Science of the Total Environment xxx (2009) xxx–xxx

Contents lists available at ScienceDirect

Science of the Total Environment

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

First results on ^{236}U levels in global fallout

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ARTICLE INFO

Article history:
Received 12 November 2008
Received in revised form 19 January 2009
Accepted 22 January 2009
Available online xxxx

Keywords:
Uranium-236
Global fallout
Accelerator mass spectrometry

ABSTRACT

The global fallout ^{236}U level in soil was deduced from measurements of ^{236}U, $^{239+240}$Pu and ^{137}Cs in surface soils which are solely influenced by global fallout. A total of 12 soil cores from the depths of 0–10, 0–20 and 0–30 cm were collected at a flat forest area in Japan. Concentrations of $^{239+240}$Pu and ^{238}U were determined by α-particle spectrometry, while the ^{236}U/^{238}U ratio was measured with accelerator mass spectrometry (AMS). Consistent ^{236}U/^{239}Pu ratios between 0.212 and 0.253 were found. Using this ratio, the total global fallout of ^{236}U on the earth is estimated to be as much as ca. 900 kg. This knowledge will contribute to the promotion of research on U isotopes, including ^{236}U, for the fields of geo-resources, waste management and geochemistry.

1. Introduction

Recently, the uranium isotope ^{236}U ($T_{1/2} = 2.342 \times 10^7$ y, α-decay) has been recognized as one of the “environmental Uranium(U) isotopes” due to the development of instruments and measuring techniques of sufficient abundance sensitivity, and applications which use ^{236}U as a proxy/tool for environmental and geochemical uranium studies are developing.

The occurrence of ^{236}U on the surface of the Earth's crust is mainly due to four origins. The first are the natural nuclear reactions of ^{235}U(n,γ) and ^{238}U(n,3n). The reaction ^{235}U(n,γ) is mainly induced by (α,n) reactions on light elements such as Na and Mg, where the α-particles are mainly emitted in the U and Th decay series. The ^{236}U/^{238}U ratios found in U ores are in the range of 10^{-12}–10^{-10} (Rokop et al., 1972; Zhao et al., 1994; Richter et al., 1999; Berkovits et al., 2000, Steier et al., 2002, 2008, Wilcken et al., 2007, 2008). Close to the surface layer, thermal neutrons derived from cosmic rays contribute to ^{236}U production:

$$dN_{236}(t)/dt = \sigma_{235n\gamma} \cdot \phi \cdot N_{235} - \lambda_{236} \cdot N_{236}$$

where N_{236} and λ_{236} are the number of ^{236}U atom per unit volume (cm^3) and decay constant of ^{236}U (9.38×10^{-16} 1/s), $\sigma_{235n\gamma}$ is thermal cross section for ^{235}U(n,γ) (86.7 barn), and ϕ is the environmental thermal neutron flux (at the latitude of the Japanese archipelago the surface flux is 0.001 n/cm^2/s, cf. around 35°N, 135°E, Endo et al., 2007). In the steady state (condition of saturation) the ^{236}U/^{238}U atom ratio can be estimated to be 7×10^{-13} for typical rocks. A minor contribution of 3×10^{-13} will arise from the reaction ^{238}U(n,3n) in which the fast cosmic neutrons are estimated from neutron flux data from (Masarik and Beer, 1999). However, this has to be considered as an upper limit, since almost no surfaces with such long exposure age exist (Schaefer et al., 2006). The isotopic ratio will generally be closer to the typical levels of 3×10^{-14} expected for deeper layers of rock (Steier et al., 2008). By assuming that the ^{238}U concentration of soil is 3 ppm (crustal average) and the soil density is 2.5 g/cm^3, the maximum inventory of natural ^{236}U in 10 cm surface soil would be calculated as 1.8×10^9 atoms/m^2. The second possible source is anthropogenic ^{236}U which was found e.g. around nuclear reprocessing facilities, the vicinity of Chernobyl and the depleted uranium (DU) contaminated areas in Kosovo (Hotchkis et al., 2000; Marsden et al., 2001; Mironov et al., 2002; Danesi et al., 2003; Ketterer et al., 2003; Boulyga and Heumann, 2006) at relatively high ^{236}U/^{238}U atom ratios from 10^{-7}–10^{-3}. The third is the growth from Plutonium-240(^{240}Pu) deposited already on land by global fallout. Plutonium isotopes have been well studied as one of the representation of global fallout nuclides, and much data has been reported for their concentrations, inventories and ^{240}Pu/^{239}Pu ratios. The ^{236}U growth from ^{240}Pu, 40 years after deposition, can be roughly estimated as 1.0×10^9–5.0×10^{11} atoms/m^2 (^{236}U/^{238}U ~ 2.6×10^{-12}–5.3×10^{-11}) by using the half life of ^{240}Pu (6563 years) and global fallout $^{239+240}$Pu values, 10–200 Bq/m^2 (4–80 Bq/m^2 for ^{240}Pu), found in Japan by Yamamoto et al. (1983).

The fourth is the global fallout of ^{236}U. Uranium-236 is produced by nuclear or thermonuclear atomic bombs. In global fallout, the

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doi:10.1016/j.scitotenv.2009.01.058

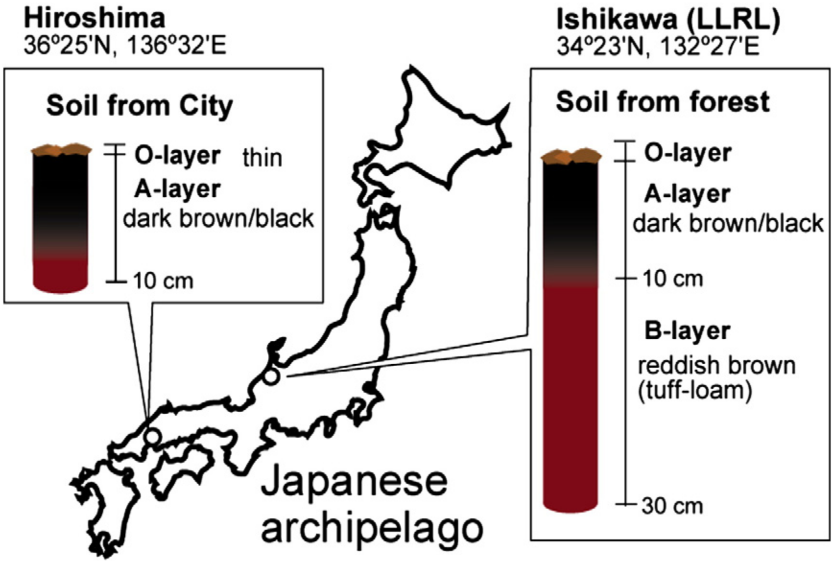

Fig. 1. Map showing surface core sampling locations (Ishikawa and Hiroshima Prefectures) in Japan, and descriptions of soil cores. The details are presented in the text.

most significant contribution will originate from ^{238}U(n,3n)^{236}U. Uranium (probably with natural isotopic composition in most cases) was used as a material for thermonuclear devices, in which the primary fusion process ^{3}H(d,n)^{4}He produces fast neutrons of 14 MeV. The cross section for ^{238}U(n,3n)^{236}U at this energy is approximately 0.4 barn (Shibata et al., 2002). From the reported ratio of ^{237} Np/^{239}Pu in global fallout, 0.44–0.59 (Beasley et al., 1998, Yamamoto et al., 1991, 1994), and the cross section of the reaction ^{238}U(n,2n) ^{237}U (^{237}U decays to ^{237}Np) of about 0.8 barn (Shibata et al., 2002), we estimate a ^{236}U/^{239}Pu production ratio of ca. 0.22–0.30. In the process of being distributed worldwide together with Pu and Cs, fractionation between U and Pu might occur, since U is generally more mobile in soils.

For the measurement of ^{236}U, ICP-MS (inductively coupled plasma mass spectrometry), TIMS (thermal ionization mass spectrometry) and AMS (accelerator mass spectrometry) have usually been employed. AMS is presently the method with the highest abundance sensitivity for ^{236}U. ICP-MS, the most commonly used mass spectrometric method for the investigation of actinides in the environment, has a detection limit above ^{236}U/^{238}U$=10^{-8}$ (Buchholz et al., 2007). TIMS can reach detection limits of 2×10^{-10} (Richter et al., 1999), but it mainly finds less favor than ICP-MS because of the more difficult sample preparation and measurement technique. For both techniques the limitation is imposed by the isobaric molecule ^{235}UH^{+} and by tails from the ^{238}U and ^{235}U peaks. Due to the natural occurrence of uranium in most minerals, the expected environmental ^{236}U/^{238}U levels resulting from global fallout are close to these limits. Precise measurements would have to be restricted to areas with exceptionally high global fallout and low natural uranium (Ketterer et al., 2007). Sample preparation would need to be optimized to minimize the extraction of natural ^{238}U from the mineral matrix, which therefore would require a compromise for the ^{236}U extraction efficiency. Compared to these techniques, AMS can measure ^{236}U/^{238}U ratios down to 10^{-14} (Wilcken et al., 2008). Therefore, the amount of natural uranium introduced into the spectrometer is of no concern, and simple and efficient chemistry will yield reliable results. AMS is also very robust against other matrix elements left in the purified sample.

In this paper, we report the ^{236}U global fallout level in surface soil which is not contaminated from any reprocessing facility, nuclear power plant, nor containing DU, etc. Emphasis was placed on (1) efficient leaching methods of ^{236}U from soil, and (2) comparison with ^{137}Cs and Pu to evaluate ^{236}U/^{137}Cs and ^{236}U/$^{239+240}$Pu ratios in surface soil which allows (3) to estimate the levels of ^{236}U in global fallout.

2. Materials and methods

2.1. Sampling

The sampling site of the present work, a forest near the vicinity of the LLRL (Low Level Radioactivity Laboratory, Ishikawa prefecture, Japan; Fig. 1) has been observed for a long time by our group. Actually, the sampling site chosen for the present work was selected among 30 sites in Japan based on results on Cs, Am and Pu presented by Yamamoto et al. (1983), as representative for global fallout. For the present work, the activity ratio $^{239+240}$Pu/^{137}Cs at the site was redetermined, yielding results in agreement with reported global fallout values (e.g. UNSCEAR, 2000; Ohtsuka et al., 2004).

The three sampling sites labeled T1, T2 and T3 lie 20 m apart from each other. The area appears to be undisturbed by human and/or animal activities. Site T2 is situated in a hollow at an altitude 1 m lower than sampling point T1, whereas T3 lies 2 m higher. A schematic representation of the soil profile is shown in Fig. 1. The surface was covered with decaying organic materials, fallen leaves and dead grasses (O-layer). We could recognize a dark brown-black mixed layer (A-layer) at the depth of 0–ca.10 cm. Below that the soil was reddish-brown (B-layer), and likely originates from weathered mother rock (tuff). It has been reported that global fallout Pu and Cs are trapped within the A-layer even in rainy Japan in the absence of severe mixture of surface soil (Takenaka et al., 1998; Koga, 2008; Fukuyama et al., 2005). As a first attempt to assess the ^{236}U/^{137}Cs and ^{236}U/$^{239+240}$Pu ratio in global fallout, up to 5 cores of 0–10 cm, 0–20 cm, and 0–30 cm depth (with diameter 4.7 cm) were collected within an area of ca. 4 m^{2} at each of the sites T1, T2, and T3.

In addition to these samples, two soil samples up to a depth of 10 cm (10 cm in diameter) collected from Hiroshima (Fig. 1) in 1976 in the framework of a different project were used for establishing appropriate leaching methods of anthropogenic uranium contamination from soil.

These samples were air-dried, and sieved through a 2-mm mesh screen to remove pebbles and plant remains, and pulverized in an agate mortar to obtain homogeneous samples. The samples used for analysis were calcinated at 450 °C overnight.

Please cite this article as: Sakaguchi A, et al, First results on ^{236}U levels in global fallout, Sci Total Environ (2009), doi:10.1016/j.scitotenv.2009.01.058

Table 1
Results of U isotope leaching rate from Hiroshima surface soil samples.

Sample	Anal. W (g)	^{238}U leached (mBq/g-soil)	^{238}U leached (%)	Atom ratio* $^{236}U/^{238}U$	^{236}U content (atoms/g-soil)
8 km NWN from hypocenter					
0.1 M HNO_3 leaching	5.00	4.62 ± 0.31	12	$(5.82 \pm 1.01) \times 10^{-8}$	5.47×10^{8}
8 M HNO_3 leaching	5.00	20.5 ± 0.98	54	$(1.48 \pm 0.22) \times 10^{-8}$	6.16×10^{8}
Decomposition	5.00	38.0 ± 1.31	100	$(7.19 \pm 3.17) \times 10^{-9}$	5.56×10^{8}
22 km NWN from hypocenter					
0.1 M HNO_3 leaching	5.00	2.00 ± 0.13	6	$(1.86 \pm 0.24) \times 10^{-7}$	7.55×10^{7}
8 M HNO_3 leaching	5.00	14.1 ± 0.87	42	$(7.58 \pm 0.52) \times 10^{-8}$	2.17×10^{8}
Decomposition	5.00	33.3 ± 1.28	100	$(3.29 \pm 0.60) \times 10^{-8}$	2.23×10^{8}
BG		n.d.		n.d.	
Vienna-KkU standard**				$(1.94 \pm 0.02) \times 10^{-11}$	

n.d.: not detected.
*Corrected value with Vienna-KkU standard.
**VERA inhouse standard with an $^{236}U/^{238}U$ ratio of 6.98×10^{-11}.

2.2. Preference of leaching/decomposition methods for ^{236}U measurement

Using the two Hiroshima soil samples, three kinds of leaching/digestion methods were attempted to check the optimal condition for the measurement of anthropogenic ^{236}U, i.e., complete recovery of ^{236}U with reasonable effort. Natural U leached from the soil imposes no problem, due to the high abundance sensitivity of AMS.

The first method is 0.1 M HNO_3 (10 ml soln./1 g soil sample) leaching for 3 h with continual agitation. The second one is 8 M HNO_3 (10 ml soln./1 g soil sample) leaching for 3 h on a hot plate (150 °C) with continual agitation. The third method is decomposition with $HF+HNO_3+HClO_4$. Each leaching/decomposition solution obtained (about 35 g in 50 g total soln.) was weighed and divided into two parts; one was used for ^{238}U concentration measurements with α-particle spectrometry and the other for $^{236}U/^{238}U$ atom ratio measurement with AMS. For the measurement of U contents, after adding a known amount of ^{232}U to the solution, the uranium was radiochemically separated and purified using an anion-exchange resin column method (Sakaguchi et al., 2004). The purified U was electroplated onto polished stainless steel discs, and the activities were determined by α-particle spectrometry conducted in LLRL, Kanazawa University.

For the AMS measurements of $^{236}U/^{238}U$ at the VERA facility at the University of Vienna, U in the solution was separated and purified in the same manner as above, but without the addition of ^{232}U tracer, since we could not rule out a ^{236}U content in the spike. The sputter targets for the AMS measurement were prepared with an iron oxide matrix by calcination at 800 °C for 2 h after co-precipitation with $Fe(OH)_3$. These targets were prepared with an ^{238}U concentration of 2–3 μg/mg Fe oxide. The resultant $^{238}U^{7+}$ currents measured ranged from 20 to 100 pA, compared to typically 50 nA observed for pure U_3O_8 samples. However, these currents are well in the range of the current amplifiers connected to our Faraday cups (SR570, Stanford Research Systems, Inc, Sunnyvale, California, USA). The process blanks read currents of 1 pA or below. To achieve sufficient precision of the current measurement, the integration time for $^{238}U^{7+}$ is extended to up to 1 s. More information on the measurement of ^{236}U by AMS is given in (Vockenhuber et al., 2003). VERA reaches an abundance sensitivity of $^{236}U/^{238}U = 10^{-13}$.

2.3. Treatment of background soil samples

The cores from the Ishikawa site were used to evaluate the ratios of $^{236}U/^{239+240}Pu$ and $^{236}U/^{137}Cs$ from global fallout.

Each soil sample (55–60 g) was packed into a plastic vessel with a diameter of 6 cm and height of 2 cm. ^{137}Cs was determined by γ-ray spectrometry using a Ge-detector (Princeton Gamma Tech, relative efficiency 40%) with measurements of 92500 s duration for each sample. The spectrometers were calibrated with standards prepared from the New Brunswick Laboratory (NBL) reference materials No. 42-1 (4.04% uranium) and analytical grade KCl. The efficiency for ^{137}Cs (662 keV) was 3.00%.

Calcinated soil samples were leached for U and Pu isotopes with 8 M HNO_3 with small amounts of H_2O_2 by heating for at least 3–4 h on a hot plate. The total amount of solution leached was weighed and separated into three aliquots: (1) 1/6 of the total leaching soln. for $^{236}U/^{238}U$ atom ratio determination by AMS, (2) 1/6 to determine the leached amount of ^{238}U, and (3) 2/3 for the $^{239+240}Pu$ content determination. Uranium analysis was performed with α-particle spectrometry in the same manner as described for the Hiroshima samples above. Plutonium, with the addition of ^{242}Pu tracer, was purified and measured with α-particle spectrometry by using the method reported by Yamamoto et al. (1996).

3. Results and discussion

The comparison of the three leaching/decomposition methods investigated using the Hiroshima soils is shown in Table 1. Assuming that the decomposition treatment represents 100% content of ^{238}U, the U extraction yields with 0.1 M and 8 M HNO_3 leaching are about 10 and 50%, respectively. The atom ratios of $^{236}U/^{238}U$ varied in a wide range of $(0.72–5.82) \times 10^{-8}$ for the samples collected 8 km from the hypocenter and

Table 2
Results of U isotopes in surface soil samples from Ishikawa Prefecture.

Sample	Depth	Number of core	Total W dry (g)	Anal. W (g)	Atom ratio* $^{236}U/^{238}U$	^{238}U leached (Bq/Anal. g)	(mBq/g)	(atoms/g)	^{236}U (atoms/g)
Point-1 T1									
T1-1	0–10 cm	2	165.85	34.94	$(1.09 \pm 0.03) \times 10^{-7}$	0.428 ± 0.019	12.26	2.497×10^{16}	2.72×10^{9}
T1-2	0–20 cm	1	250.75	35.09	$(5.48 \pm 0.15) \times 10^{-8}$	0.528 ± 0.020	15.06	3.066×10^{16}	1.68×10^{9}
T1-3	0–30 cm	1	413.07	35.72	$(2.62 \pm 0.13) \times 10^{-8}$	0.689 ± 0.029	19.30	3.929×10^{16}	1.03×10^{9}
Point-2 T2									
T2-1	0–10 cm	2	294.95	35.70	$(1.09 \pm 0.03) \times 10^{-7}$	0.605 ± 0.025	16.94	3.449×10^{16}	3.76×10^{9}
T2-2	0–20 cm	1	282.30	35.70	$(2.14 \pm 0.09) \times 10^{-8}$	0.731 ± 0.023	20.48	4.170×10^{16}	8.92×10^{8}
Point-3 T3									
T3-1	0–10 cm	2	247.58	35.10	$(6.18 \pm 0.13) \times 10^{-8}$	0.837 ± 0.031	23.85	4.855×10^{16}	3.00×10^{9}
T3-2	0–20 cm	2	473.14	35.64	$(2.44 \pm 0.10) \times 10^{-8}$	1.122 ± 0.036	31.48	6.409×10^{16}	1.56×10^{9}
T3-3	0–30 cm	1	402.41	35.08	$(1.85 \pm 0.10) \times 10^{-8}$	1.080 ± 0.062	30.80	6.270×10^{16}	1.16×10^{9}

*A part of the leached solution was used for the AMS measurement.

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Table 3
Results of radionuclides in surface soil samples from Ishikawa Prefecture.

Sample	Depth	^{236}U	^{239,240}Pu	^{137}Cs	^{239,240}Pu/^{137}Cs	^{236}U/^{239,240}Pu	^{236}U/^{137}Cs
		(atoms/g)	(Bq/g)	(Bq/g)	Activity ratio	(atoms/Bq)	(atoms/Bq)
Point-1							
T1-1	0–10 cm	2.72×10^9	1.943 ± 0.052	71.78 ± 0.93	0.0271	1.40×10^{11}	3.79×10^9
T1-2	0–20 cm	1.68×10^9	1.111 ± 0.031	37.34 ± 0.54	0.0298	1.51×10^{11}	4.50×10^9
T1-3	0–30 cm	1.03×10^9	0.667 ± 0.020	16.55 ± 0.30	0.0403	1.54×10^{11}	6.22×10^9
Point-2							
T2-1	0–10 cm	3.76×10^9	2.244 ± 0.051	82.50 ± 0.73	0.0272	1.68×10^{11}	4.56×10^9
T2-2	0–20 cm	8.92×10^8	0.578 ± 0.023	20.85 ± 0.53	0.0277	1.54×10^{11}	4.28×10^9
Point-3							
T3-1	0–10 cm	3.00×10^9	2.021 ± 0.069	66.87 ± 0.79	0.0302	1.48×10^{11}	4.49×10^9
T3-2	0–20 cm	1.56×10^9	0.925 ± 0.030	26.24 ± 0.56	0.0353	1.69×10^{11}	5.96×10^9
T3-3	0–30 cm	1.16×10^9	0.730 ± 0.024	22.01 ± 0.50	0.0332	1.59×10^{11}	5.27×10^9
Mean ± σ					0.0313 ± 0.005	$(1.56\pm0.10)\times10^{11}$	$(4.88\pm0.85)\times10^9$

$(3.29–18.6)\times10^{-8}$ for the samples collected 22 km from the hypocenter. However, in this case, the more meaningful number is probably the ^{236}U concentration (^{236}U atoms/g soil) because the ^{236}U/^{238}U atom ratios might be changed by varying leaching efficiencies of natural ^{238}U from the soil matrix. The concentrations of ^{236}U were determined to be $(5.47–6.16)\times10^8$ atoms/g for the 8 km samples and $(0.76–2.23)\times10^8$ atoms/g for the 22 km samples, respectively. For the 22 km samples, the ^{236}U concentration estimated from the 0.1 M HNO_3 leaching is evidently lower than those for 8 M HNO_3 leaching and complete decomposition. The ^{236}U recovered with 8 M HNO_3 leaching and complete decomposition showed nearly the same values in both samples. As a result, leaching with 8 M HNO_3 is recommended as an appropriate approach for ^{236}U leaching from soil samples; full digestion is not required.

The Hiroshima surface soil is expected to contain also ^{236}U from the nuclear explosion. The estimation of this contribution, by considering also the isotopic composition of other nuclides such as $^{239+240}$Pu and ^{137}Cs, will be future work.

Table 2 shows the results for ^{236}U in soil samples from the Ishikawa Prefecture. ^{236}U/^{238}U atom ratios were detected in the range from 1.85×10^{-8} to 1.09×10^{-7}.

Our samples do not allow us to obtain a local inventory of ^{236}U for the Ishikawa site, since no depth profiles were determined. A strong local variability, well known for other global fallout nuclides, is also evident for ^{236}U: The 0–10 cm sample from site T2 contains more ^{236}U atoms than the 0–30 cm sample originating from the immediate neighborhood. However, the $^{239+240}$Pu/^{137}Cs activity ratio averages 0.0313 ± 0.0046, which is consistent with previous data as listed in Table 3 (e.g. Yamamoto et al., 1983; UNSCEAR, 2000; Ohtsuka et al., 2004). The average ratios of ^{236}U atoms to ^{137}Cs and $^{239+240}$Pu activity (atoms/Bq) in our Ishikawa samples were $(4.88\pm0.85)\times10^9$ and $(1.56\pm0.10)\times10^{11}$, respectively. By using the commonly accepted value of 0.18 as the ^{240}Pu/^{239}Pu ratio for global fallout the ^{236}U/^{239}Pu atom ratio in our samples can be estimated to be 0.235 ± 0.014. We think that this number is also representative for global fallout.

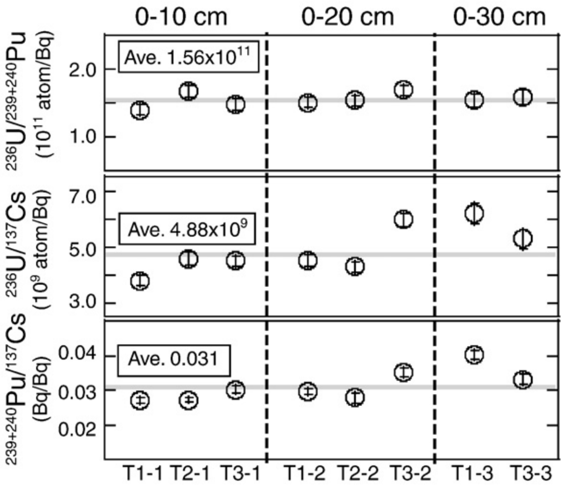

Fig. 2. Comparison of ratios among fallout nuclides, $^{239+240}$Pu/^{137}Cs, ^{236}U/^{137}Cs and ^{236}U/$^{239+240}$Pu in the soil cores form Ishikawa pref. Japan. Gray lines show the average of ratios.

Fractionation of ^{236}U and $^{239+240}$Pu in soil by different migration behavior could impair this conclusion. However, as can be seen in Fig. 2, identical values of ^{236}U/$^{239+240}$Pu are found for all samples. If ^{236}U and Pu migrated differently, it is unlikely that the same result would be obtained on all the samples, which differ in measured concentration, core length, and sampling location. This suggests that ^{236}U behaves in the nearly same manner as Pu at our site. Cs is known to migrate differently from Pu, and the correlation between ^{137}Cs and Pu is much worse in our data. The same holds for ^{236}U/^{137}Cs. The total deposition of ^{236}U as global fallout is roughly estimated to be as much as ca. 900 kg by using the global deposition of $^{239+240}$Pu on earth (14.8 PBq, Harley, 1980).

4. Conclusion

The global fallout situation of ^{236}U, together with $^{239+240}$Pu and ^{137}Cs, was investigated using soil core samples up to a depth of 30 cm in Ishikawa, Japan to serve as an aid as representative background data of areas which were not influenced by local or regional fallout from nuclear facilities, DU contamination, or the Chernobyl accident. ^{236}U was easily measured with AMS for environmental samples, and the isotopic ratios of ^{236}U/^{238}U and ^{236}U concentrations ranged from 1.85×10^{-8} to 1.09×10^{-7} and 8.92×10^8 to 3.76×10^9 atoms/g dry soil, respectively. The average atom ratios (atoms/Bq) of ^{236}U/$^{239+240}$Pu in our samples were $(1.56\pm0.10)\times10^{11}$, respectively, which translates into a ^{236}U/^{239}Pu atom ratio of 0.212 to 0.253, which corresponds with a rough estimate based on the production process in nuclear weapons, and with values reported by (Ketterer et al., 2007) (0.05–0.5). The total deposition of ^{236}U as global fallout on the earth was estimated as ca. 900 kg. Between ^{236}U and ^{137}Cs less correlation was found, which we attribute to different migration behavior in the soil. As already mentioned in other publications (Lee et al., 2008; Steier et al., 2008), ^{236}U might be a potential tracer not only for uranium contamination from nuclear fuel or waste, but also for geochemical dynamics. In the case of measurements of low level ^{236}U in the environment especially for surface soil samples, the effect of global fallout has to be considered. In this sense, the value estimated here will be very helpful in future applications.

Please cite this article as: Sakaguchi A, et al, First results on ^{236}U levels in global fallout, Sci Total Environ (2009), doi:10.1016/j.scitotenv.2009.01.058

Acknowledgement

We are grateful to ME. Ketterer, Northern Arizona University for scientific discussion, and T. Imanaka, University of Kyoto, for helpful comments and suggestions.

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Please cite this article as: Sakaguchi A, et al, First results on ^{236}U levels in global fallout, Sci Total Environ (2009), doi:10.1016/j.scitotenv.2009.01.058