>Pu/<sup>239</sup>Pu atomic ratios are found in Fe/Mn fraction, the lowest ratios in the organic matter for IAEA-375 soil and in the exchangeable fraction for NIST-4354 sediment. These results should be signatures related to the differing anthropogenic source terms, particular for Chernobyl and the AECL site, however, they are still not well interpreted in this work due to the limited data and large variations. It is noted that the <sup>240</sup>Pu/<sup>239</sup>Pu atomic ratios for some measurements, e.g., in Fe/Mn oxides fraction from IAEA-375 soil (<sup>240</sup>Pu/<sup>239</sup>Pu (atom/atom) = 0.72 ± 0.41), are higher than the one previously reported. This might be a consequence of low concentration of <sup>239</sup>Pu and <sup>240</sup>Pu in the fraction along with the high uncertainty in the measurement.

### 4.3. Comparison with other reported results

Association of <sup>238</sup>U and <sup>239+240</sup>Pu in NIST-SRM-4354 sediment has been studied by Outola et al. (2009) using a five-step sequential extraction method. Theirs results showed that most of U (~75%) presented in the fraction attacked with ammonium acetate which can be regarded as carbonate associated fraction, while most of Pu (~75%) was found in the fraction extracted with hydrogen peroxide which can be regarded as organic matter related fraction. Our results are not notably comparable with their results, since a different sequential extraction procedure was utilized in this work. As the fractionation analysis is very critical and sensitive to the sequential extraction procedure, even using the same procedure but conducted in different laboratories, different results still can be obtained due to different detailed operational conditions (Hou, et al., 2009a). No fractionation results



for  $^{129}\text{I}$ ,  $^{232}\text{Th}$  and  $^{137}\text{Cs}$  in NIST-4354 sediment have been reported so far. But since iodine is readily to bind to organic matter, being a fresh lake sediment which contains 50% of organic content by weight (Inn, 1987), it is reasonable to observe around 75% of  $^{127}\text{I}$  and 50% of  $^{129}\text{I}$  in the organic matter fraction. The fractionation analysis of IAEA-375 soil has not been well studied, while Hou et al. (2003) have studied the fractionation of  $^{129}\text{I}$  and  $^{137}\text{Cs}$  in Chernobyl contaminated soil, and reported similar results as our work for IAEA-375 soil, which was prepared by dilution of Chernobyl accident contaminated soil with some less contaminated soil.

#### **4.4. Sequential extraction procedure applied in this work**

It should be noted that in the widely used sequential extraction procedures the organic matter associated fraction is normally extracted with  $\text{H}_2\text{O}_2\text{-HNO}_3$  mixture, however leached iodine is unstable in  $\text{H}_2\text{O}_2\text{-HNO}_3$  solution since iodine is readily converted to  $\text{I}_2$  in acidic solution and thus lost from the leachate (Hou, et al., 2003). Therefore, the iodine bound to organic matter has to be calculated as the difference of iodine content in the samples before and after  $\text{H}_2\text{O}_2\text{-HNO}_3$  leaching. For this purpose, alkaline fusion or combustion processing needs to be applied to analyse the solid sample for  $^{129}\text{I}$  and  $^{127}\text{I}$  which makes the analytical workload more burdensome. NaOH has also been often used to extract organic bound iodine (Englund, et al., 2010; Sheppard and Thibault, 1992). In this work, we used 3M NaOH solution to extract organic matter associated iodine in the leachate, and 5% NaOCl solution to further decompose organic matter meanwhile to oxidize iodine to iodate which could remain in the leachate. In addition, TMAH (tetramethylammonium hydroxide,  $(\text{CH}_3)_4\text{NOH}$ ) as another alkali solution has also been applied instead of NaOH to extract iodine, mainly organic associated iodine from soil and sediment, but in principle the function of NaOH and TMAH are identical (Yamada, et al., 2002).

The sequential extraction procedure applied in this work is a very useful and comprehensive tool to investigate the characteristic of partitioning behaviour for different nuclides in different samples even though there might be somehow shortcomings to clearly segregate one fraction from another and the crossover between fractions could be encountered. For example, in the F5B fraction, organic bound nuclide is released into the leachate by decomposing organic matter with  $\text{H}_2\text{O}_2$ , but it might also be possible that, to some extent, U(IV) in the minerals phase is oxidized to U(VI) and released into the leachate.

## 5. Conclusion

From the above results and discussion, we can conclude that the partitioning behaviour varies from one radionuclide to another. Iodine is more readily to be bound to the organic matter fraction and Pu is mainly related to organic matter and 7 M HNO<sub>3</sub> leachable fractions. The most proportion of Th was predominated in 7 M HNO<sub>3</sub> leachable fraction while Cs was strongly bound to 7 M HNO<sub>3</sub> and *aqua regia* leachable fractions and the final residue. About half of U might be still remaining in the residue after multi-step extraction, even could not be leached with *aqua regia*. It might be possible to assume that the potential mobility and bioavailability of these radionuclides in the four samples investigated in this work is generally in a descending order as: I > Pu > Th > Cs > U. It is also important to interpret the partitioning behaviour of individual radionuclide by radio-ecologically linking the character of the source terms of radionuclides in different samples. For example, the low leachability of uranium from IAEA-375 soil shows good consistency with the highly fired particulate form of uranium in this soil. The atomic ratios of <sup>129</sup>I/<sup>127</sup>I are different for different fractions in all samples, revealing different originations of <sup>127</sup>I and <sup>129</sup>I.

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Table 1. Sequential fractionation procedure used in this work

Fraction	Main associated species	measured isotopes	Extracting Reagents	pH	Temperature	Contact time, hr
F1	Water soluble	$^{127}\text{I}$ , $^{129}\text{I}$ , $^{137}\text{Cs}$ , $^{232}\text{Th}$ , $^{238}\text{U}$ , $^{239}\text{Pu}$ , $^{240}\text{Pu}$	Water (Milli Q)		Room temperature	2
F2	Exchangeable	$^{127}\text{I}$ , $^{129}\text{I}$ , $^{137}\text{Cs}$ , $^{232}\text{Th}$ , $^{238}\text{U}$ , $^{239}\text{Pu}$ , $^{240}\text{Pu}$	1M $\text{NH}_4\text{Ac}$ -HAc	7	20°C	2
F3	Carbonates	$^{127}\text{I}$ , $^{129}\text{I}$ , $^{137}\text{Cs}$ , $^{232}\text{Th}$ , $^{238}\text{U}$ , $^{239}\text{Pu}$ , $^{240}\text{Pu}$	1M $\text{NH}_4\text{Ac}$ -HAc	5	20°C	2
F4	Fe/Mn oxides	$^{127}\text{I}$ , $^{129}\text{I}$ , $^{137}\text{Cs}$ , $^{232}\text{Th}$ , $^{238}\text{U}$ , $^{239}\text{Pu}$ , $^{240}\text{Pu}$	0.04M $\text{NH}_2\text{OH}$ HCl in 25 % (v/v) HAc	2	80°C	6
F5A*	Organic matter	$^{127}\text{I}$ , $^{129}\text{I}$	step (1) 3 M NaOH step (2) 5% NaOCl to residue		85°C 85°C	3 2
F6A*	Residue	$^{127}\text{I}$ , $^{129}\text{I}$	The residue is decomposed by combustion			
F5B§	Organic matter	$^{137}\text{Cs}$ , $^{232}\text{Th}$ , $^{238}\text{U}$ , $^{239}\text{Pu}$ , $^{240}\text{Pu}$	step (1) 30 % $\text{H}_2\text{O}_2$ - $\text{HNO}_3$ (pH2) step (2) 3.2 M $\text{CH}_3\text{COONH}_4$ ( $\text{NH}_4\text{Ac}$ ) in 20 % (v/v) $\text{HNO}_3$	2	80°C 20°C	5.5 0.5
F6B§	7M nitric acid leachable	$^{137}\text{Cs}$ , $^{232}\text{Th}$ , $^{238}\text{U}$ , $^{239}\text{Pu}$ , $^{240}\text{Pu}$	7 M $\text{HNO}_3$		80°C	6
F7B§	<i>Aqua regia</i> leachable	$^{137}\text{Cs}$ , $^{232}\text{Th}$ , $^{238}\text{U}$ , $^{239}\text{Pu}$ , $^{240}\text{Pu}$	<i>Aqua regia</i>		Heat under reflux (100°C)	2
F8B§	Residue	$^{137}\text{Cs}$				

\* 20% of residue from F4 (by weight) was used; § 80% of residue from F4 (by weight) was used;

Table 2. Analytical results of  $^{127}\text{I}$  and  $^{129}\text{I}$  fractionation in soil and sediment samples

IAEA-SRM-375 soil			
Fraction	$^{127}\text{I}$ Concentration, $\mu\text{g/g}$	$^{129}\text{I}$ Concentration, $\text{mBq/kg}$	$^{129}\text{I}/^{127}\text{I}$ $10^{-8}$ , at/at
F1	$0.064 \pm 0.003$	$0.106 \pm 0.014$	$25.01 \pm 3.51$
F2	$0.028 \pm 0.003$	$0.064 \pm 0.006$	$34.54 \pm 4.46$
F3	$0.054 \pm 0.005$	$0.120 \pm 0.016$	$33.76 \pm 5.30$
F4	$0.279 \pm 0.016$	$0.345 \pm 0.018$	$18.61 \pm 1.43$
F5A	$0.840 \pm 0.109$	$0.476 \pm 0.067$	$8.53 \pm 1.63$
F6A	$0.251 \pm 0.010$	$0.138 \pm 0.024$	$8.29 \pm 1.47$
Sum(1-6)	$1.515 \pm 0.144$	$1.249 \pm 0.144$	$12.41 \pm 1.86$
Total (combustion)	$1.94 \pm 0.05$	$1.671 \pm 0.038$	$12.94 \pm 1.70$
Recommended value	-	$1.7 \pm 0.4$	-
NIST-SRM-4354 lake sediment			
Fraction	$^{127}\text{I}$ Concentration, $\mu\text{g/g}$	$^{129}\text{I}$ Concentration, $\text{mBq/kg}$	$^{129}\text{I}/^{127}\text{I}$ $10^{-8}$ , at/at
F1	$0.118 \pm 0.004$	$1.855 \pm 0.221$	$236.89 \pm 29.31$
F2	$0.064 \pm 0.009$	$1.138 \pm 0.157$	$267.36 \pm 53.99$
F3	$0.060 \pm 0.012$	$1.014 \pm 0.078$	$256.36 \pm 53.86$
F4	$0.580 \pm 0.050$	$11.763 \pm 1.262$	$305.35 \pm 42.02$
F5A	$5.351 \pm 0.446$	$23.021 \pm 4.029$	$64.75 \pm 12.55$
F6A	$0.903 \pm 0.046$	$4.835 \pm 0.307$	$80.59 \pm 6.56$
Sum(1-6)	$7.075 \pm 0.567$	$43.626 \pm 6.054$	$92.81 \pm 14.87$
Total (combustion)	NM	NM	NM
Recommended value	-	-	-
Lake Heimdalen sediment-1			
Fraction	$^{127}\text{I}$ Concentration, $\mu\text{g/g}$	$^{129}\text{I}$ Concentration, $\text{mBq/kg}$	$^{129}\text{I}/^{127}\text{I}$ $10^{-8}$ , at/at
F1	$0.003 \pm 0.001$	$0.031 \pm 0.005$	$139.40 \pm 37.30$
F2+F3	$0.011 \pm 0.001$	$0.036 \pm 0.009$	$51.85 \pm 13.77$
F4	$0.646 \pm 0.080$	$0.101 \pm 0.011$	$2.35 \pm 0.38$
F5A	$3.439 \pm 0.286$	$0.519 \pm 0.213$	$2.27 \pm 0.95$
F6A	$0.626 \pm 0.078$	$0.231 \pm 0.032$	$5.54 \pm 1.03$
Sum(1-6)	$4.725 \pm 0.446$	$0.917 \pm 0.270$	$2.92 \pm 0.90$
Total (combustion)	NM	NM	NM
Lake Heimdalen sediment-2			
Fraction	$^{127}\text{I}$ Concentration, $\mu\text{g/g}$	$^{129}\text{I}$ Concentration, $\text{mBq/kg}$	$^{129}\text{I}/^{127}\text{I}$ $10^{-8}$ , at/at
F1	$0.004 \pm 0.001$	$0.032 \pm 0.004$	$123.15 \pm 31.88$
F2+F3	$0.016 \pm 0.001$	$0.072 \pm 0.007$	$69.55 \pm 7.41$
F4	$0.596 \pm 0.065$	$0.160 \pm 0.008$	$4.05 \pm 0.49$
F5A	$2.341 \pm 0.462$	$0.406 \pm 0.039$	$2.61 \pm 0.57$
F6A	$0.450 \pm 0.017$	$0.127 \pm 0.008$	$4.25 \pm 0.30$
Sum(1-6)	$3.406 \pm 0.546$	$0.798 \pm 0.065$	$3.52 \pm 0.63$
Total (combustion)	NM	NM	NM

\* NM: not measured

Table 3. Analytical results of  $^{137}\text{Cs}$  fractionation in different samples

$^{137}\text{Cs}$ Fraction	IAEA-SRM-375 soil	$^{137}\text{Cs}$ Fraction	Lake Heimdalen sediment-1	Lake Heimdalen sediment-2
	Concentration Bq/g		Concentration Bq/g	Concentration Bq/g
F1	0.003 ± 0.002	F1	0.010 ± 0.003	0.011 ± 0.008
F2	0.148 ± 0.047	F2+F3	0.178 ± 0.030	0.225 ± 0.063
F3	0.060 ± 0.007	F4	0.144 ± 0.021	0.252 ± 0.037
F4	0.026 ± 0.004	F5B	0.098 ± 0.029	0.127 ± 0.031
F5B	0.117 ± 0.043	F6B	0.402 ± 0.068	1.087 ± 0.088
F6B	1.325 ± 0.282	F7B	0.120 ± 0.045	0.295 ± 0.066
F7B	1.122 ± 0.314	F8B	0.529 ± 0.030	1.130 ± 0.004
F8B	0.271 ± 0.028	Sum(1-8)	1.480 ± 0.225	3.126 ± 0.297
Sum(1-8)	3.071 ± 0.728	Total	1.469 ± 0.044	3.185 ± 0.052
Total	3.080 ± 0.056			
Recommended value	3.553 ± 0.054*			

\*The value was recalculated for decay correction to reference date: 1st March 2009.



Table 4. Fractionation of  $^{232}\text{Th}$  in different samples

$^{232}\text{Th}$ Fraction	IAEA-SRM-375 soil	NIST-SRM-4354 lake sediment
	Concentration Bq/kg	Concentration Bq/kg
F1	$0.016 \pm 0.001$	$0.236 \pm 0.064$
F2	$0.017 \pm 0.003$	$0.197 \pm 0.050$
F3	$0.099 \pm 0.013$	$0.590 \pm 0.055$
F4	$0.036 \pm 0.004$	$0.546 \pm 0.239$
F5B	$0.015 \pm 0.016$	$0.039 \pm 0.022$
F6B	$14.750 \pm 1.260$	$15.984 \pm 3.716$
F7B	$2.022 \pm 0.734$	$0.305 \pm 0.091$
Sum(1-7)	$16.956 \pm 2.031$	$17.897 \pm 4.237$
Total ( <i>aqua regia</i> )	$13.897 \pm 1.390$	$17.165 \pm 2.717$
Recommended value	$20.5 \pm 1.4$	$26.8 \pm 0.9$

$^{232}\text{Th}$ Fraction	Lake Heimdalen sediment-1	Lake Heimdalen sediment-2
	Concentration Bq/kg	Concentration Bq/kg
F1	$0.0007 \pm 0.0002$	$0.0005 \pm 0.0002$
F2+F3	$0.050 \pm 0.033$	$0.028 \pm 0.004$
F4	$0.303 \pm 0.066$	$0.255 \pm 0.030$
F5B	$0.002 \pm 0.001$	$0.033 \pm 0.038$
F6B	$4.493 \pm 1.402$	$4.661 \pm 0.353$
F7B	$0.563 \pm 0.047$	$0.813 \pm 0.169$
Sum(1-7)	$5.412 \pm 1.550$	$5.790 \pm 0.594$
Total ( <i>aqua regia</i> )	$4.870 \pm 0.487$	$5.816 \pm 0.582$

Table 5. Fractionation of  $^{238}\text{U}$  in different samples

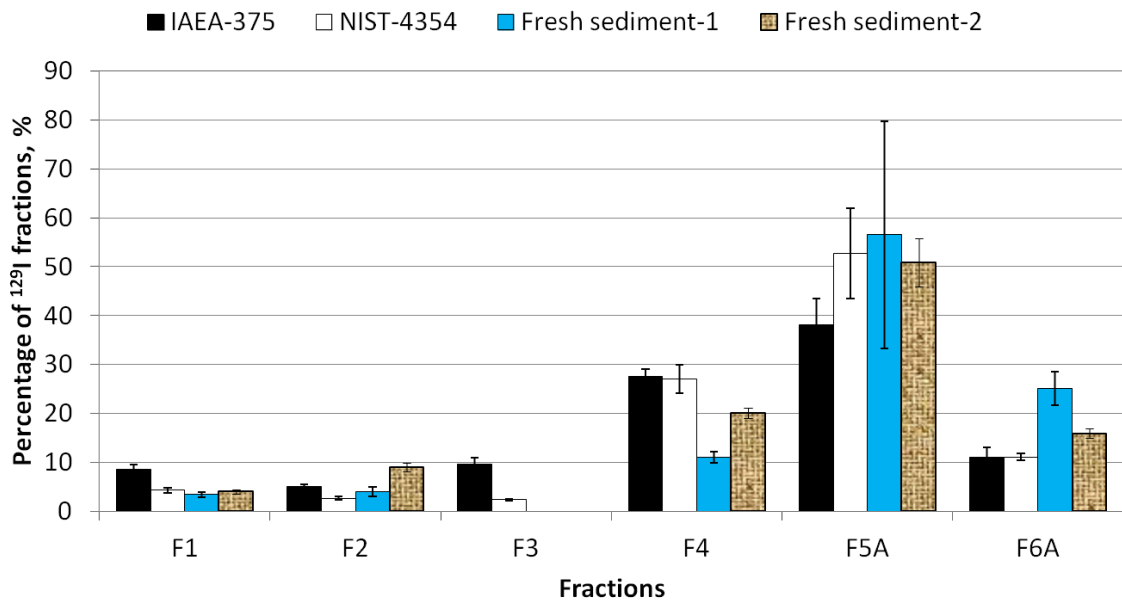
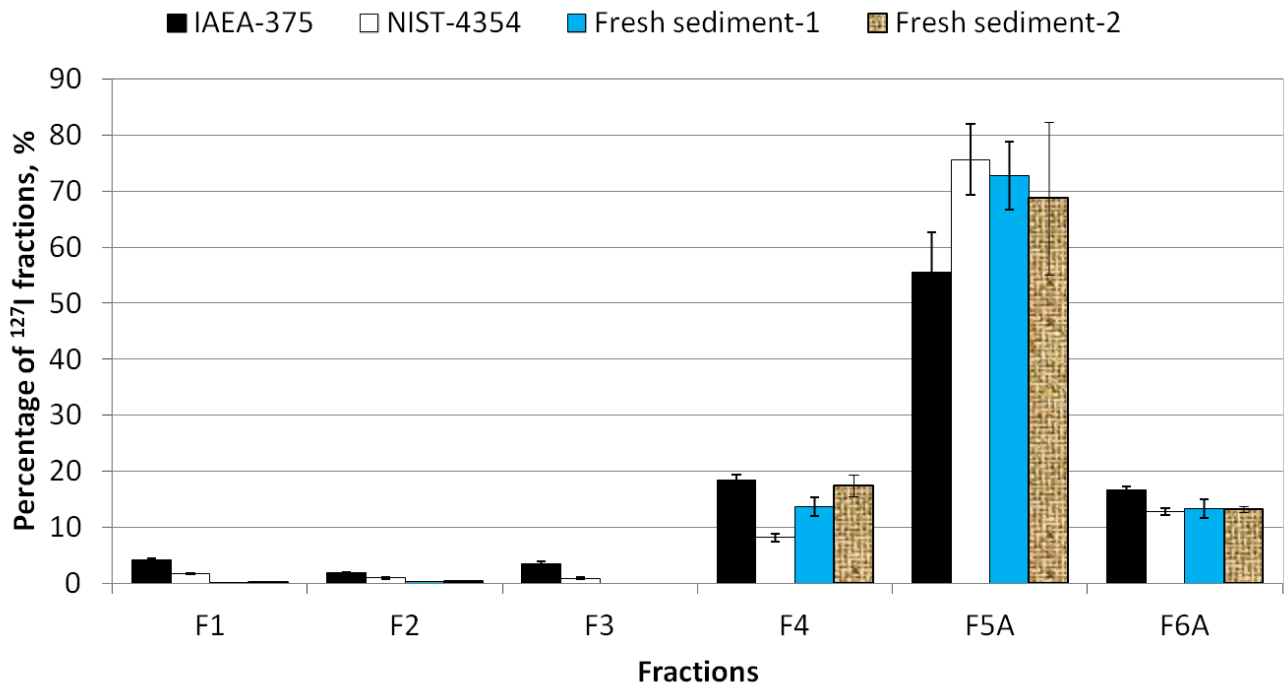
$^{238}\text{U}$ Fraction	IAEA-SRM-375 soil	NIST-SRM-4354 lake sediment
	Concentration Bq/kg	Concentration Bq/kg
F1	$0.068 \pm 0.007$	$0.153 \pm 0.040$
F2	$0.201 \pm 0.031$	$0.520 \pm 0.077$
F3	$2.129 \pm 0.195$	$3.340 \pm 0.317$
F4	$0.980 \pm 0.086$	$2.832 \pm 1.077$
F5B	$2.612 \pm 0.905$	$1.594 \pm 0.254$
F6B	$4.284 \pm 1.132$	$0.450 \pm 0.071$
F7B	$0.889 \pm 0.344$	$0.126 \pm 0.013$
Sum(1-7)	$11.163 \pm 2.699$	$9.014 \pm 1.850$
Total (aqua regia)	-	-
Recommended value	$24.4 \pm 5.4$	$17.400 \pm 0.418$

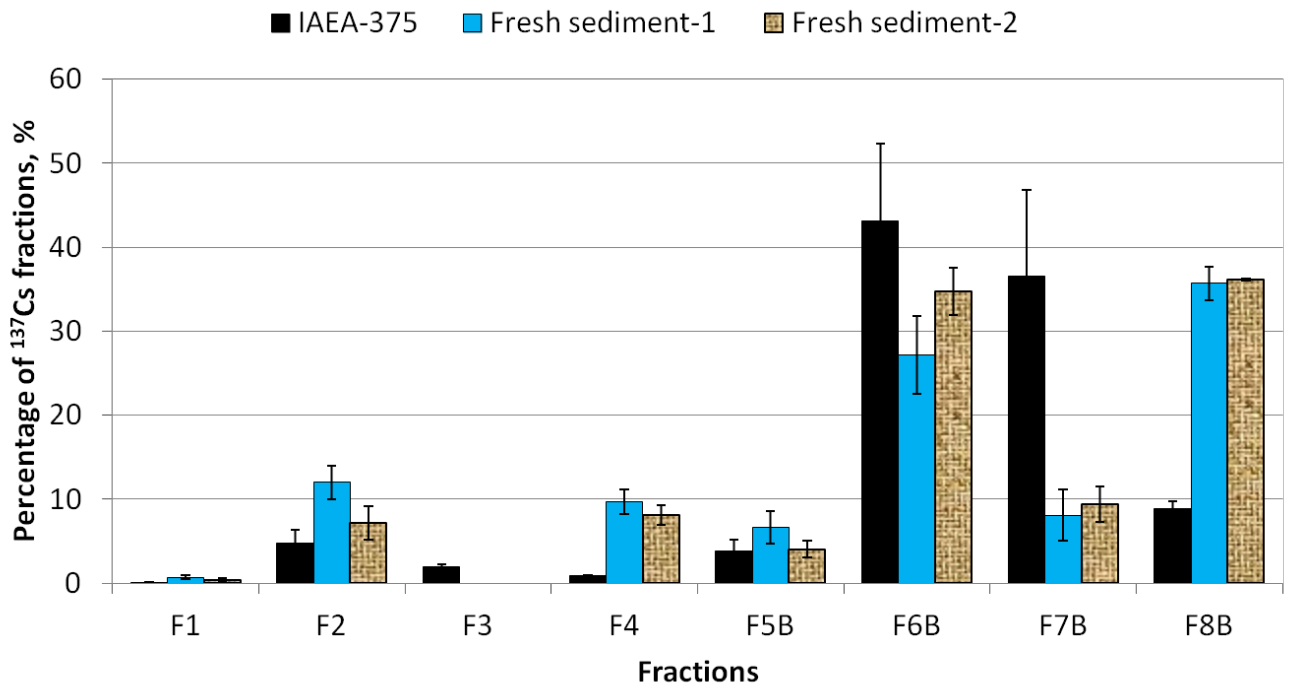
$^{238}\text{U}$ Fraction	Lake Heimdalen sediment-1	Lake Heimdalen sediment-2
	Concentration Bq/kg	Concentration Bq/kg
F1	$0.007 \pm 0.007$	$0.0006 \pm 0.0002$
F2+F3	$2.743 \pm 1.150$	$1.889 \pm 0.331$
F4	$7.144 \pm 0.708$	$6.154 \pm 0.931$
F5B	$0.822 \pm 0.379$	$1.265 \pm 0.366$
F6B	$2.858 \pm 0.504$	$2.426 \pm 0.345$
F7B	$0.532 \pm 0.047$	$0.747 \pm 0.140$
Sum(1-7)	$14.106 \pm 2.795$	$12.481 \pm 2.113$
Total (aqua regia)	-	-

Table 6. Fractionation of  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  in different samples

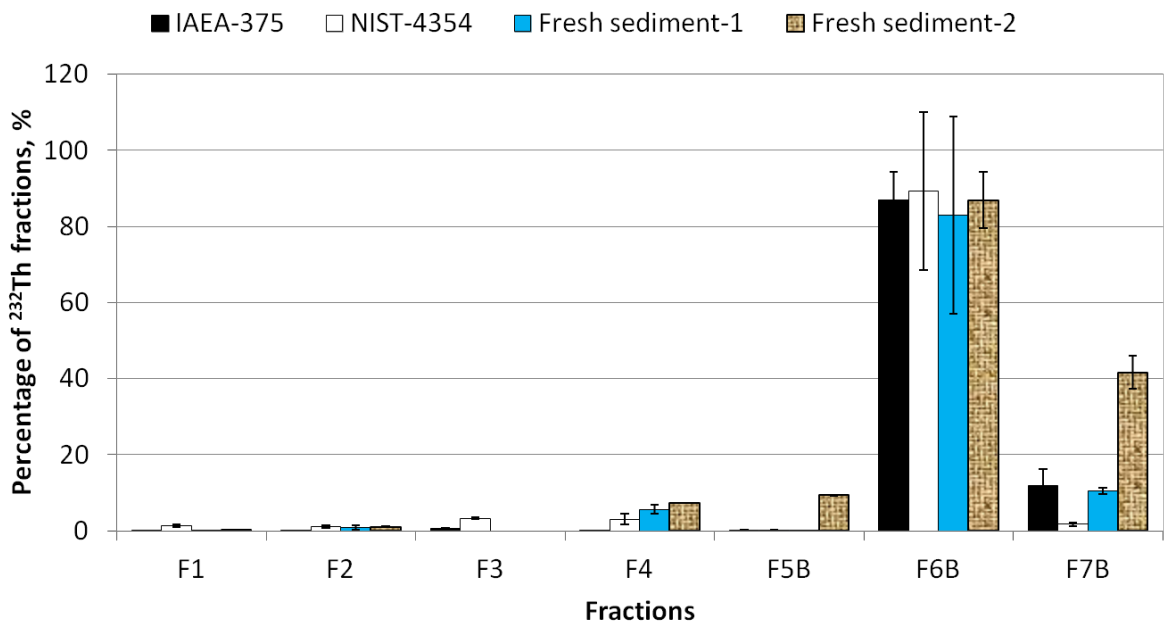
Sample	IAEA-SRM-375 soil			
Fraction	$^{239}\text{Pu}$ Concentration, Bq/kg	$^{240}\text{Pu}$ Concentration, Bq/kg	$^{239+240}\text{Pu}$ Concentration, Bq/kg	$^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio
F1	0.006 ± 0.004	0.011 ± 0.005	0.017 ± 0.006	0.51 ± 0.39
F2	0.016 ± 0.004	0.027 ± 0.003	0.043 ± 0.005	0.45 ± 0.12
F3	0.006 ± 0.003	0.012 ± 0.005	0.018 ± 0.006	0.54 ± 0.36
F4	0.005 ± 0.002	0.012 ± 0.004	0.017 ± 0.005	0.72 ± 0.28
F5B	0.062 ± 0.014	0.064 ± 0.010	0.125 ± 0.017	0.28 ± 0.08
F6B	0.025 ± 0.019	0.033 ± 0.006	0.058 ± 0.020	0.36 ± 0.28
F7B	0.005 ± 0.001	0.005 ± 0.004	0.010 ± 0.004	0.31 ± 0.23
Sum(1-7)	0.125 ± 0.047	0.165 ± 0.037	0.290 ± 0.063	0.36 ± 0.16
Total (aqua regia)	0.123 ± 0.012	0.082 ± 0.008	0.205 ± 0.021	0.18 ± 0.03
Recommended value	-	-	0.30 ± 0.04	-
Sample	NIST-SRM-4354 lake sediment			
Fraction	$^{239}\text{Pu}$ Concentration, Bq/kg	$^{240}\text{Pu}$ Concentration, Bq/kg	$^{239+240}\text{Pu}$ Concentration, Bq/kg	$^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio
F1	0.056 ± 0.005	0.032 ± 0.013	0.088 ± 0.013	0.16 ± 0.06
F2	0.068 ± 0.015	0.051 ± 0.023	0.119 ± 0.028	0.21 ± 0.10
F3	0.071 ± 0.025	0.051 ± 0.004	0.123 ± 0.025	0.20 ± 0.07
F4	0.037 ± 0.010	0.038 ± 0.007	0.075 ± 0.012	0.28 ± 0.09
F5B	0.274 ± 0.032	0.176 ± 0.087	0.450 ± 0.093	0.18 ± 0.09
F6B	0.813 ± 0.157	0.698 ± 0.123	1.511 ± 0.200	0.23 ± 0.06
F7	0.040 ± 0.015	0.031 ± 0.003	0.071 ± 0.015	0.21 ± 0.08
Sum(1-7)	1.359 ± 0.258	1.077 ± 0.260	2.436 ± 0.386	0.22 ± 0.07
Total (aqua regia)	2.406 ± 0.241	1.197 ± 0.120	3.603 ± 0.360	0.14 ± 0.02
Recommended value	-	-	4.0 ± 0.3	-
Sample	Lake Heimdalen sediment-1			
Fraction	$^{239}\text{Pu}$ Concentration, Bq/kg	$^{240}\text{Pu}$ Concentration, Bq/kg	$^{239+240}\text{Pu}$ Concentration, Bq/kg	$^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio
F1	0.053 ± 0.011	0.071 ± 0.008	0.124 ± 0.013	0.36 ± 0.08
F2+F3	0.058 ± 0.009	0.066 ± 0.010	0.125 ± 0.013	0.31 ± 0.07
F4	0.105 ± 0.034	0.110 ± 0.010	0.216 ± 0.035	0.29 ± 0.10
F5B	0.345 ± 0.046	0.307 ± 0.077	0.651 ± 0.090	0.24 ± 0.07
F6B	0.164 ± 0.030	0.184 ± 0.064	0.347 ± 0.071	0.31 ± 0.12
F7B	0.028 ± 0.013	0.042 ± 0.010	0.070 ± 0.016	0.41 ± 0.22
Sum(1-7)	0.753 ± 0.143	0.780 ± 0.178	1.533 ± 0.239	0.28 ± 0.08
Total (aqua regia)	0.896 ± 0.090	0.565 ± 0.057	1.461 ± 0.146	0.17 ± 0.02
Sample	Lake Heimdalen sediment-2			
Fraction	$^{239}\text{Pu}$ Concentration, Bq/kg	$^{240}\text{Pu}$ Concentration, Bq/kg	$^{239+240}\text{Pu}$ Concentration, Bq/kg	$^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio
F1	0.126 ± 0.026	0.118 ± 0.030	0.244 ± 0.040	0.26 ± 0.08
F2+F3	0.026 ± 0.011	0.046 ± 0.018	0.072 ± 0.021	0.48 ± 0.27
F4	0.061 ± 0.007	0.056 ± 0.009	0.117 ± 0.011	0.25 ± 0.05
F5B	0.193 ± 0.019	0.155 ± 0.036	0.348 ± 0.041	0.22 ± 0.06
F6B	0.408 ± 0.255	0.395 ± 0.124	0.803 ± 0.283	0.26 ± 0.19
F7B	0.014 ± 0.003	0.018 ± 0.003	0.032 ± 0.005	0.36 ± 0.11
Sum(1-7)	0.828 ± 0.321	0.788 ± 0.220	1.616 ± 0.401	0.26 ± 0.12
Total (aqua regia)	1.154 ± 0.115	0.550 ± 0.055	1.703 ± 0.170	0.13 ± 0.02



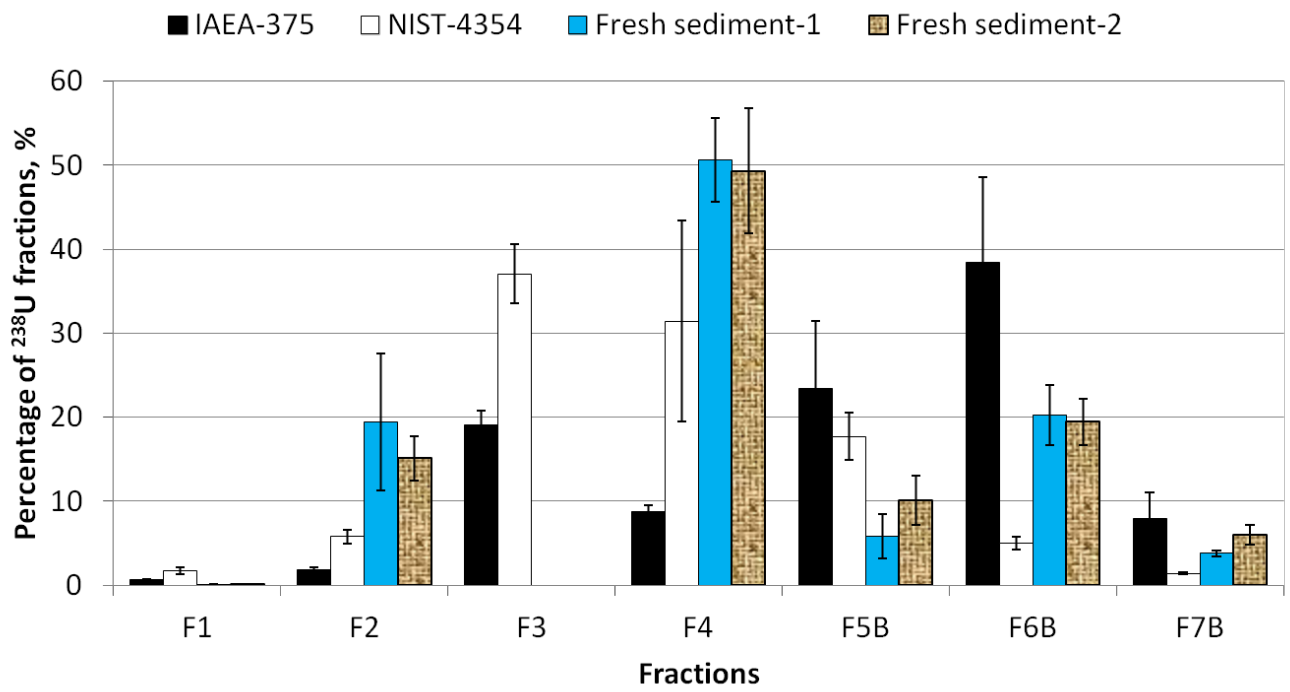
**Fig. 1** Distribution of  $^{127}\text{I}$  and  $^{129}\text{I}$  in the fractions from sequential extraction of IAEA-375 soil, NIST-4354 sediment and two sediments from Lake Heimdalne (for the two Lake Heimdalne sediments, F2 is represented F2+F3)



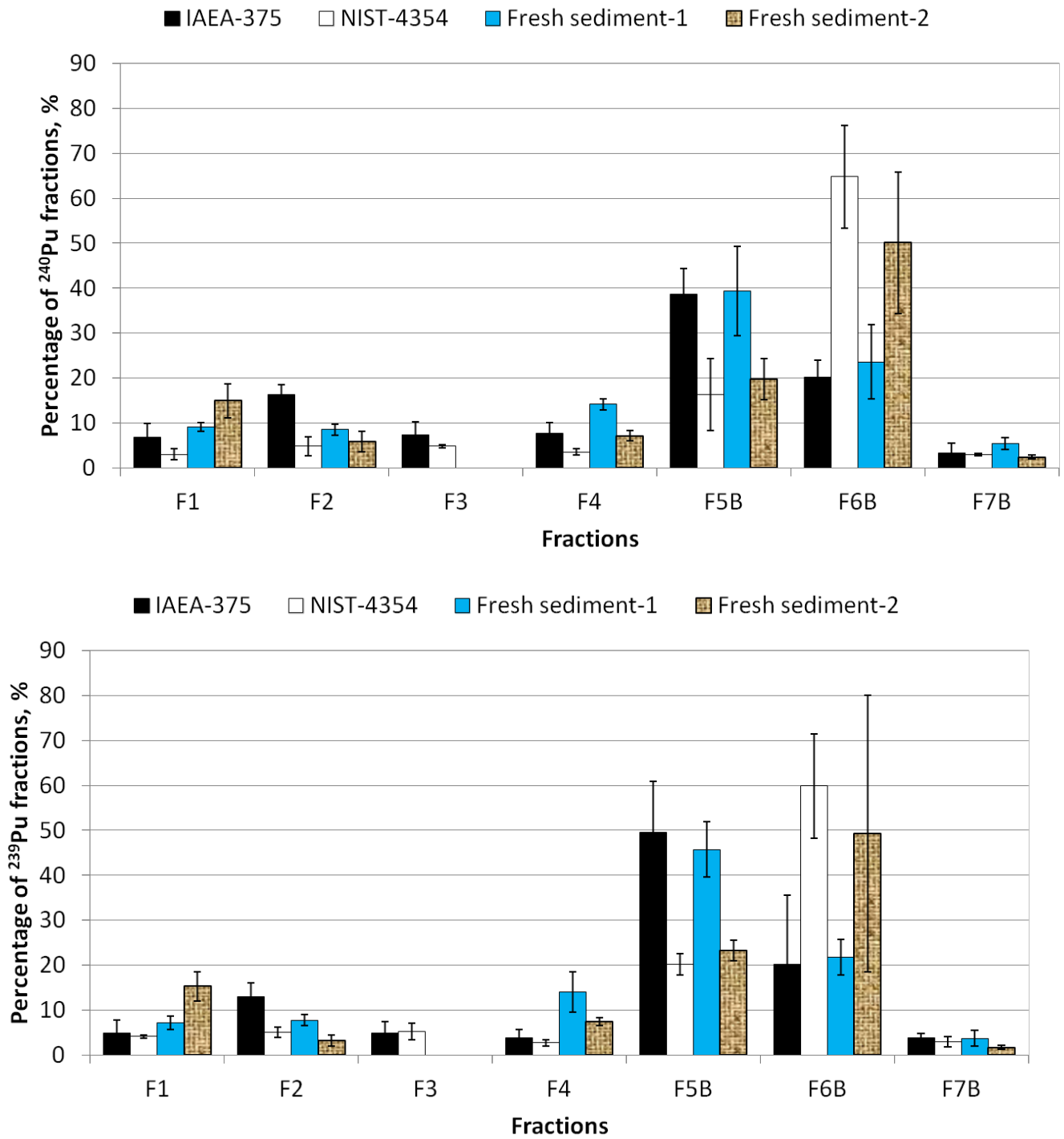
**Fig. 2** Distribution of <sup>137</sup>Cs in the fractions from sequential extraction of IAEA-375 soil and two sediments from Lake Heimdalne (for the two Lake Heimdalne sediments, F2 is represented F2+F3)



**Fig. 3** Distribution of <sup>232</sup>Th in the fractions from sequential extraction of IAEA-375 soil, NIST-4354 sediment and two sediments from Lake Heimdalne (for the two Lake Heimdalne sediments, F2 is represented F2+F3)



**Fig. 4** Distribution of  $^{238}\text{U}$  in the fractions from sequential extraction of IAEA-375 soil, NIST-4354 sediment and two sediments from Lake Heimdalne (for the two Lake Heimdalne sediments, F2 is represented F2+F3)



**Fig. 5** Distribution of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  in the fractions from sequential extraction of IAEA-375 soil, NIST-4354 sediment and two sediments from Lake Heimdalne (for the two Lake Heimdalne sediments, F2 is represented F2+F3)