

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

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22
23 **ABSTRACT**

24 We present a feasibility study for using ²³⁶U as an oceanic circulation tracer based on
25 depth profiles of ²³⁶U and ¹³⁷Cs in the Japan Sea. The concentration of the
26 predominantly anthropogenic ²³⁶U, measured with Accelerator Mass Spectrometry
27 (AMS), decreased from $(13 \pm 3) \times 10^6$ atom/kg in surface water to $(1.6 \pm 0.3) \times 10^6$
28 atom/kg close to the sea floor (2800 m). The profile has a smooth trend with depth and
29 concentration values are generally proportional to that of ¹³⁷Cs for the same water
30 samples, but with a slightly lower ratio of ¹³⁷Cs/²³⁶U below 2000 m. The cumulative
31 inventory of dissolved ²³⁶U in the water column was estimated to be $(13.7 \pm 0.9) \times 10^{12}$
32 atom/m², which is similar to the global-fallout level (17.8×10^{12} atom/m²) in Japan.

1 Additional analyses of suspended solids (SS) and bottom sediments yielded negligible
2 amounts of ^{236}U . Our results suggest that ^{236}U behaves as a conservative nuclide in
3 seawater, with potential advantages over other tracers of oceanic circulation.

4 5 **1. Introduction**

6 The Japan Sea in the western Pacific Ocean is separated by four shallow channels
7 (50-100 m in depth). The average and maximum depths are ca. 1700 m and 3800 m,
8 respectively (Chronological scientific table, 2009). This deep bowl-like sea has one
9 large input from the surrounding ocean through the Tsushima strait, forming the
10 Tsushima Warm Current which dominates the surface layer. Without inflow from the
11 open ocean, formation of deepwater occurs locally in the north west part of the Japan
12 Sea and is similar to the processes in the world oceans (e.g. Uda, 1934; Nitani, 1972;
13 Gamo and Horibe, 1983; Sudo, 1986). The product is a stable deep water body, the
14 Japan Sea Proper Water, which has an extremely narrow range in water temperature
15 (0.0-0.1°C) and low salinity (34.0-34.1psu), and is highly oxygenated (dissolved
16 oxygen: 0.2-0.25 mM) (Asaoka 1987). Therefore, past and ongoing efforts by
17 oceanographers have concentrated on the role of the Japan Sea as a “Miniature Ocean”
18 (e.g. Uda, 1934). Extensive oceanographic observations have been carried out,
19 employing advanced techniques to measure currents and water properties, but also
20 three-dimensional numerical models with real topography and time-varying
21 meteorological forcing, to improve our understanding of sophisticated phenomena
22 occurring in the Japan Sea (Kim et al., 2008).

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

1 more detailed studies on the circulation of water and migration of materials are needed.

2 Cs-137 ($T_{1/2}=30.2$ y) has been spread worldwide as a fission product of atmospheric
3 nuclear weapons testing in the 1960s. This nuclide has proved to be a powerful tool for
4 oceanography due to its well-defined origin and conservative behaviour in water (e.g.
5 Aoyama and Hirose, 1995; Miyao et al., 1998; Ito et al., 2003). However, the number of
6 atoms has now decayed to about one third compared with initial levels, and will become
7 even more difficult to measure in the future. Given this situation, we have focused on
8 ^{236}U ($T_{1/2}=2.342\times 10^7$ y) as a candidate for a new isotopic tracer for oceanography.
9 Reliable measurement of ^{236}U in the environment has become possible only recently, as
10 a result of progress in high sensitivity measurement based on accelerator mass
11 spectrometry (AMS) (Steier et al., 2008). Sakaguchi et al. (2009, 2010) showed that
12 global fallout from nuclear weapons testing contains ^{236}U , which is mainly produced via
13 the nuclear reaction $^{238}\text{U}(n,3n)$ and about 900 kg of ^{236}U have been distributed in the
14 surface environment. Thus, ^{236}U may have the potential to act as a tracer for
15 environmental dynamics similar to that of ^{137}Cs .

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

20 **2. Materials and Methods**

21 **2.1 Samples**

22 The Japan Sea samples were collected on cruise KH10-02 of the research vessel
23 Hahuko Maru, July to August 2010. Bottom sediments (CR 14; $39^{\circ}32.56'\text{N}$,
24 $136^{\circ}40.35'\text{E}$) and seawater samples (CR 58; $40^{\circ}25.66'\text{N}$, $135^{\circ}55.21'\text{E}$) were collected
25 around the Yamato Basin, as shown in Fig. 1. The water depths at CR 14 and CR 58
26 were 2653 m and 2803 m, respectively. Samples of about 20 L seawater were taken with
27 Niskin bottles at 12 different depths. Water temperature, dissolved oxygen (DO), pH,
28 salinity and some other basic properties of seawater were measured precisely at 25
29 different depths with routine protocols established for the GEOTRACES project.
30 Immediately after sample collection, the water was filtered with $0.45\ \mu\text{m}$ pore-size
31 membrane filters using a Teflon[®] filtration system. The filtrate was stored in

1 polyethylene containers after adjustment to pH 1 by the addition of conc. HNO₃. The SS
2 collected on the filters was packed into a double plastic bag. Sediment core samples
3 were taken by a multi-corer device with a diameter of 8.9 cm. The surface of the cores
4 was not disturbed during sampling and handling of the sediment core samples. The
5 cores were extruded in 1 cm segments, which were cut off and stored in a refrigerator at
6 4°C within a few hours of sampling. Samples from the surface to 5 cm in depth were
7 used for the present study. The procedure does lend itself to the possibility for cross-
8 contamination from smearing between the layers. This is especially true for the topmost
9 sample of the core (0-2 cm), which had a watery consistency.

11 2.2 Chemical treatments and measurements

12 The filtered seawater sample was put into a polyethylene vessel (approximately 20
13 L). The following chemical tracers and carriers were added: 30 µg of an in-house
14 standard "Vienna-UPu1" used at the VERA Laboratory which is a mix of IRMM-058
15 and IRMM-085 with a gravimetric dilution to $(4.76 \pm 0.05) \times 10^{10}$ atom ²³³U/g, and
16 $(2.69 \pm 0.03) \times 10^{11}$ atoms ²⁴²Pu/g, 200 mg of ¹³³Cs (Wako, analytical grade CsCl), and
17 200 mg of iron (Fe) as Fe/HCl solution. The seawater sample was heated for three hours
18 with stirring and then stood for 12 hours. After re-adjustment of the pH to 1 with HNO₃
19 and ammonium solution, 4 g of ammonium-phosphomolybdate (AMP) powder was
20 added to adsorb Cs. After stirring for 1 hour and standing for 24 hours at room
21 temperature, the supernatant was removed first with a siphon, followed by
22 centrifugation (the supernatant was kept for U and Pu analyses). The Cs-adsorbed AMP
23 was dried at 105°C for 12 hours, and packed into a plastic tube (1.4 cmx4.0 cm) for the
24 measurement of ¹³⁷Cs.

25 U and Pu in the supernatant were co-precipitated with Fe(OH)₃ at pH 8. After
26 removing the supernatant by a siphon and centrifugation, the Fe(OH)₃ was dissolved in
27 10 M HCl. Further purification of U and Pu from this solution was conducted by using
28 columns packed with anion-exchange resin as described by Sakaguchi et al. (2009,
29 2010).

30 The sea floor sediment samples were dried at 105°C for 12 hours. An aliquot (about
31 4 g) of sediment was totally digested with the methods described by Sakaguchi et al.

1 (2004) after the addition of known amounts of ^{233}U , ^{242}Pu , and ^{133}Cs as yield tracers for
2 each element. A part of the digested solution was precisely sub sampled (by weight) for
3 measurement of ^{133}Cs by ICP-MS. After dilution with MQ water to 1 L and adjusting
4 the remaining solution to pH 1 with HNO_3 and ammonium solution, 2 g of AMP was
5 added to the solution. Analysis of Cs-adsorbed AMP and purification of U and Pu were
6 conducted in a similar way as that for seawater.

7 The treatment for the SS samples was the same as used for the sediments, but
8 measurement of ^{137}Cs was not attempted. Inoue et al. (personal comm.) previously had
9 tried to measure ^{137}Cs in SS, filtered from more than 80 L of seawater (Japan Sea) using
10 a Ge-detector with an ultra-low background level installed at the Ogoya Underground
11 Laboratory (OUL), LLRL, Kanazawa University, but was unsuccessful.

12 For analysis of the AMP samples from the seawater and sediment samples, the 662-
13 keV line of ^{137}Cs was measured by γ -ray spectrometry using a well-type Ge detector
14 (ORTEC, GWL-120230-S) installed at Hiroshima University. For the sediment samples
15 a low-background well-type Ge detector (EURISYS, EGPC 150 P16) installed at LLRL
16 was used, as the ^{137}Cs level was expected to be very low. The detection limits for ^{137}Cs
17 in both detectors were estimated as 0.007 Bq/day and 0.002 Bq/day, respectively. The
18 uncertainty for each sample was about a few percent expressed as 1σ . The spectrometer
19 was calibrated with a mixed standard prepared by the Japan Radioisotope Association
20 (No. MX-033).

21 A part of the AMP was dissolved with 1.25% tetramethylammonium hydroxide
22 (TMAH) and the ^{133}Cs concentration was measured to assess the yield of adsorbed ^{137}Cs .
23 We calculated the recovery for Cs as 98-100 %.

24 For the measurement of ^{236}U , ^{238}U , ^{239}Pu , and ^{240}Pu with AMS, the samples were
25 prepared as U or Pu oxides in typically 3 mg of Fe_2O_3 matrix. A detailed description of
26 the AMS measurement procedures for ^{236}U and $^{239}\text{Pu}/^{240}\text{Pu}$ at VERA are described in
27 Steier et al. (2010) and Sakaguchi et al. (2010). The solid sample is sputtered with a Cs
28 beam, negative sample ions (U^{16}O^- and Pu^{16}O^-) are extracted, and pass through the first
29 mass spectrometer. The selected ions are accelerated to an energy of 3 MeV, the
30 molecular ions are broken up and stripped to positive charge states in a gas cell, and the
31 atomic breakup products undergo a second acceleration. The actinide ions of interest

1 (U^{5+} or $^{239}Pu^{5+}$, at approximately 18 MeV) are separated in a second mass spectrometer.
2 The high particle energy provided by the accelerator, the destruction of molecular
3 isobars in the stripping process, and the repeated deployment of high-resolving electric
4 and magnetic analyzers explains the exceptional abundance sensitivity of AMS.

5 The spectrometers are programmed to rapidly switch between the different isotopes.
6 The uranium isotopes were determined by counting $^{233}U^{5+}$ and $^{236}U^{5+}$ with a gas
7 ionization detector, and by measuring the $^{238}U^{5+}$ beam current in a Faraday cup. The
8 resultant $^{238}U^{5+}$ current for samples, containing about 50 μg of uranium, was of the
9 order of 100 pA, compared to typically 50 nA observed for pure U_3O_8 samples.
10 However, these currents are well in the range of the current amplifiers (SR570, Stanford
11 Research Systems, Inc, Sunnyvale, California, USA) connected to the Faraday cups.
12 The process blanks gave currents of 1 pA or below. To achieve sufficient precision for
13 the current measurement, the integration time for $^{238}U^{5+}$ was extended to up to 1 s.
14 VERA achieves an abundance sensitivity for $^{236}U/^{238}U$ of 10^{-13} . The overall detection
15 efficiency is 1 count in the detector per ~ 3000 actinide atoms in the sample, with the
16 major losses taking place during sputtering (1% negative ion yield) and stripping (5%
17 yield of 5+). This limits the sensitivity for plutonium, where no abundant isotope exists.
18 Concentrations of ^{236}U and ^{238}U in seawater and SS samples were determined by using
19 the measured ratios of $^{236}U/^{233}U$ and $^{233}U/^{238}U$ and the known amount of ^{233}U tracer
20 added. The results for Pu will be presented elsewhere. The recovery of U for chemical
21 purification was 80-95%.

22 23 **3. Results and discussion**

24 **3.1 Concentrations and inventories of ^{236}U and ^{137}Cs in the Japan Sea**

25 The fundamental oceanographic parameters such as temperature ($^{\circ}C$), salinity (psu), pH
26 (SWS scale $25^{\circ}C$) and dissolved oxygen (ml/l) in water from CR58 site are shown in
27 Fig.2. The θ -T diagram of this station is also presented in Fig. 3. From consideration of
28 the depth profiles for these parameters and Fig. 3, it can be said that the compositional
29 data are typical for Japan Sea samples: composition of the deeper water-mass at this
30 observation site is very stable compared with the Pacific and Antarctic oceans although
31 the water massed in 500-2000 m and deeper than 2000 m might show the very tiny
32 variation.

1 The results for the $^{236}\text{U}/^{238}\text{U}$ atom ratios, ^{238}U and the ^{236}U concentrations (atom/kg) in
2 seawater and SS are shown in Table 1 and Fig. 2, along with the depth profiles for ^{137}Cs .
3 The concentration of ^{137}Cs in the dissolved phase was in the range of 0.12-1.18 mBq/kg.
4 The observed concentrations and the depth profile were similar to data reported
5 previously in the Japan Sea (e.g. Ito et al., 2003; Nakano 2009). As can be seen from
6 Fig. 2, ^{236}U was successfully measured in all seawater samples from the surface to the
7 sea floor using 20 L sample volumes. The $^{236}\text{U}/^{238}\text{U}$ atom ratios for dissolved ^{236}U in
8 seawater were in the range of $(0.21-1.65) \times 10^{-9}$, and varied by a factor of about 8. The
9 concentration values showed a subsurface maximum of $(12.7 \pm 2.8) \times 10^6$ atom/kg in the
10 layer at 50 m below the surface and decreased steeply with depth. The minimum value
11 of $(1.56 \pm 0.34) \times 10^6$ atoms/kg was found at a depth of 2500 m. It is not surprising that
12 this profile is markedly different from that of natural ^{238}U which is nearly constant over
13 depth, further supporting the recent anthropogenic origin of the ^{236}U . In the SS samples,
14 derived from 20 L of water, ^{236}U could not be detected (current rate similar to the blank
15 value). This corresponds to an upper limit of 2% particle bound ^{236}U compared to the
16 ^{236}U found in solution. The total ^{236}U inventory of the water column was estimated as
17 $(13.7 \pm 0.9) \times 10^{12}$ atom/m². This value is nearly the same as the global fallout level
18 $(17.8 \times 10^{12}$ atom/m²) estimated from the analyses of soil samples in Japan (Sakaguchi et
19 al., 2010). ^{236}U was also found in the bottom sediments of the Japan Sea (Table 2). The
20 $^{236}\text{U}/^{238}\text{U}$ atom ratios and ^{236}U concentrations in bottom sediments were in the range of
21 $(0.55 \pm 0.16) \times 10^{-9}$ - $(9.63 \pm 1.35) \times 10^{-9}$ and $(0.36 \pm 0.11) \times 10^7$ - $(4.86 \pm 0.76) \times 10^7$ atoms/g,
22 respectively. The $^{236}\text{U}/^{238}\text{U}$ atom ratios in the topmost sediment layer are higher than
23 those in the seawater immediately above, which implies that the ^{236}U in the sediment
24 does not originate from the dissolved phase. It is our understanding that only deposition
25 of particle-bound ^{236}U can explain this observation. Whether this can compromise the
26 use of ^{236}U as a conservative radiotracer, depends on the relative importance of particle
27 deposition, which can be gauged by comparison of the dissolved and the deposited ^{236}U
28 inventory. The concentration in seawater, integrated up to the depth of the sediment
29 sample, corresponds to $(1.47 \pm 0.13) \times 10^{10}$ atom/m², which is about 100 times more than
30 in the sediment. Also for ^{137}Cs , we have observed deposition with an inventory in the
31 bottom sediment of 37 Bq/m². The inventory for ^{137}Cs in sediments in the Japan Sea has
32 been reported by Ito and Otosaka (2007), and our results are comparable. This inventory

1 for ^{137}Cs is about 1/40 of that in the water column (Table 2). In proportion, this is about
2 twice as high as for ^{236}U . The details of the processes scavenging Cs are not yet clear.
3 However, these results indicate that ^{236}U from global fallout has not been effectively
4 scavenged from the water column by sedimentation.

5 6 **3.2. Depth distributions of ^{236}U and ^{137}Cs**

7 The concentration of ^{236}U is relatively high and constant (9.6×10^6 atom/kg), except
8 one samples from 50 m, in the surface water (above 250 m depth). The ^{236}U
9 concentration profile can be described by the diffusion equation:

$$10 \quad \partial C / \partial t = \kappa \cdot \partial^2 C / \partial z^2$$

11
12
13 In this equation, C is the concentration (atom/kg) of ^{236}U , t is the time (y) after
14 deposition of ^{236}U to the surface of the Japan Sea (50 years), κ is the diffusion
15 coefficient (cm^2/s) and z (m) is the depth. As an example, fitting of the whole depth
16 profile with a normal distribution (corresponding to the simplest eddy diffusion case)

$$17 \quad C(z, t) = (4\pi\kappa t)^{-1/2} \cdot \exp\{-z^2/(4\kappa t)\}$$

18
19
20 is shown in Fig. 4A. Another fit with the difference-method is shown in Fig. 4B. The
21 parameters in the latter example are: a bottom depth $z_{(\text{bottom})} = 3000$ m, $dC/dz_{(\text{bottom})} = 0$
22 and the surface concentration $C_{(\text{surface})} = 9.6 \times 10^6$ atom/kg. It is assumed that the
23 concentration for ^{236}U in the surface water (0-250 m) has been nearly constant for about
24 50 years. The same models were also fitted to the ^{137}Cs data. In this case, the surface
25 concentration was assumed as 1.09 mBq/kg. The results are shown in Figs. 4C and 4D.
26 The diffusion coefficient of ^{236}U , obtained by fitting a normal distribution, is $5.6 \text{ cm}^2/\text{s}$;
27 using the difference-method, a value of $4.2 \text{ cm}^2/\text{s}$ was calculated. For ^{137}Cs , the
28 diffusion coefficient obtained for the normal distribution was $4.9 \text{ cm}^2/\text{s}$ and $4.7 \text{ cm}^2/\text{s}$
29 for the difference-method, respectively. As can be seen, the diffusion coefficients of
30 ^{236}U are almost the same as those of ^{137}Cs . These results are similar to those which have
31 been reported for conservative nuclides such as ^{137}Cs ($1-10 \text{ cm}^2/\text{s}$; Tsumune et al., 1996)

1 or natural Ra isotopes ($6 \text{ cm}^2/\text{s}$; Tanaka et al., 2006) for depths from the surface to ca.
2 1000 m.

3 Actually, the concentrations of ^{236}U and ^{137}Cs show a very good correlation (Fig. 5,
4 $R=0.99$, filled circles) except for depths of 50 and 700 m (hollow circles), which
5 deviated by about 2σ . Such a deviation is usually not considered sufficiently robust.
6 Additionally, at the depths of 50 and 700 m, water temperature, salinity and dissolved
7 oxygen showed no drastic change. Further measurements with higher precision would
8 be required to confirm that the deviation in $^{236}\text{U}/^{237}\text{Cs}$ is real before attempting an
9 oceanographic interpretation.

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

24 25 **4. Conclusions**

26 ^{236}U , introduced into the environment by nuclear weapons testing in the 1960's, was
27 measured together with ^{137}Cs in seawater, suspended materials and sediments around
28 the Yamato basin/ridge of the Japan Sea with a view to developing a new oceanic tracer.
29 In the feasibility study, one depth profile for ^{236}U in the Japan Sea was presented. Our
30 results for ^{236}U in seawater demonstrate that the ^{236}U concentration can be accurately
31 determined for 20 L samples using AMS, even at a depth of 2500-2800 m which

1 showed the lowest ^{236}U concentration. The depth profiles of ^{236}U in the seawater column
2 showed a similar profile to ^{137}Cs . The inventory of ^{236}U in seawater was nearly the same
3 as that from global fallout on land in Japan. Only one hundredth of the ^{236}U inventory of
4 the water column was detected in the seafloor sediments. No ^{236}U above the blank value
5 could be detected in the SS. These results suggest that ^{236}U behaves as a conservative
6 nuclide and is dissolved in the water column without being subject to any effective
7 scavenging after 50 years. Our results provide important information on the behavior of
8 ^{236}U in the ocean and suggest that ^{236}U has great potential as an oceanic tracer.
9 Encouraged by the promising results obtained for this first depth profile, we plan a
10 comprehensive study of the circulation of sea water in the Japan Sea, including the
11 Japan Sea Proper Water, using a complete dataset for U, Pu and Cs isotopes in water, SS
12 and sediments from seven sites of the cruise KH10-02.

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26 References

- 27 Aoyama, M., Hirose, K., 1995. The temporal and spatial variation of ^{137}Cs
28 concentration in the western North Pacific and its marginal seas during the period
29 from 1979 to 1988, *J. Environ. Radioactivity* 29, 57-74.
- 30 Asaoka, O., 1987. *Encyclopedia of Oceanography*, Tokyodoshuppan, Co., Tokyo. pp.
31 333-384. (in Japanese).
- 32 Chronological scientific table, National Astronomical Observatory, Maruzen, 2009.

- 1 Gamo, T and Horibe, Y., 1983. Abyssal circulation in the Japan Sea, *J. Oceanogr. Soc. Japan* 39, 220-230.
- 2
- 3 Ito, T., Aramaki, T., Kitamura, T., Otsuka, S., Suzuki, T., Togawa, O., Kobayashi, T.,
4 Senjyu, T., Chaykovskaya, E.L., Karasev, E.V., Lishavskaya, T.S., Novichkov, V.P.,
5 Tkalin, A.V., Shcherbinin, A.F., Volkov, Y.N., 2003. Anthropogenic radionuclides
6 in the Japan Sea: their distributions and transport processes, *J. Environ.*
7 *Radioactivity* 68, 249–267.
- 8 Ito, T. and Otsuka, S., 2007. Transport process of radionuclides in the Japan Sea
9 obtained by JAEA's expeditions. *Jpn. J. Health Phys.* 42, 234-246.
- 10 Kim, K., Kim, K.R., Min, D.H., Yoon, J.H., Takematsu, M., 2001. Warming and
11 Structural Changes in the East(Japan) Sea: A Clue to Future Changes in Global
12 Oceans? *Geophys. Res. Lett.*, 28, 3293-3296.
- 13 Kim, K., Chang, K.I., Kang, D.J., Kim, Y. H., Kim, H., Lee JH. 2008. Review of Recent
14 Findings on the Water Masses and Circulation in the East Sea (Sea of Japan), *J.*
15 *Oceanography*, 64, 721-735.
- 16 Min, D.-H., Warner, M.J., 2005. Basin-wide circulation and ventilation study in the East
17 Sea (Sea of Japan) using chlorofluorocarbon tracers, *Deep-Sea Res. II* 52, 1580-
18 1616.
- 19 Miyao, T., Hirose. K., Aoyama, M., Igarashi, Y., 1998. Temporal variation of ^{137}Cs and
20 $^{239,240}\text{Pu}$ in the Sea of Japan. *J. Environ. Radioactivity* 40, 239-250.
- 21 Nitani, H., 1972. On the deep and the bottom waters in the Japan Sea. In: D. Shoji,
22 Editor, *Researches in Hydrography and Oceanography*, Hydrographic Department
23 of Japan, Maritime Safety Agency, Tokyo, pp. 151-201.
- 24 Nuclear Emergency Response Headquarters Government of Japan. 2011. Report of the
25 Japanese Government to the IAEA Ministerial Conference on Nuclear Safety- The
26 Accident at TEPCO's Fukushima Nuclear Power Stations -. VI Discharge of
27 Radioactive Materials to the Environment, VI-1-VI-7 and appendix VI.
- 28 Sakaguchi, A., Kawai, K., Steier, P., Quinto, F., Mino, M., Tomita, J., Hosh,i M.,
29 Whitehead, N., Yamamoto, M., 2009. First results on ^{236}U levels in global fallout.
30 *Sci. Total Environ.* 407, 4238-4242.

- 1 Sakaguchi, A., Kawai, K., Steier, P., Imanaka, T., Hoshi, M., Endo, S., Zhumadilov, K.,
2 Yamamoto, M., 2010. Feasibility of using ^{236}U to reconstruct close-in fallout
3 deposition from the Hiroshima Atomic Bomb. *Sci. Total Environ.* 408, 5392-5398.
- 4 Sakaguchi, A., Yamamoto, M., Shimizu, T., Koshimizu, S., 2004. Geochemical record
5 of U and Th isotopes in bottom sediments of Lake Kawaguchi at the foot of Mt.
6 Fuji, Central Japan, *J. Radioanal. Nucl. Chem.* 262, 617-628.
- 7 Senjyu, T., Shin, H.R., Yoon, J.H., Nagano, Z., An, H.S., Byun, S.K., Lee, C.K., 2005.
8 Deep flow field in the Japan/East Sea as deduced from direct current measurements,
9 *Deep-Sea Res. II* 52, 1726–1741.
- 10 Steier, P., Bichler, M., Fifield, L.K., Golser, R., Kutschera, W., Priller, A., Quinto, F.,
11 Richter, S., Srnecik, M., Terrasi, P., Wacker, L., Wallner, A., Wallner, G., Wilcken,
12 K.M., Wild, E.M., Natural and anthropogenic ^{236}U in environmental samples. 2008.
13 *Nucl. Instr. and Meth. B* 266, 2246-2250.
- 14 Steier, P., Dellinger, F., Forstner, O., Golser, R., Knie, K., Kutschera, W., Priller, A.,
15 Quinto, F., Srnecik, M., Terrasi, F., Vockenhuber, C., Wallner, A., Wallner, G., Wild,
16 E.M. 2010. Analysis and application of heavy isotopes in the environment. *Nucl*
17 *Instr and Meth B* 268, 1045-1049.
- 18 Sudo, H., 1986. A note on the Japan Sea Proper Water, *Progr Oceanogr*, 17, 313-336.
- 19 Tanaka, K., Inoue, M., Misono, J., Komura, K., 2006. Vertical profiles of ^{226}Ra , ^{228}Ra
20 and ^{137}Cs activities in seawater around the Yamato Ridge and coastal areas of the
21 Sea of Japan, *Chikyukagaku*, 40, 167-176 (in Japanese).
- 22 Tsumune, D., Suzuki, H., Saegusa, T., Maruyama, K., Ito, C., Watabe, N., 1999. Study
23 on transport safety of fresh MOX fuel-Radiation dose from package hypothetical
24 submerged into sea, Abiko research laboratory report, Central Research Institute of
25 Electric Power Industry, U98029, pp. 1-18 (in Japanese).
- 26 Uda, M., 1934. The results of simultaneous oceanographical investigations in the Japan
27 Sea and its adjacent waters in May and June, 1932. *J. imp. Fish. Exp. Sta.*, 5, 57-
28 190 (in Japanese).
- 29 Yamamoto, M., Tsumura, A., Katayama, Y., Tsukatani, T., 1996. Plutonium isotopic
30 composition in soil from the Former Semipalatinsk Nuclear Test Site, *Radiochim.*
31 *Acta*, 72, 209–215.

1 Yamamoto, M., Hoshi, M., Takada, J., Sakaguchi, A., Apsalikov, K.N., Gusev, B.I.,
2 2004. Plutonium, ¹³⁷Cs and U in some pond and lake sediments from areas
3 surrounding the Semipalatinsk Nuclear Test Site: With emphasis on anomalously
4 high U accumulation, J. Radioanal. Nuc. Chem. 262, 607-616.

Highlights:

- > We present a first study using ^{236}U as a new oceanic circulation tracer.
- > We could measure anthropogenic global-fallout ^{236}U in sea water and sea sediments.
- > Depth profile of ^{236}U was nearly proportional to that of ^{137}Cs observed in the same water samples.
- > ^{236}U behaves conservative in seawater.

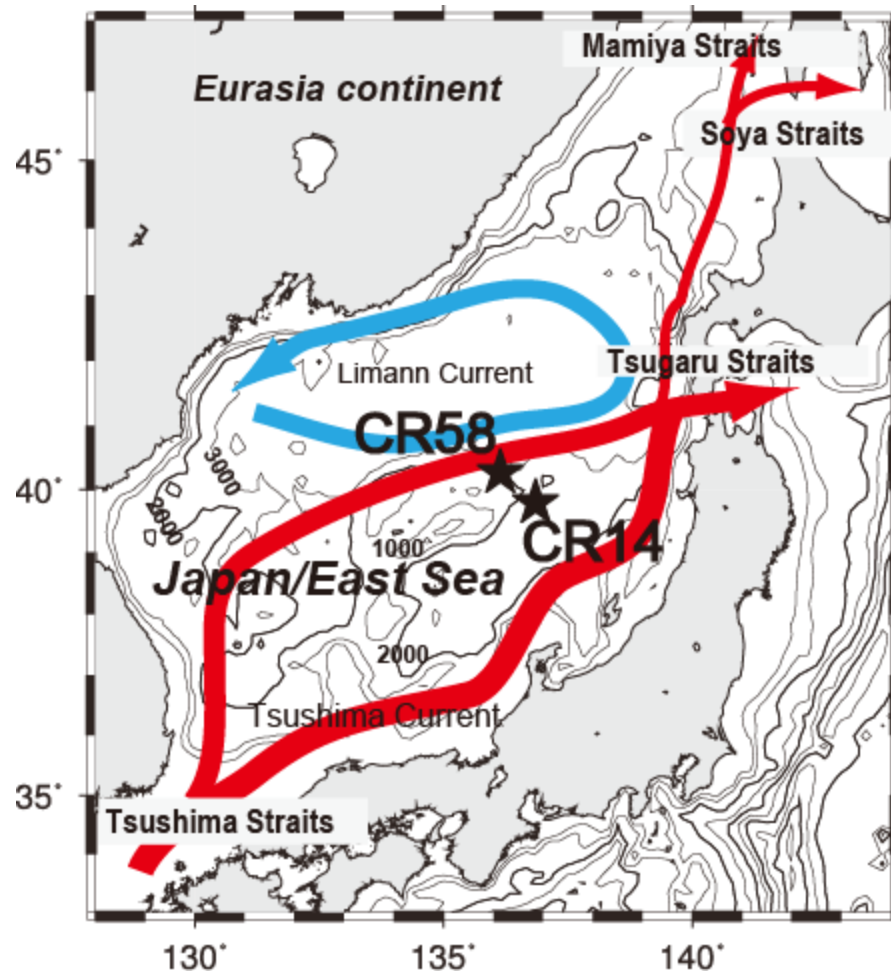


Fig. 1

Figure2

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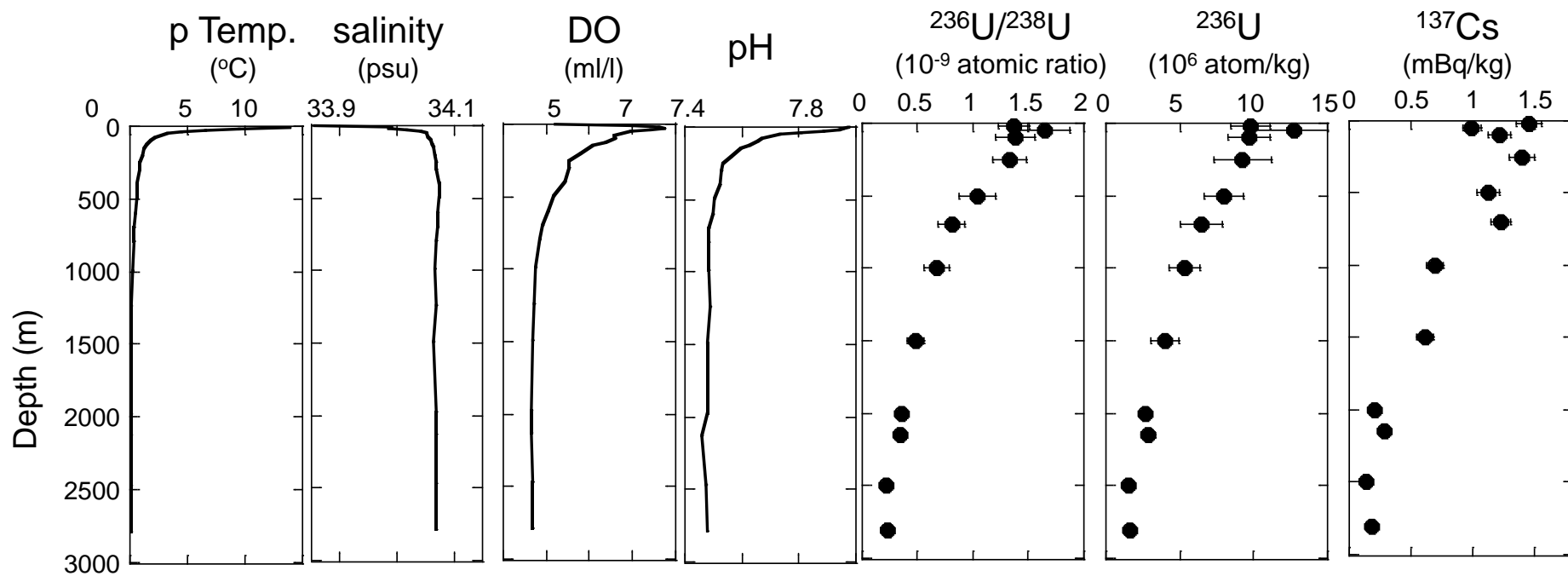


Fig. 2

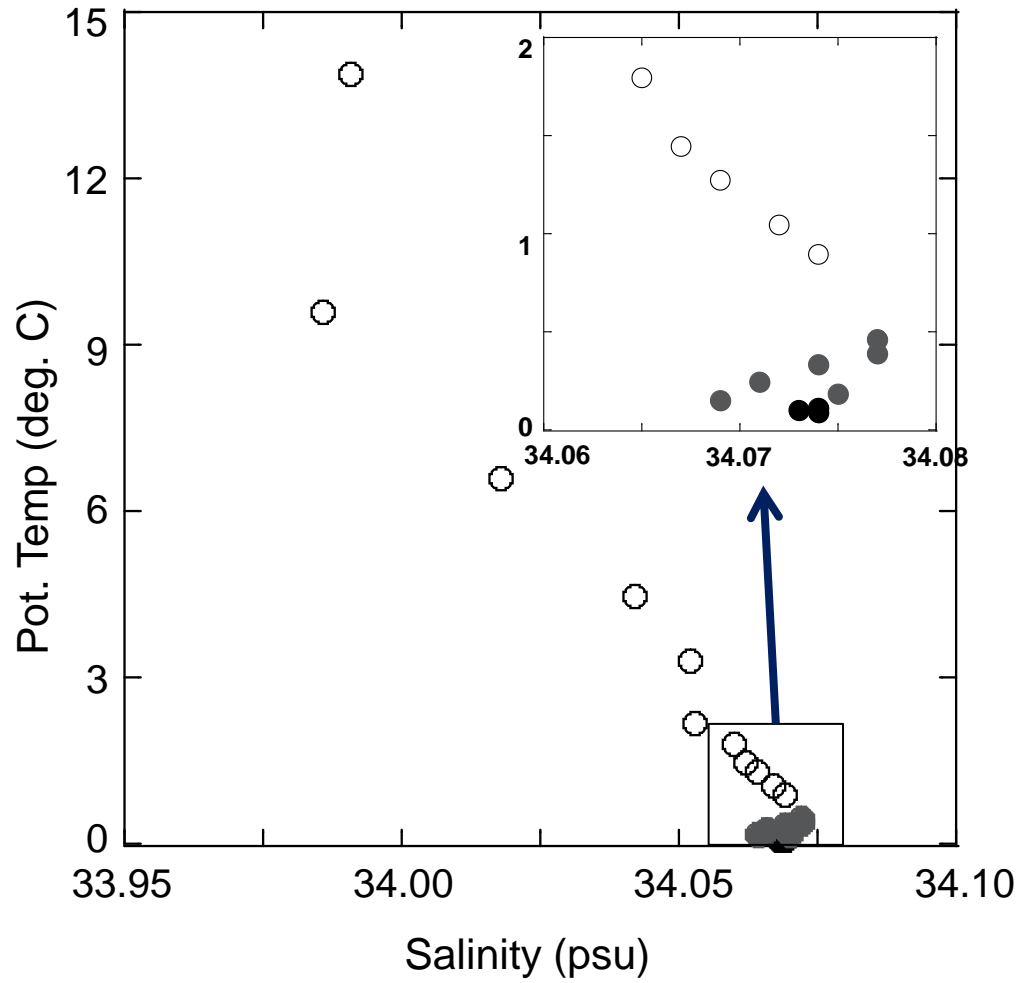


Fig. 3

Figure4

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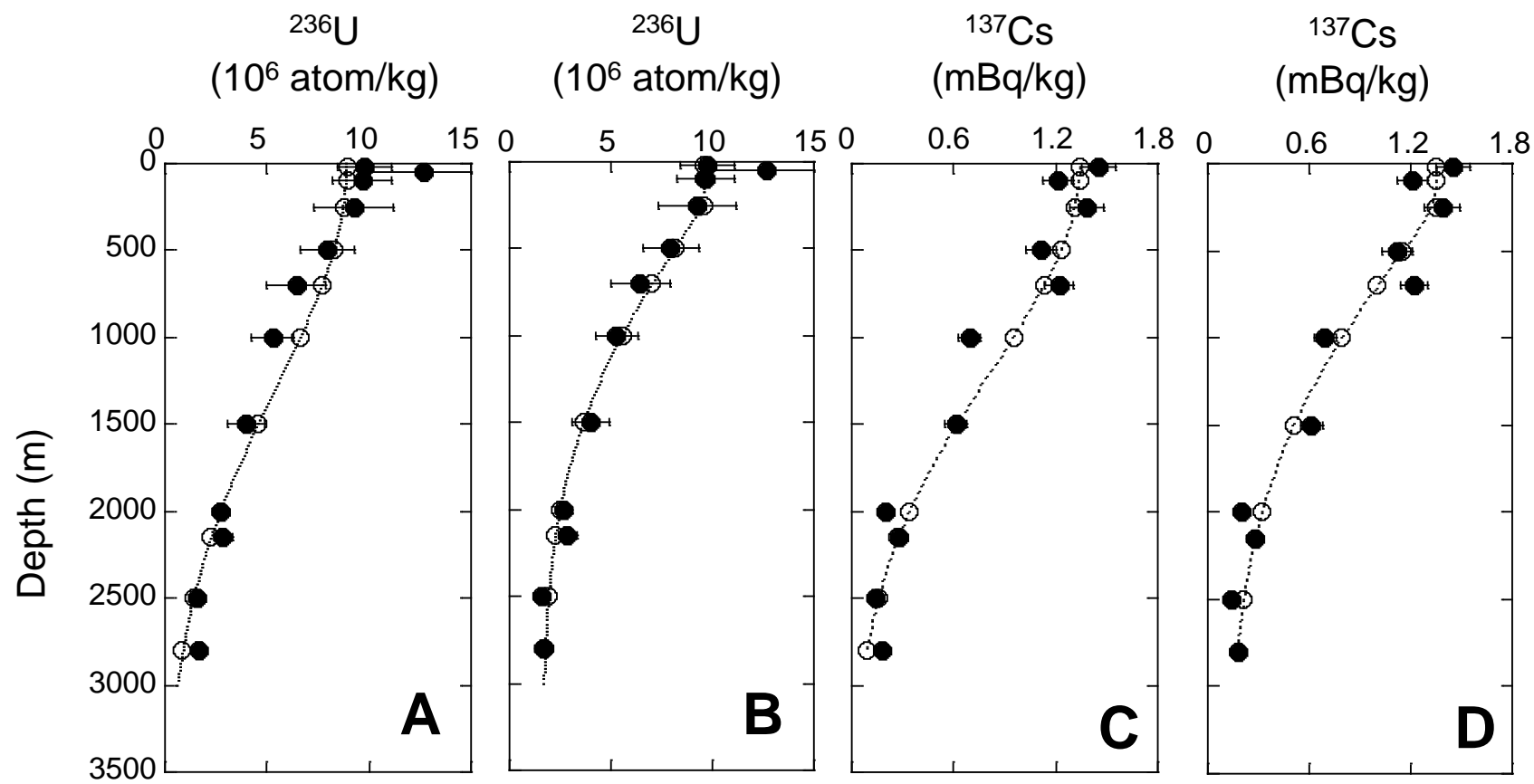


Fig. 4

Figure5

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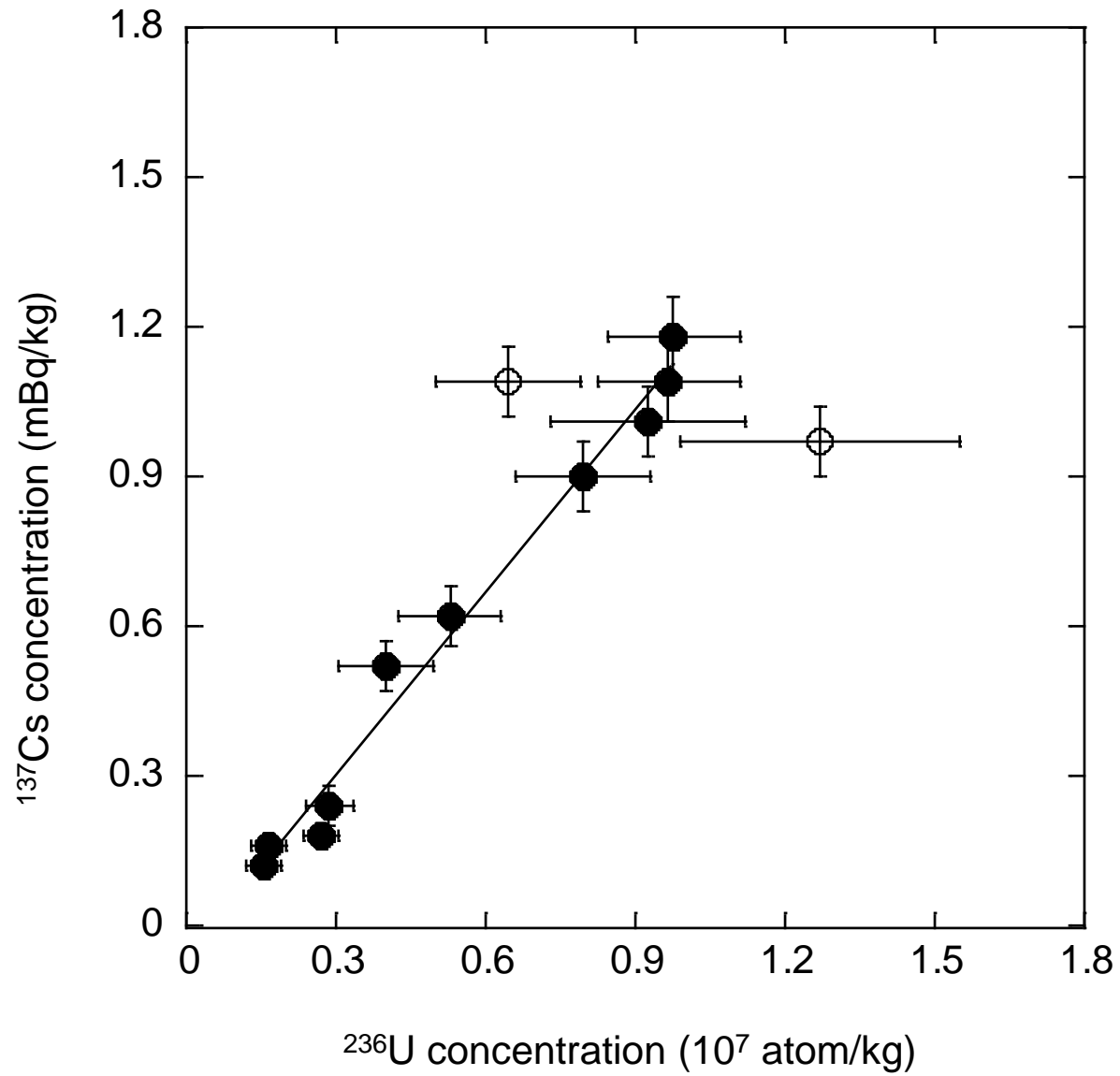


Fig. 5

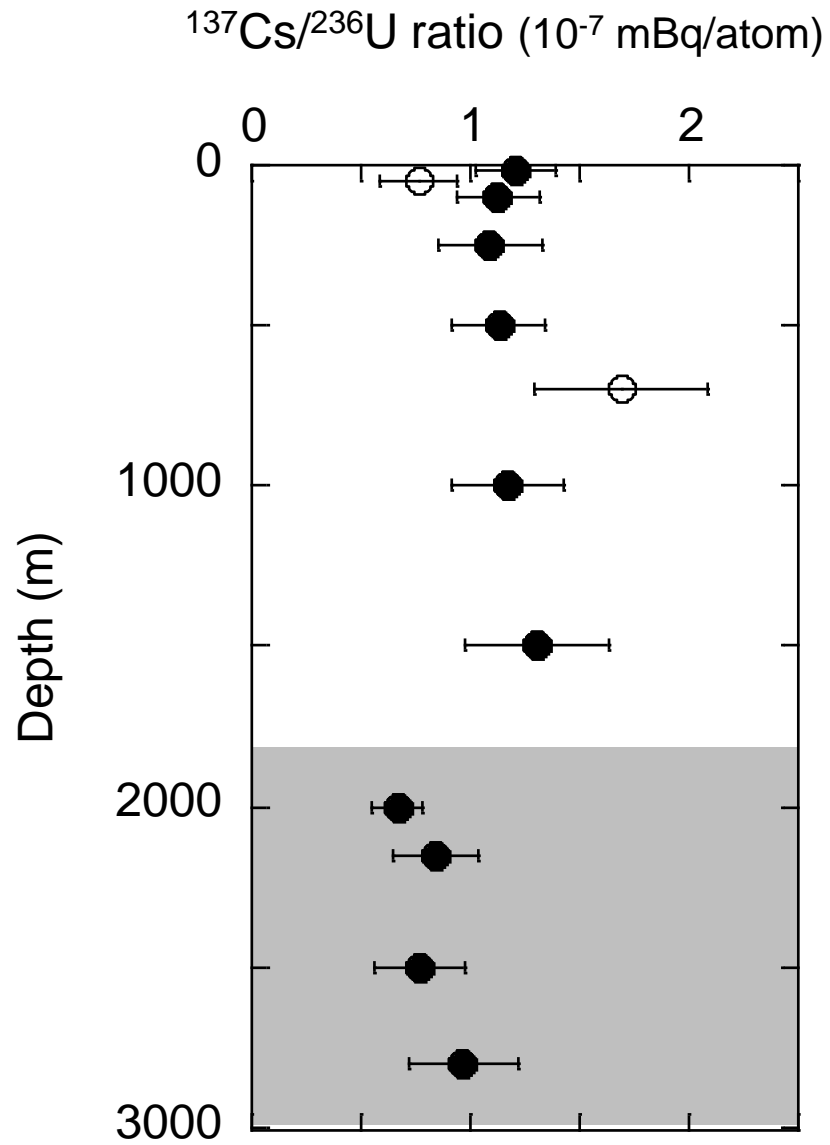


Fig. 6

Figure captions

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.

Fig. 2. Depth profiles of $^{236}\text{U}/^{238}\text{U}$ 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 ($^{\circ}\text{C}$), salinity (psu), pH (SWS scale 25°C) and dissolved oxygen (ml/l) data are also shown.

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. 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 (atoms/kg) and ^{137}Cs (mBq/kg) in sea water, while the open circles and the broken line represent the fit.

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. 6. Depth profile for $^{137}\text{Cs}/^{236}\text{U}$ (10^{-7} mBq/atom) in the water column at station CR58. Error bars represent one standard deviation.

Table1

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Table 1 The results of $^{236}\text{U}/^{238}\text{U}$ 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	depth m	Water				depth range m	^{236}U inventory 10^{12} atom/m ²	^{137}Cs inventory Bq/m ²	SS
		$^{236}\text{U}/^{238}\text{U}$ 10^{-9} atom ratio	^{238}U conc. 10^{15} atom/kg	^{236}U conc. 10^6 atom/kg	^{137}Cs conc. mBq/kg				^{236}U conc. 10^4 atom/kg-water
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.

Table2

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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	anal. sediments	dry bulk sediments	$^{236}\text{U}/^{238}\text{U}$	^{236}U conc.	^{236}U inventory	^{137}Cs conc.	^{137}Cs inventory
cm	g	g	10^{-9} atom ratio	10^7 atom/g	10^{10} atom/m ²	Bq/g	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 ± 0.000 ₈	5.53 ± 0.69
3-4	3.91	10.75	2.87 ± 0.61	1.67 ± 0.37	2.82 ± 0.63	0.004 ± 0.000 ₇	4.35 ± 0.76
4-5	3.96	11.69	0.55 ± 0.16	0.36 ± 0.11	0.66 ± 0.20	0.001 ± 0.000 ₆	0.93 ± 0.70
total					15.0 ± 1.3		36.8 ± 1.9