Uranium-236 as a new oceanic tracer: A first depth profile in the Japan Sea and comparison with caesium-137

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ABSTRACT

We present a feasibility study for using ^{236}U as an oceanic circulation tracer based on depth profiles of ^{236}U and ^{137}Cs in the Japan/East Sea. The concentration of the predominantly anthropogenic ^{236}U, measured with Accelerator Mass Spectrometry (AMS), decreased from $(13 \pm 3) \times 10^6$ atom/kg in surface water to $(1.6 \pm 0.3) \times 10^6$ atom/kg close to the sea floor (2800 m). The profile has a smooth trend with depth and concentration values are generally proportional to that of ^{137}Cs for the same water samples, but with a slightly lower ratio of $^{137}Cs/^{236}U$ below 2000 m. The cumulative inventory of dissolved ^{236}U in the water column was estimated to be $(13.7 \pm 0.9) \times 10^{12}$ atom/m², which is similar to the global-fallout level $(17.8 \times 10^{12}$ atom/m²) in Japan. Additional analyses of suspended solids (SS) and bottom sediments yielded negligible amounts of ^{236}U. Our results suggest that ^{236}U behaves as a conservative nuclide in seawater, with potential advantages over other tracers of oceanic circulation.

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Introduction

The Japan Sea in the western Pacific Ocean is separated by four shallow channels (50–100 m in depth). The average and maximum depths are ca. 1700 m and 3800 m, respectively (Chronological Scientific Table, 2009). This deep bowl-like sea has one large input from the surrounding ocean through the Tsushima strait, forming the Tsushima Warm Current which dominates the surface layer. Without inflow from the open ocean, formation of deepwater occurs locally in the north west part of the Japan Sea and is similar to the processes in the world oceans (e.g. Uda, 1934; Nitani, 1972; Gamo and Horibe, 1983; Sudo, 1986). The product is a stable deep water body, the Japan Sea Proper Water, which has an extremely narrow range in water temperature (0.0–0.1 °C) and low salinity (34.0–34.1 psu), and is highly oxygenated (dissolved oxygen: 0.2–0.25 mM) (Asaoka 1987). Therefore, past and ongoing efforts by oceanographers have concentrated on the role of the Japan Sea as a "Miniature Ocean" (e.g. Uda, 1934). Extensive oceanographic observations have been carried out, employing advanced techniques to measure currents and water properties, but also three-dimensional numerical models with real topography and time-varying meteorological forcing, to improve our understanding of sophisticated phenomena occurring in the Japan Sea (Kim et al., 2008).

During the last several decades, a series of events such as a significant increase of water temperature, a weakening of water circulation and a decrease of dissolved oxygen in the water column have been reported for the Japan Sea (e.g. Gamo et al., 1986; Kim and Kim, 1996; Minami et al., 1999). Furthermore, Min and Warner (2005) suggested that since a near-complete cessation of deep water circulation during 1950–1975 the ventilation of deep water has been weak, but recently has increased. These observations have been interpreted as a result of global warming, and as a predictor of a possible outcome of global ocean ventilation. The semi-closed system in the Japan Sea has also raised concerns about the accumulation of contamination from sewage, industrial effluents and dumping of waste. To serve as an aid to understanding these problems, more detailed studies on the circulation of water and migration of materials are needed.

Cs-137 ($T_{1/2} = 30.2$ yr) has been spread worldwide as a fission product of atmospheric nuclear weapons testing in the 1960s. This nuclide has proved to be a powerful tool for oceanography due to its well-defined origin and conservative behaviour in water (e.g. Aoyama and Hirose, 1995; Miyao et al., 1998; Ito et al., 2003).

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However, the number of atoms has now decayed to about one third compared with initial levels, and will become even more difficult to measure in the future. Given this situation, we have focused on global fallout ^{236}U ($T_{1/2}=2.342 \times 10^7$ yr) as a candidate for a new isotopic tracer for oceanography. Reliable measurement of ^{236}U in the environment has become possible only recently, as a result of progress in high sensitivity measurement with small amount of sample based on accelerator mass spectrometry (AMS) (e.g. Steier et al., 2008; Srncik et al., 2010). ^{236}U is mainly induced by the nuclear reaction $^{235}U(n,\gamma)$ with thermal neutrons in reactors which use ^{235}U fuel, and this anthropogenic U isotope has been used as a fingerprint of environmental contamination arising from use of U fuel, e.g. from nuclear reprocessing facilities, Chernobyl NPP and so on (e.g. Boulyga and Heumann, 2006; Lee et al., 2008; Srncik et al., 2010).

Sakaguchi et al. (2009, 2010) showed that global fallout from nuclear weapons testing contains ^{236}U, which is mainly produced via the nuclear reaction $^{238}U(n,3n)$ and about 900 kg of ^{236}U have been distributed in the surface environment. Thus, ^{236}U may have the potential to act as a tracer for environmental dynamics similar to that of ^{137}Cs.

In a first study to characterise and clarify the environmental behaviour of ^{236}U, comprehensive studies have been made to measure the concentration of the isotope in marine samples such as waters, suspended solids (SS) and bottom sediments.

Materials and methods

Samples

The Japan Sea samples were collected on cruise KH10-02 of the research vessel Hahuko Maru, July to August 2010. Bottom sediments (CR 14; 39°32.56′N, 136 °40.35′E) and seawater samples (CR 58; 40°25.66′N, 135°55.21′E) were collected around the Yamato Basin, as shown in Fig. 1. The water depths at CR 14 and CR 58 were 2653 m and 2803 m, respectively. Samples of about 20 l seawater were taken with Niskin bottles at 12 different depths. Water temperature, dissolved oxygen (DO), pH, salinity
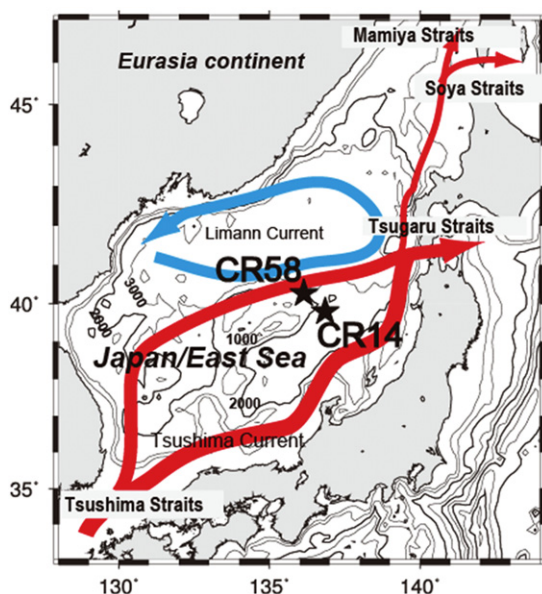

Fig. 1. Map of the sampling sites. CR58: Water and suspended solid (SS). CR14: Sea floor sediments. The major currents in the Japan Sea are Tsushima Warm Current through the Tsushima strait. The deep water might be conformed around the Limann Cold current in the severe winter.

and some other basic properties of seawater were measured precisely at 25 different depths with routine protocols established for the GEOTRACES project. Immediately after sample collection, the water was filtered with 0.45 μm pore-size membrane filters using a Teflon® filtration system. The filtrate was stored in polyethylene containers after adjustment to pH 1 by the addition of concentrated HNO_3. The SS collected on the filters was packed into a double plastic bag. Sediment core samples were taken by a multi-corer device with a diameter of 8.9 cm. The surface of the cores was not disturbed during sampling and handling of the sediment core samples. The cores were extruded in 1 cm segments, which were cut off and stored in a refrigerator at 4 °C within a few hours of sampling. Samples from the surface to 5 cm in depth were used for the present study. The procedure does lend itself to the possibility for cross-contamination from smearing between the layers. This is especially true for the topmost sample of the core (0–2 cm), which had a watery consistency.

Chemical treatments and measurements

The filtered seawater sample was put into a polyethylene vessel (approximately 20 l). The following chemical tracers and carriers were added: 30 μg of an in-house standard "Vienna-UPu1" used at the VERA Laboratory which is a mix of IRMM-058 and IRMM-085 with a gravimetric dilution to $(4.76 \pm 0.05) \times 10^{10}$ atom $^{233}U/g$, and $(2.69 \pm 0.03) \times 10^{11}$ atoms $^{242}Pu/g$, 200 mg of ^{133}Cs (Wako, analytical grade CsCl), and 200 mg of iron (Fe) as Fe/HCl solution. The seawater sample was heated for 3 h with stirring and then stood for 12 hs. After re-adjustment of the pH to 1 with HNO_3 and ammonium solution, 4 g of ammonium-phosphomolybdate (AMP) powder was added to adsorb Cs. After stirring for 1 h and standing for 24 h at room temperature, the supernatant was removed first with a siphon, followed by centrifugation (the supernatant was kept for U and Pu analyses). The Cs-adsorbed AMP was dried at 105 °C for 12 h, and packed into a plastic tube (1.4 cm × 4.0 cm) for the measurement of ^{137}Cs.

U and Pu in the supernatant were co-precipitated with $Fe(OH)_3$ at pH 8. After removing the supernatant by a siphon and centrifugation, the $Fe(OH)_3$ was dissolved in 10 M HCl. Further purification of U and Pu from this solution was conducted by using columns packed with anion-exchange resin as described by Sakaguchi et al. (2009, 2010).

The sea floor sediment samples were dried at 105 °C for 12 h. An aliquot (about 4 g) of sediment was totally digested with the methods described by Sakaguchi et al. (2004) after the addition of known amounts of ^{233}U, ^{242}Pu, and ^{133}Cs as yield tracers for each element. A part of the digested solution was precisely sub sampled (by weight) for measurement of ^{133}Cs by ICP-MS. After dilution with MQ water to 1 l and adjusting the remaining solution to pH 1 with HNO_3 and ammonium solution, 2 g of AMP was added to the solution. Analysis of Cs-adsorbed AMP and purification of U and Pu were conducted in a similar way as that for seawater.

The treatment for the SS samples was the same as used for the sediments, but measurement of ^{137}Cs was not attempted. Tanaka et al. (2006) previously had tried to measure 137Cs in SS, filtered from more than 60 l of seawater (Japan Sea) using a Ge-detector with an ultra-low background level installed at the Ogoya Underground Laboratory (OUL), LLRL, Kanazawa University, but was unsuccessful.

For analysis of the AMP samples from the seawater and sediment samples, the 662-keV line of ^{137}Cs was measured by γ-ray spectrometry using a well-type Ge detector (ORTEC, GWL-120230-S) installed at Hiroshima University. For the sediment samples a low-background well-type Ge detector (EURISYS, EGPC 150 P16) installed at LLRL was used, as the ^{137}Cs level was

expected to be very low. The detection limits for ^{137}Cs in both detectors were estimated as 0.007 Bq/d and 0.002 Bq/d, respectively. The uncertainty for each sample was about a few per cent expressed as 1σ. The spectrometer was calibrated with a mixed standard prepared by the Japan Radioisotope Association (no. MX-033).

A part of the AMP was dissolved with 1.25% tetramethylammonium hydroxide (TMAH) and the [133]Cs concentration was measured to assess the yield of the adsorbed [137]Cs. We calculated the recovery for Cs as 98–100%.

For the measurement of [236]U, [238]U, [239]Pu, and [240]Pu with AMS, the samples were prepared as U or Pu oxides in typically 3 mg of Fe_2O_3 matrix. A detailed description of the AMS measurement procedures for [236]U and [239]Pu/[240]Pu at VERA are described in Steier et al. (2010) and Sakaguchi et al. (2010). The solid sample is sputtered with a Cs beam, negative sample ions ($U^{16}O^-$ and $Pu^{16}O^-$) are extracted, and passed through the first mass spectrometer. The selected ions are accelerated to an energy of 3 MeV, the molecular ions are broken up and stripped to positive charge states in a gas cell, and the atomic breakup products undergo a second acceleration. The actinide ions of interest (U^{5+} or $^{239}Pu^{5+}$, at approximately 18 MeV) are separated in a second mass spectrometer. The high particle energy provided by the accelerator, the destruction of molecular isobars in the stripping process, and the repeated deployment of high-resolving electric and magnetic analysers explain the exceptional abundance sensitivity of AMS.

The spectrometers are programmed to rapidly switch between the different isotopes. The uranium isotopes were determined by counting $^{233}U^{5+}$ and $^{236}U^{5+}$ with a gas ionisation detector, and by measuring the $^{238}U^{5+}$ beam current in a Faraday cup. The resultant $^{238}U^{5+}$ current for samples, containing about 50 µg of uranium, was of the order of 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 (SR570, Stanford Research Systems, Inc., Sunnyvale, California, USA) connected to the Faraday cups. The process blanks gave currents of 1 pA or below. To achieve sufficient precision for the current measurement, the integration time for $^{238}U^{5+}$ was extended to up to 1 s. VERA achieves an abundance sensitivity for $^{236}U/^{238}U$ of 10^{-13}. The overall detection efficiency is 1 count in the detector per ~ 3000 actinide atoms in the sample, with the major losses taking place during sputtering (1% negative ion yield) and stripping (5% yield of 5+). This limits the sensitivity for plutonium, where no abundant isotope exists. Concentrations of [236]U and [238]U in

seawater and SS samples were determined by using the measured ratios of $^{236}U/^{233}U$ and $^{233}U/^{238}U$ and the known amount of [233]U tracer added. The results for Pu will be presented elsewhere. The recovery of U for chemical purification was 80–95%.

Results and discussion

Concentrations and inventories of [236]U and ^{137}Cs in the Japan Sea

The fundamental oceanographic parameters such as temperature (°C), salinity (psu), pH (SWS scale 25 °C) and dissolved oxygen (ml/l) in water from CR58 site are shown in Fig. 2. The θ–T diagram of this station is also presented in Fig. 3. From consideration of the depth profiles for these parameters and Fig. 3, it can be said that the compositional data are typical for Japan Sea samples: composition of the deeper water-mass at this
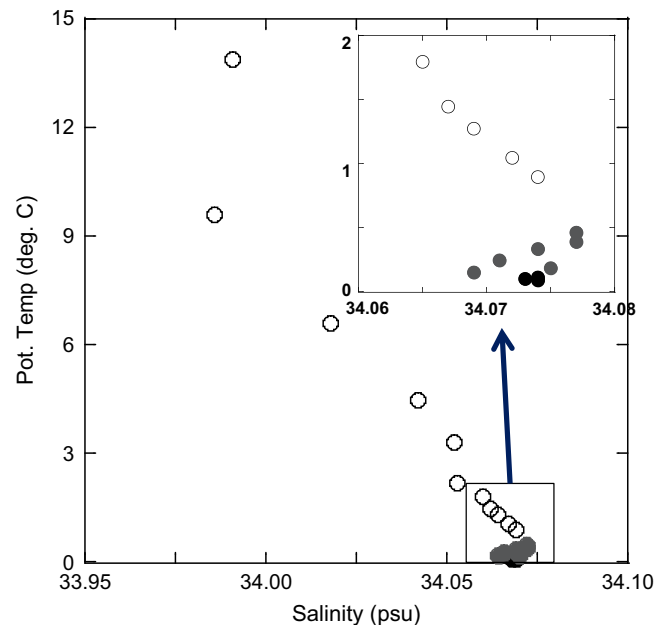

Fig. 3. θ–T (salinity vs. temperature) diagram of station CR58. Open circle: surface: 500 m, grey filled circle: 500–2000 m, and black filled circle: deeper than 2000 m.
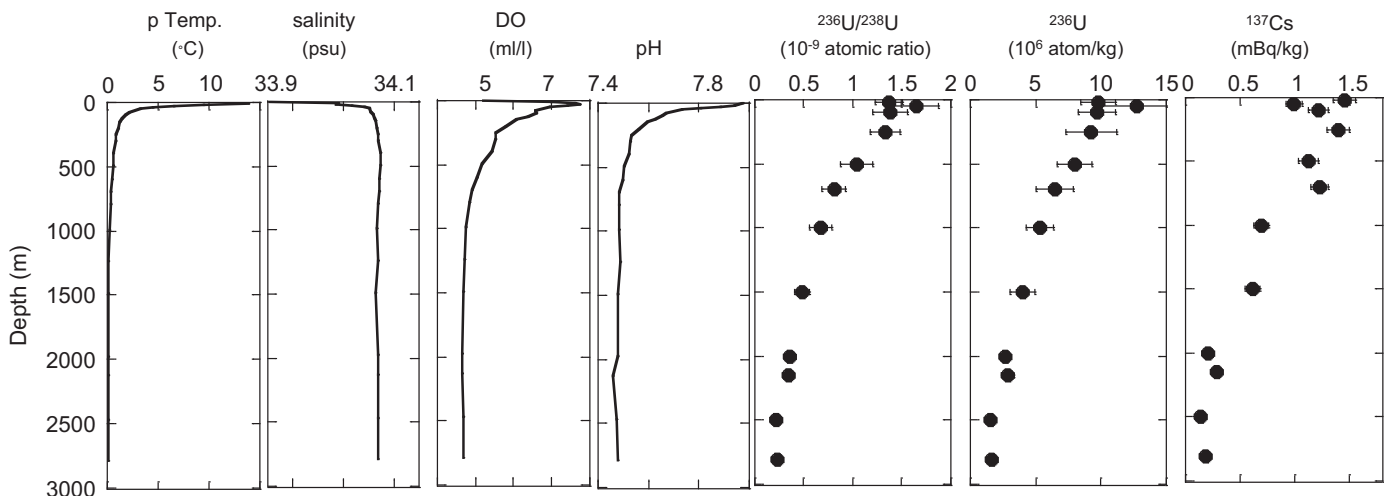

Fig. 2. Depth profiles of $^{236}U/^{238}U$ atom ratio and the concentration of [236]U (atom/kg) together with [137]Cs (mBq/kg) in the water column at the CR58 site. Error bars are one standard deviation. Potential temperature (°C), salinity (psu), pH (SWS scale 25 °C) and dissolved oxygen (ml/l) data are also shown.

observation site is very stable compared with the Pacific and Antarctic oceans although the water massed in 500–2000 m and deeper than 2000 m might show the very tiny variation.

The results for the $^{236}U/^{238}U$ atom ratios, ^{238}U and the ^{236}U concentrations (atom/kg) in seawater and SS are shown in Table 1 and Fig. 2, along with the depth profiles for ^{137}Cs. The concentration of ^{137}Cs in the dissolved phase was in the range of 0.12–1.18 mBq/kg. The observed concentrations and the depth profile were similar to data reported previously in the Japan Sea (e.g. Ito et al., 2003; Tanaka et al., 2006). As can be seen from Fig. 2, ^{236}U was successfully measured in all seawater samples from the surface to the sea floor using 20 l sample volumes. The $^{236}U/^{238}U$ atom ratios for dissolved ^{236}U in seawater were in the range of $(0.21-1.65) \times 10^{-9}$, and varied by a factor of about 8. This value is nearly same or a little bit larger than that obtained from the seawater of the Atlantic Ocean (Christl et al., 2012), and three orders of magnitude larger than that of standard seawater (IAEA-381) which was contaminated with the radioactive release from the Sellafield reprocessing plant (Lee et al., 2008). The concentration values showed a subsurface maximum of $(12.7 \pm 2.8) \times 10^6$ atom/kg in the layer at 50 m below the surface and decreased steeply with depth. The minimum value of $(1.56 \pm 0.34) \times 10^6$ atom/kg was found at a depth of 2500 m. It is not surprising that this profile is markedly different from that of natural ^{238}U which is nearly constant over depth, further supporting the recent anthropogenic origin of the ^{236}U. In the SS samples, derived from 20 l of water, ^{236}U could not be detected (count rate similar to the blank value). This corresponds to an upper limit of 2% particle bound ^{236}U compared to the ^{236}U found in solution. The total ^{236}U inventory of the water column was estimated as $(13.7 \pm 0.9) \times 10^{12}$ atom/m^2. This value is nearly the same as the global fallout level (17.8×10^{12} atom/m^2) estimated from the

analyses of soil samples in Japan (Sakaguchi et al., 2010). ^{236}U was also found in the bottom sediments of the Japan Sea (Table 2). The $^{236}U/^{238}U$ atom ratios and ^{236}U concentrations in bottom sediments were in the range of $(0.55 \pm 0.16) \times 10^{-9}-(9.63 \pm 1.35) \times 10^{-9}$ and $(0.36 \pm 0.11) \times 10^7-(4.86 \pm 0.76) \times 10^7$ atoms/g, respectively. The $^{236}U/^{238}U$ atom ratios in the topmost sediment layer are higher than those in the seawater immediately above, which implies that the ^{236}U in the sediment does not originate from the dissolved phase. It is our understanding that only deposition of particle-bound ^{236}U can explain this observation. Whether this can compromise the use of ^{236}U as a conservative radiotracer, depends on the relative importance of particle deposition, which can be gauged by comparison of the dissolved and the deposited ^{236}U inventory. The concentration in seawater, integrated up to the depth of the sediment sample, corresponds to $(1.47 \pm 0.13) \times 10^{10}$ atom/m^2, which is about 100 times more than in the sediment. Also for ^{137}Cs, we have observed deposition with an inventory in the bottom sediment of 37 Bq/m^2. The inventory for ^{137}Cs in sediments in the Japan Sea has been reported by Ito and Otosaka (2007), and our results are comparable. This inventory for ^{137}Cs is about 1/40 of that in the water column (Table 2). In proportion, this is about twice as high as for ^{236}U. The details of the processes scavenging Cs are not yet clear. However, these results indicate that ^{236}U from global fallout has not been effectively scavenged from the water column by sedimentation.

3.2. Depth distributions of ^{236}U and ^{137}Cs

The concentration of ^{236}U is relatively high and constant (9.6×10^6 atom/kg), except one samples from 50 m, in the surface

Table 1
The results of $^{236}U/^{238}U$ atomic ratio, concentration and inventory of ^{236}U and ^{137}Cs in sea water and suspended solid (SS) samples in the Japan Sea.

Sample name	Water								SS
	Depth (m)	$^{236}U/^{238}U$ (10^{-9} atom ratio)	^{238}U concentration (10^{15} atom/kg)	^{236}U concentration (10^6 atom/kg)	^{137}Cs concentration (mBq/kg)	Depth range (m)	^{236}U inventory	^{137}Cs inventory	^{236}U conc. 10^4 atom/kg water
							10^{12} atom/m^2	Bq/m^2	
CR58_1U	20	1.37 ± 0.14	7.12 ± 0.19	9.76 ± 1.33	1.18 ± 0.08	0–20	0.20 ± 0.03	30 ± 2	< 7.99
CR58_2U	50	1.65 ± 0.23	7.70 ± 0.08	12.7 ± 2.81	0.97 ± 0.07	20–50	0.39 ± 0.09	30 ± 3	< 8.25
CR58_3U	100	1.38 ± 0.18	7.01 ± 0.10	9.67 ± 1.43	1.09 ± 0.08	50–100	0.50 ± 0.07	62 ± 5	< 4.54
CR58_4U	250	1.33 ± 0.15	6.95 ± 0.04	9.25 ± 1.94	1.01 ± 0.07	100–250	1.43 ± 0.30	214 ± 18	5.85 ± 5.08
CR58_5U	500	1.04 ± 0.16	7.65 ± 0.11	7.96 ± 1.35	0.90 ± 0.07	250–500	2.05 ± 0.35	287 ± 26	< 7.05
CR58_6U	700	0.81 ± 0.12	7.99 ± 0.07	6.45 ± 1.45	1.09 ± 0.07	500–700	1.32 ± 0.30	251 ± 20	9.22 ± 8.01
CR58_7U	1000	0.67 ± 0.11	7.87 ± 0.08	5.28 ± 1.04	0.62 ± 0.06	700–1000	1.63 ± 0.32	214 ± 22	6.89 ± 9.76
CR58_8U	1500	0.48 ± 0.08	8.28 ± 0.05	3.99 ± 0.94	0.52 ± 0.05	1000–1500	2.05 ± 0.48	316 ± 37	< 4.65
CR58_9U	2000	0.36 ± 0.04	7.50 ± 0.04	2.70 ± 0.36	0.18 ± 0.02	1500–2000	1.39 ± 0.19	101 ± 13	< 7.32
CR58_10U	2150	0.34 ± 0.05	8.31 ± 0.08	2.86 ± 0.48	0.24 ± 0.04	2000–2150	1.47 ± 0.25	42 ± 7	7.53 ± 10.1
CR58_11U	2500	0.21 ± 0.04	7.43 ± 0.09	1.56 ± 0.34	0.12 ± 0.02	2150–2500	0.80 ± 0.17	49 ± 10	< 7.56
CR58_12U	2803	0.23 ± 0.05	7.30 ± 0.10	1.65 ± 0.36	0.16 ± 0.02	2500–2803	0.51 ± 0.11	56 ± 6	9.60 ± 8.35
						total	13.7 ± 0.9	1652 ± 60	

Errors show one sigma standard deviation. ^{137}Cs concentrations are determined in January/2011. Values are not decay corrected.

Table 2
The results on the concentration, isotopic composition and inventory of U isotope and ^{137}Cs in bottom sediments in the Japan Sea.

Depth (cm)	Anal. sediments (g)	Dry bulk sediments (g)	$^{236}U/^{238}U$ (10^{-9} atom ratio)	^{236}U concentration (10^7 atom/g)	^{236}U inventory (10^{10} atom/m^2)	^{137}Cs concentration (Bq/g)	^{137}Cs inventory (Bq/m^2)
0–1	3.43	6.61	9.63 ± 1.35	4.86 ± 0.76	5.06 ± 0.79	0.017 ± 0.001	10.9 ± 0.90
1–2	3.61	7.41	8.70 ± 0.65	4.41 ± 0.39	5.14 ± 0.46	0.020 ± 0.001	15.1 ± 1.11
2–3	3.99	10.48	1.11 ± 0.69	0.59 ± 0.37	0.97 ± 0.61	0.006 ± 0.000₈	5.53 ± 0.69
3–4	3.91	10.75	2.87 ± 0.61	1.67 ± 0.37	2.82 ± 0.63	0.004 ± 0.000₇	4.35 ± 0.76
4–5	3.96	11.69	0.55 ± 0.16	0.36 ± 0.11	0.66 ± 0.20	0.001 ± 0.000₆	0.93 ± 0.70
Total					15.0 ± 1.3		36.8 ± 1.9

water (above 250 m depth). The ^{236}U concentration profile can be described by the diffusion equation

$$\partial C/\partial t = \kappa \times \partial^2 C/\partial z^2$$

In this equation, C is the concentration (atom/kg) of ^{236}U, t is the time (yr) after deposition of ^{236}U to the surface of the Japan Sea (50 yr), κ is the diffusion coefficient (cm^2/s) and z (m) is the depth. As an example, fitting of the whole depth profile with a normal distribution (corresponding to the simplest eddy diffusion case)

$$C(z,t) = (4\pi\kappa t)^{-1/2} \times \exp\{-z^2/(4\kappa t)\}$$

is shown in Fig. 4A. Another fit with the difference-method is shown in Fig. 4B. The parameters in the latter example are: a bottom depth $z_{(bottom)} = 3000$ m, $dC/dz_{(bottom)} = 0$ and the surface concentration $C_{(surface)} = 9.6 \times 10^6$ atom/kg. It is assumed that the concentration for ^{236}U in the surface water (0–250 m) has been nearly constant for about 50 yr. The same models were also fitted to the ^{137}Cs data. In this case, the surface concentration was assumed as 1.09 mBq/kg. The results are shown in Fig. 4C and D. The diffusion coefficient of ^{236}U, obtained by fitting a normal distribution, is 5.6 cm^2/s; using the difference-method, a value of 4.2 cm^2/s was calculated. For ^{137}Cs, the diffusion coefficient obtained for the normal distribution was 4.9 cm^2/s and 4.7 cm^2/s for the difference-method, respectively. As can be seen, the diffusion coefficients of ^{236}U are almost the same as those of ^{137}Cs. These results are similar to those which have been reported for conservative nuclides such as ^{137}Cs (1–10 cm^2/s; Tsumune et al.,1999) or natural Ra isotopes (6 cm^2/s; Tanaka et al., 2006) for depths from the surface to ca. 1000 m.

Actually, the concentrations of ^{236}U and ^{137}Cs show a very good correlation (Fig. 5, $R = 0.99$, filled circles) except for depths of 50 and 700 m (hollow circles), which deviated by about 2σ. Such a deviation is usually not considered sufficiently robust. Additionally, at the depths of 50 and 700 m, water temperature, salinity and dissolved oxygen showed no drastic change. Further measurements with higher precision would be required to confirm that the deviation in ^{236}U/^{237}Cs is real before attempting an oceanographic interpretation.

Plotting the ^{137}Cs/^{236}U ratios versus depth (Fig. 6), suggests a change between 1500 and 2000 m. The average ratios, above and below $\sim$1750 m (except for depths of 50 m and 700 m) are 1.17 $\pm$ 0.05, and 0.81 $\pm$ 0.13, respectively. According to the work of Gamo and Horibe (1983), the Japan Sea Proper Water can be divided into three parts: the Upper Portion Proper Water, the Deep Water, and the Bottom Water. The water column below 2000 m, where we observed a lower ^{137}Cs/^{236}U value, corresponds to the bottom water layer. As can be seen from Fig. 3, for these depths the concentration of ^{137}Cs does not show a good agreement with fitted curves, while the ^{236}U agrees well. Considering the larger proportion of ^{137}Cs in the bottom sediment, it seems plausible that the deficit of ^{137}Cs in the Bottom Water has been scavenged to the sediment. Apparently, the scavenging process with precipitated materials and/or resuspended sediments is less effective for uranium. Another possibility is that the bottom water is derived from a water mass which has a different origin than the upper part. Further investigations of these nuclides for other areas in the Japan Sea are needed to clarify these observations.
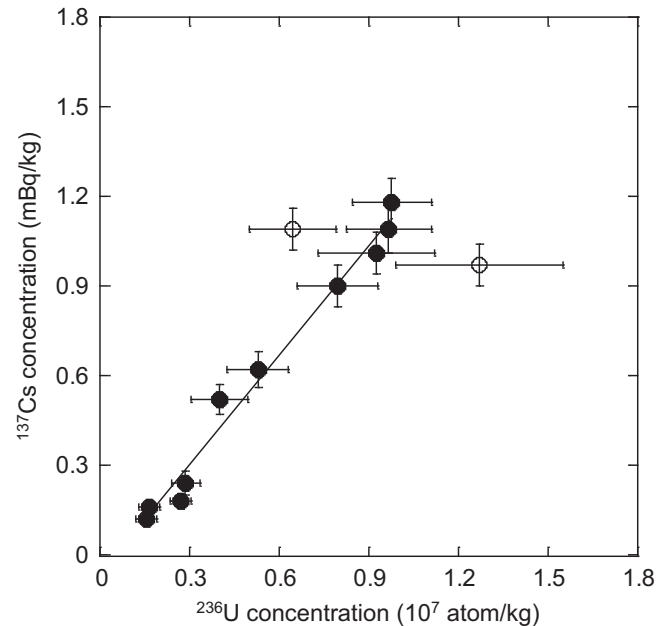

Fig. 5. Concentrations of dissolved ^{236}U (atom/kg) and ^{137}Cs (mBq/kg) in the water column as a function of depth. Error bars are one standard deviation. (See text for explanation further details).
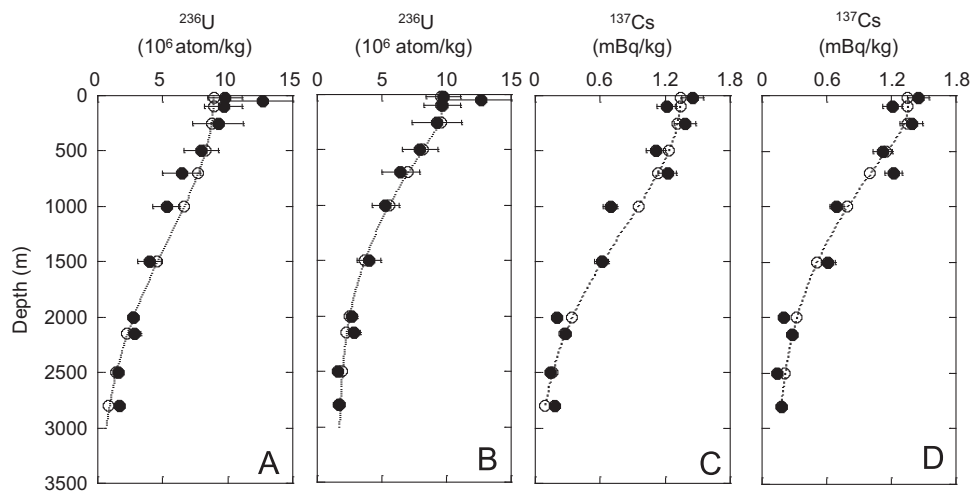

Fig. 4. Comparison of fit for a normal distribution (eddy-diffusion) A and C, and the difference-method B and D. Filled circles show the measured concentrations of ^{236}U (atom/kg) and ^{137}Cs (mBq/kg) in sea water, while the open circles and the broken line represent the fit.
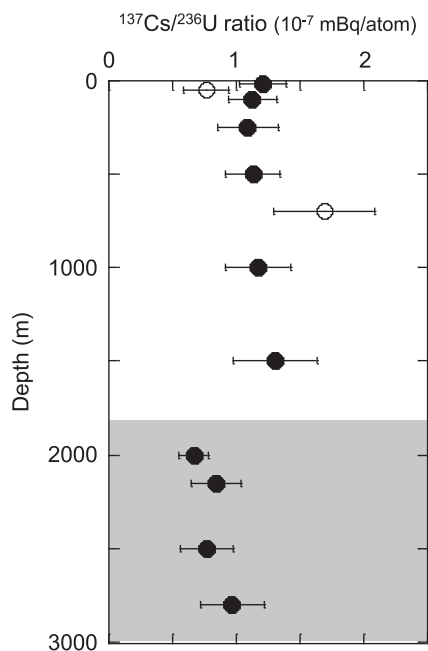
Fig. 6. Depth profile for $^{137}Cs/^{236}U$ $(10^{-7}$ mBq/atom) in the water column at station CR58. Error bars represent one standard deviation.

Conclusions

^{236}U, introduced into the environment by nuclear weapons testing in the 1960s, was measured together with ^{137}Cs in seawater, suspended materials and sediments around the Yamato basin/ridge of the Japan Sea with a view to developing a new oceanic tracer.

In the feasibility study, one depth profile for ^{236}U in the Japan Sea was presented. Our results for ^{236}U in seawater demonstrate that the ^{236}U concentration can be accurately determined for 20 l samples using AMS, even at a depth of 2500–2800 m which showed the lowest ^{236}U concentration. The depth profiles of ^{236}U in the seawater column showed a similar profile to ^{137}Cs. The inventory of ^{236}U in seawater was nearly the same as that from global fallout on land in Japan. Only one hundredth of the ^{236}U inventory of the water column was detected in the seafloor sediments. No ^{236}U above the blank value could be detected in the SS. These results suggest that ^{236}U behaves as a conservative nuclide and is dissolved in the water column without being subject to any effective scavenging after 50 yr. Our results provide important information on the behaviour of ^{236}U in the ocean and suggest that ^{236}U has great potential as an oceanic tracer. Encouraged by the promising results obtained for this first depth profile, we plan a comprehensive study of the circulation of sea water in the Japan Sea, including the Japan Sea Proper Water, using a complete dataset for U, Pu and Cs isotopes in water, SS and sediments from seven sites of the cruise KH10-02.

Acknowledgements

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