

## RESEARCH ARTICLE

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Temporal and vertical distributions of anthropogenic  $^{236}\text{U}$  in the Japan Sea using a coral core and seawater samplesAya Sakaguchi<sup>1,2</sup>, Tomoya Nomura<sup>2</sup>, Peter Steier<sup>3</sup>, Robin Golser<sup>3</sup>, Keiichi Sasaki<sup>4</sup>,  
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## Key Points:

- The input history of  $^{236}\text{U}$  to the surface water of the Japan Sea was reconstructed
- The depth profiles of  $^{236}\text{U}$  in the water column of the Japan Sea could be simulated
- Artificial nuclide  $^{236}\text{U}$  has great potential as a world oceanic circulation tracer

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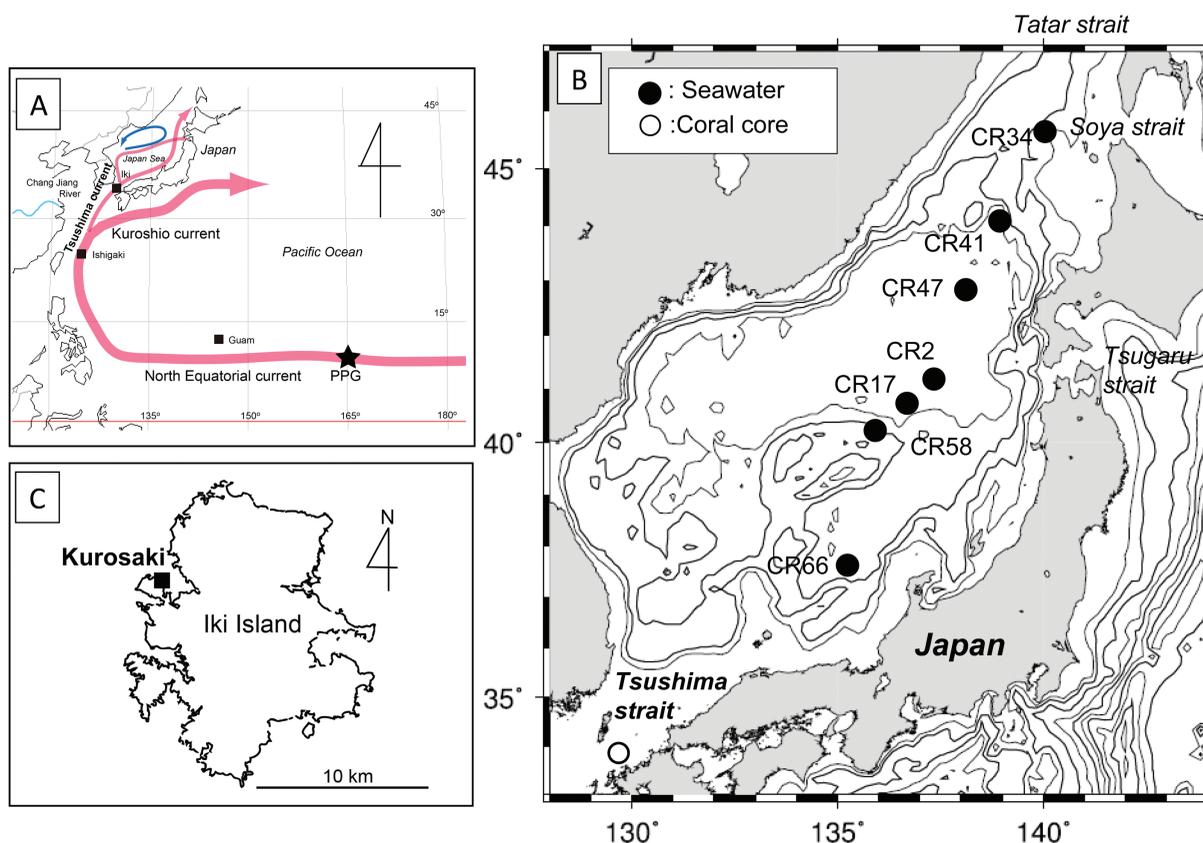
**Abstract** The input history of  $^{236}\text{U}$  to the surface water of the Japan Sea was reconstructed through measurement of the  $^{236}\text{U}/^{238}\text{U}$  atom ratio in annual bands of a coral skeleton which was collected at Iki Island in the Tsushima Strait, the main entrance to the Japan Sea. The  $^{236}\text{U}/^{238}\text{U}$  atom ratios and concentrations of U isotopes were measured for the period 1935–2010 using AMS and ICP-MS. The  $^{236}\text{U}/^{238}\text{U}$  atom ratios revealed three prominent peaks:  $4.51 \times 10^{-9}$  in 1955,  $6.15 \times 10^{-9}$  in 1959 and  $4.14 \times 10^{-9}$  in 1963; thereafter the isotope ratios gradually decreased over the next several decades, attaining a value of  $ca. 1.3 \times 10^{-9}$  for the present day. A simplified depth profile model for  $^{236}\text{U}$  in the Japan Sea, using the reconstructed  $^{236}\text{U}$  value for the surface water together with observed depth profiles for  $^{236}\text{U}$  in the water column in 2010, yielded diffusion coefficients of 3.4–5.6  $\text{cm}^2/\text{s}$  for 6 sampling points. The diffusion coefficient values obtained for the northern stations were relatively large, and fitting uncertainty was also larger for stations in the northern region. It may be presumed that the distribution of  $^{236}\text{U}$  in the water columns have been influenced not only by diffusion but also by subduction of the surface water in the Japan Sea.

## 1. Introduction

Due to recent improvements in instrumentation and chemical pretreatments,  $^{236}\text{U}$  ( $T_{1/2} = 2.342 \times 10^7$  y,  $\lambda_{236} = 2.960 \times 10^{-8} \text{ y}^{-1}$ ) is increasingly being used as an oceanic circulation tracer instead of the short half-life artificial nuclide,  $^{137}\text{Cs}$  [e.g., Christl *et al.*, 2012; Winkler *et al.*, 2012; Casacuberta *et al.*, 2014]. The dominant sources for anthropogenic U input to the North Sea and the North Atlantic Ocean are considered to be the direct and ongoing discharges from the nuclear reprocessing plants at Sellafield (UK) and La Hague (France) [Casacuberta *et al.*, 2014]. In the case of other regions, the maximum input to the surface environment happened in the early 1960s as a result of atmospheric weapons testing as revealed by the analysis of coral samples in the Caribbean Sea [Winkler *et al.*, 2012]. Thus,  $^{236}\text{U}$  can serve as an oceanic tracer from which valuable information can be obtained, concerning the origin, input history and amount of this nuclide in the ocean.

The Japan Sea, a marginal sea in the northwest Pacific, which is linked to the Pacific Ocean by four shallow straits of 20–130 m in depth, has a maximum depth of about 3800 m and an average depth of 1700 m. In this deep bowl-like semienclosed sea, there are two main large surface currents: the cold Limann Current and the warm Tsushima Current (Figure 1). In the Japan Sea, very homogeneous water exists at depth of below a few-hundred meters. This water mass, which is called the “Japan Sea Proper Water,” has extremely narrow range of water temperature (0.0–0.6°C) and salinity (34.06–34.08), and is highly oxygenated (0.2–0.23 mM) [e.g., Uda, 1934; Yasui *et al.*, 1967; Gamo *et al.*, 2014]. Convection and formation of deepwater caused by dry and cold winds are reported for the Japan Sea [e.g., Nitani, 1972; Gamo and Horibe, 1983; Sudo, 1986; Gamo, 1999], which is similar to that observed for the global conveyor belt in the world’s oceans [Broecker, 1990]. For these reasons, the Japan Sea, although being a marginal body of water, has been referred as a “Miniature Ocean” [e.g., Gamo *et al.*, 2014], that can serve as a model for the world’s oceans.

Studies are in progress, using  $^{236}\text{U}$ , to clarify deep water circulation/formation in the Japan Sea [Sakaguchi *et al.*, 2012, 2014]. It was found that this conservative nuclide has different depth profiles from that of



**Figure 1.** Map of the North Pacific area. (a) Surface sea currents, North Equatorial current, Kuroshio Current and Tsushima Current. Star symbol shows the Pacific Proving Ground (PPG). (b) Sampling stations for coral core at Iki Island and seawater samples in the Japan Sea. (c) Sampling station of coral care at Kurosaki, Iki Island.

anthropogenic  $^{137}\text{Cs}$ . Furthermore, the vertical distributions of  $^{236}\text{U}$  from some water columns did not show the same simple diffusion profiles that were simulated under the assumption of a single prominent input of  $^{236}\text{U}$  in 1963 from the atmosphere, as happened with  $^{137}\text{Cs}$ . However, the history of  $^{236}\text{U}$  input to the surface water of the Japan Sea is unknown, and therefore accurate/precise distributions of  $^{236}\text{U}$  cannot be simulated for the Japan Sea.

The single supply of surface water to the Japan Sea is the warm Tsushima Current with input from the Pacific Ocean through the Tsushima Strait. This warm current is a part of the Kuroshio Current which originates from the North Equatorial Current. The North Equatorial Current flows north of the equator from east to west; in this region, nuclear test sites of the U.S. army were located (Pacific Proving Ground, PPG, Figure 1). During the period 1946–1958, about 60 nuclear tests including the detonation of hydrogen bombs were conducted at the Bikini and Eniwetok Atolls in the Marshall Islands (total yield: 110 Mt). If the production rate of  $^{236}\text{U}$  due to the reaction  $^{238}\text{U}(n,3n)^{236}\text{U}$  is large, as proposed by Sakaguchi *et al.* [2009],  $^{236}\text{U}$  must have been directly supplied to the Japan Sea through the surface current since the early 1950s before the majority of global fallout occurred in the 1960s.

Corals, distributed at ocean depths of a few meters, retain compositional information on surface seawater and/or environmental history in their carbonate skeleton. Particularly the U isotopic composition of seawater is generally reflected in the skeleton with negligibly small fractionation for the typical precision of the AMS measurements, which is a few percent [e.g., Weyer *et al.*, 2008]. Thus, the U isotopes (only  $^{236}\text{U}$  and  $^{238}\text{U}$  are considered in this work) in each annual ring of the coral skeleton should permit reconstruction of the past U isotopic composition in surface seawater.

In this study, we have focused on the analysis of a coral core from Iki Island in the Tsushima Strait, Japan (Figure 1), in an attempt to reconstruct the input history of  $^{236}\text{U}$  to the surface seawater of the Japan Sea. The distribution of  $^{236}\text{U}$  in the water column is simulated using reconstructed  $^{236}\text{U}$  concentrations in the coral core in order to understand the diffusion/distribution process of  $^{236}\text{U}$  and deep water circulation in the Japan Sea.

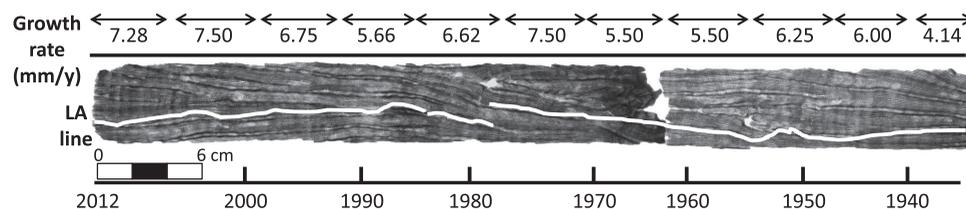


Figure 2. X-ray imaging for coral slab. White line in the image is trace for the analysis of LA-ICP-MS.

## 2. Materials and Methods

### 2.1. Sampling and Sample Pretreatment

The coral core sample (*Dipsastraea speciosa*) was collected in November 2012 from a depth of 3 m at Kurosaki, Iki Island (Figure 1, N33° 48'22.5, E129° 40'02.9). The diameter of the core was 5 cm and the length was 89.5 cm. Iki Island is situated in the Tsushima Strait. This area has been recognized as the northern limit of coral reefs [Yamano *et al.*, 2001, 2012]. This coral sample may reflect chemical information as a direct result of input via the ocean current and global fallout.

The coral slab (thin vertical coral plate), which was 5 mm thick, was subsampled from the centre of the core using a coral cutter installed at Hokkaido University. The annual growth bands were confirmed by X-ray imaging and Sr/Ca analysis with laser ablation (LA)-ICP-MS. The X-ray imaging was conducted according to Nagata *et al.* [2013]. As for the LA-ICP-MS analysis, the instrumentation consisted of a quadrupole ICP-MS unit (Agilent 7500) interfaced to a Nd-YAG laser ablation system (UP-213, New Wave Research) operating at 213 nm [Katsube *et al.*, 2012]. The Sr(<sup>87</sup>Sr)/Ca(<sup>43</sup>Ca) ratios were measured every 500 μm along the coral using a laser beam diameter of 30 μm. The total number of annual rings observed was 78 from the top of the slab to 54 cm inside the core. Consequently, this coral slab (0–53 cm) represented the period from 1935 to present (from the middle of 1934 to the middle of 2012) (Figure 2).

The seawater samples were collected during the cruise KH-10-02 on *Research Vessel Hakuho-maru*, 2010, under the GEOTRACES project (Figure 1). Samples of about 20 kg of seawater were taken with Niskin bottles at different depths at each sampling station. Immediately after sampling, the water was filtered with 0.45 μm pore-size membrane-filters using a Teflon® filtration system. The obtained water fractions were stored in polyethylene containers after adjustment of the pH to 1 by the addition of a small amount of HNO<sub>3</sub>. Details of sampling and pretreatment of seawater samples have been reported by Sakaguchi *et al.* [2012, 2014].

### 2.2. Chemical Treatment of Coral and Seawater Samples

The following procedures were conducted using analytical grade or ultra-pure reagents and with pre-cleaned labware. The coral sections were cut into annual segments in accordance with the X-ray image and the variation of the Sr/Ca ratios. In fact, each segment was cut at the lowest density part between the high density bands to minimize the loss of information, that is, the annual segments were sectioned at the summer zone which has the largest growth rate. Each sample section, which typically weighed between 0.45 and 1.2 g, was homogenized with an agate mortar and pestle. About 0.3–0.5 g of coral was subsampled for the analysis. The sample was put into a beaker with about 20 ml of 4 M HNO<sub>3</sub> (per g of coral) and heated on a hotplate (140°C) for 3 h. During the digestion, 1 ml of hydrogen peroxide (35% w/w) was added every 15 min. After cooling, 0.2 ml of the digest was subsampled for measurement of the <sup>238</sup>U concentration. The residual solution was then evaporated to dryness. After adding a few ml of 10 M HCl, the sample solution was again evaporated to dryness. The residue was dissolved in 20 ml of 10 M HCl on a hotplate (about 100–140°C). The solution was cooled and then passed through a Dowex anion exchange resin (chloride form; 100–200 mesh; column length, 7 cm; column diameter, 8 mm) which was prewashed and activated. The column was washed with 20 ml of 10 M HCl followed by a small amount of 8 M HNO<sub>3</sub> (5 ml×2), and again with about 15 mL of 10 M HCl, to remove matrix and interfering elements. Uranium was then eluted from the resin with 50–80 mL of 2 M HCl. The eluent was dried on a hotplate and evaporated to dryness after adding a few ml of HClO<sub>4</sub> for the decomposition of organic substances originating from the resin and the sample. Concentrated HCl solution (3 ml) was used to dissolve the residue completely. Ultra-pure Fe<sup>3+</sup>

(Kanto Kagaku, AAS standard; 3 ml of 1000 mg/L) was added to the sample solution and diluted to 50 ml with ultra-pure water. A part of the solution was subsampled for determination the chemical yield of U for the sample treatments. Coprecipitation of U with  $\text{Fe}(\text{OH})_3$  was achieved by adding ultra-pure ammonium hydroxide solution. Finally the precipitate was dried in an oven at  $80^\circ\text{C}$  for 12 h. Sputter targets for the AMS measurement were prepared with an iron oxide matrix by calcination of the dried Fe precipitate at  $800^\circ\text{C}$  for 2 h.

The filtered seawater sample was put into a polyethylene vessel (approximately 20 kg). Thirty  $\mu\text{g}$  of an in-house standard which was gravimetrically diluted IRMM-058 to  $4.76 \pm 0.05 \times 10^{10}$  atoms  $^{233}\text{U}/\text{g}$  was added to the sample. The seawater sample was heated for 3 h ( $60\text{--}80^\circ\text{C}$ ) with stirring and then stood for 12 h. Uranium dissolved in seawater was collected by Fe hydroxide coprecipitation (20 mg  $\text{Fe}^{3+}/\text{kg}$ -seawater). The chemical separation (purification) for the U isotope measurement was the same for water and coral samples.

### 2.3. Measurement of U Isotopes

Sample digests and subsample solution were analyzed by ICP-MS (Agilent 7700) after appropriate dilution with 2%  $\text{HNO}_3$  to determine the concentration of  $^{238}\text{U}$ . To reduce and/or correct for matrix effects in ICP-MS measurement, the High-Matrix-Mode of the instrument was used. A mixed solution of In and Bi (each 10  $\mu\text{g}/\text{L}$ ) served as an online-internal standard. It was found that the chemical yield of U following the sample pretreatment was, on average, 85%. In the case of  $^{238}\text{U}$  measurement in the subsampled seawater, the "Ultra-Robust" mode was employed with the samples being analyzed with directly without any chemical treatment.

For the AMS measurement of the isotopic ratio  $^{236}\text{U}/^{238}\text{U}$  in the coral samples, the U oxide samples in the  $\text{Fe}_2\text{O}_3$  matrix were pressed in suitable sample holders and sputtered with a  $\text{Cs}^+$  beam, negative sample ions ( $\text{U}^{16}\text{O}^-$ ) being extracted. After acceleration to 1.7 MeV and fragmentation of the molecular ions in a helium gas cell,  $\text{U}^{3+}$ , with an energy of 6.5 MeV, was separated by high-resolving electric and magnetic sectors before detection of  $^{236}\text{U}^{5+}$  with a gas ionization detector, and measurement of the  $^{238}\text{U}^{3+}$  beam current by a Faraday cup. The use of He and the 3+ charge state provides higher detection efficiency than the previously used method employing Ar and 5+ as described in Sakaguchi *et al.* [2012]; the earlier method was, however still used for the sea water samples described in this work. Full details of AMS measurements have been described by Steier *et al.* [2010], Sakaguchi *et al.* [2012], and Winkler *et al.* [2015]. All AMS measurements of the  $^{236}\text{U}/^{238}\text{U}$  isotopic ratio were performed at the Vienna Environmental Research Accelerator (VERA) facility at the University of Vienna.

## 3. Results and Discussion

### 3.1. Uranium Isotope Variation in Annual Rings From a Coral Sample

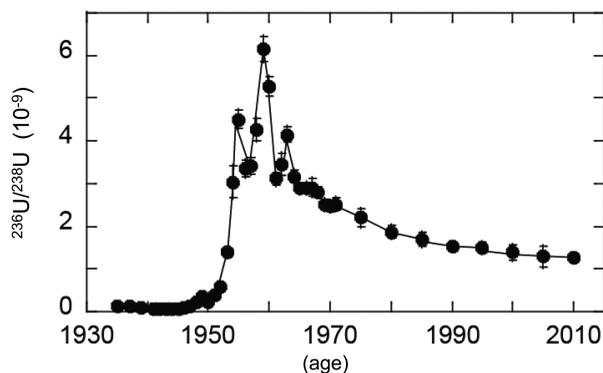
The results for the  $^{236}\text{U}/^{238}\text{U}$  atom ratios and the  $^{238}\text{U}$  and the  $^{236}\text{U}$  concentrations in the 42 coral samples measured are shown in Table 1 and Figure 3. The concentrations of  $^{238}\text{U}$  cover the range of  $1.15 \pm 0.01$  to  $3.29 \pm 0.02$  ppm, and the U concentration factor was estimated to be 400–1000 relative to the surrounding seawater.  $^{236}\text{U}$  was successfully measured in all the annual samples, covering a period from the middle of 1935 to the middle of 2010. The  $^{236}\text{U}/^{238}\text{U}$  atom ratios, which reflect the past ratios in the surface seawater of the Japan Sea, were in the range of  $2.1 \times 10^{-10}$  to  $6.2 \times 10^{-9}$ . These values were greater than those observed for a coral core from the Caribbean Sea, Turneffe Atoll (Belize) [Winkler *et al.*, 2012]. Moreover, we noted a slight elevation in the  $^{236}\text{U}$  values for the preatomic bomb period 1935–1944 compared to values expected for naturally occurring  $^{236}\text{U}$  ( $^{236}\text{U}/^{238}\text{U} < 10^{-13}$ ) [Steier *et al.*, 2008]. This might be due to subsequent adsorption of U into the older parts of coral due to the percolation of seawater into the skeleton. However, this effect can be considered to be negligible when discussing the variation of  $^{236}\text{U}$  input to the Japan Sea in the time period in question.

The variation of  $^{236}\text{U}/^{238}\text{U}$  obtained in this study was totally different from the histories of  $^{236}\text{U}$  input to the North Atlantic Ocean and the Caribbean Sea. The ratios show an abrupt increase in the growth bands from 1953 (from the middle 1952 to the middle 1953) onward, and whose increase is explained by hydrogen bomb testing at the Pacific Proving Ground (PPG). Table 2 lists the nuclear weapons tests performed at the PPG. The following discussion uses information from the web site "Nuclear Weapons Archive" [Sublette,

**Table 1.** Uranium Isotopes in Coral From Iki Island and Estimated  $^{236}\text{U}$  Concentration in Surface Water by Assuming the Surface  $^{238}\text{U}$  Concentration of 3.0 ppb (See Text).

Age (year)	$^{238}\text{U}$ Conc. (ppm)	$^{236}\text{U}/^{238}\text{U}$ ( $\times 10^{-9}$ )	$^{236}\text{U}$ in Seawater ( $10^6$ atom/kg)
1935	2.82 ± 0.04	0.14 ± 0.03	1.05 ± 0.22
1937	2.86 ± 0.01	0.12 ± 0.03	0.92 ± 0.21
1939	2.85 ± 0.01	0.10 ± 0.03	0.74 ± 0.24
1941	2.07 ± 0.02	0.06 ± 0.01	0.48 ± 0.11
1942	2.61 ± 0.03	0.08 ± 0.02	0.61 ± 0.14
1943	2.56 ± 0.01	0.08 ± 0.02	0.59 ± 0.16
1944	2.89 ± 0.03	0.07 ± 0.02	0.54 ± 0.15
1945	2.65 ± 0.07	0.08 ± 0.04	0.62 ± 0.30
1946	2.42 ± 0.05	0.10 ± 0.03	0.72 ± 0.19
1947	2.44 ± 0.02	0.14 ± 0.04	1.04 ± 0.31
1948	1.99 ± 0.03	0.21 ± 0.05	1.63 ± 0.41
1949	2.91 ± 0.04	0.35 ± 0.05	2.64 ± 0.40
1950	2.33 ± 0.01	0.24 ± 0.05	1.84 ± 0.36
1951	2.02 ± 0.04	0.39 ± 0.06	2.95 ± 0.43
1952	2.88 ± 0.02	0.57 ± 0.07	4.36 ± 0.55
1953	2.54 ± 0.03	1.39 ± 0.12	10.6 ± 0.9
1954	1.88 ± 0.01	3.04 ± 0.37	23.0 ± 2.8
1955	2.00 ± 0.02	4.51 ± 0.22	34.2 ± 1.7
1956	2.63 ± 0.03	3.35 ± 0.19	25.5 ± 1.4
1957	3.29 ± 0.02	3.42 ± 0.21	26.0 ± 1.6
1958	1.15 ± 0.01	4.28 ± 0.26	32.5 ± 2.0
1959	2.10 ± 0.02	6.15 ± 0.29	46.7 ± 2.2
1960	3.00 ± 0.03	5.27 ± 0.23	40.0 ± 1.7
1961	2.27 ± 0.02	3.12 ± 0.15	23.7 ± 1.1
1962	2.04 ± 0.01	3.46 ± 0.25	26.3 ± 1.9
1963	2.84 ± 0.05	4.14 ± 0.18	31.4 ± 1.4
1964	3.16 ± 0.02	3.17 ± 0.14	24.1 ± 1.1
1965	3.11 ± 0.04	2.89 ± 0.11	21.9 ± 0.9
1966	3.01 ± 0.05	2.90 ± 0.12	22.0 ± 0.9
1967	2.43 ± 0.00	2.91 ± 0.21	22.1 ± 1.6
1968	3.13 ± 0.08	2.81 ± 0.11	21.4 ± 0.9
1969	2.10 ± 0.01	2.51 ± 0.14	19.1 ± 1.1
1970	3.11 ± 0.03	2.46 ± 0.13	18.7 ± 1.0
1971	3.06 ± 0.02	2.52 ± 0.16	19.1 ± 1.2
1976	3.08 ± 0.02	2.20 ± 0.20	16.7 ± 1.5
1981	3.03 ± 0.01	1.86 ± 0.14	14.1 ± 1.1
1986	2.73 ± 0.02	1.69 ± 0.17	12.8 ± 1.3
1991	2.92 ± 0.01	1.52 ± 0.10	11.5 ± 0.8
1996	2.96 ± 0.03	1.50 ± 0.13	11.4 ± 1.0
2001	2.57 ± 0.01	1.39 ± 0.18	10.5 ± 1.4
2006	3.26 ± 0.03	1.29 ± 0.23	9.80 ± 1.77
2010	2.99 ± 0.01	1.28 ± 0.09	9.72 ± 0.70

2007], which has to be considered with caution, as it is not a traceable scientific publication but seems to compile data released by military intelligence and in-official sources. However, our measurements generally seem to confirm this information. The first fusion bomb test, "Mike" in Operation Ivy which had a yield of several-megaton, was conducted in 1952 at Eniwetok Atoll, Marshall Islands. As a result of the nuclear reaction  $^{238}\text{U}(n,3n)$  occurring in the "tamper" of the device, built of natural U,  $^{236}\text{U}$  was produced rather efficiently and locally/regionally deposited radionuclides were transported into the Japan Sea by the North Equatorial, Kuroshio, and Tsushima Currents. The first prominent isotope ratio maximum,  $4.51 \pm 0.22 \times 10^{-9}$ , was found in the 1955 annual band (from the middle of 1954 to the middle of 1955). This date is exactly 1 year after Operation Castle, which consisted of five tests at megaton yields. In the period immediately following the peak, the U isotopic ratio dropped abruptly, reflecting the fact that uncontaminated seawater was drawn into the area by the North Equatorial Current. The  $^{236}\text{U}/^{238}\text{U}$  ratio increased again in 1958 and achieved a maximum value of  $6.15 \pm 0.29 \times 10^{-9}$  in 1959 (from the middle of 1958 to the middle of 1959), 1 year after a large number of devices were tested at the PPG under operation Hardtack. The above maximum value is about 5 times larger than that in surface water in the Japan Sea at the present time [Sakaguchi et al., 2012, 2014].



**Figure 3.** Variation of  $^{236}\text{U}/^{238}\text{U}$  atom ratios reconstructed from the annual rings in the coral core from Iki Island.

The surface velocity of the North Equatorial current varies between about 18 cm/s at 10 to 15°N to 5 cm/s to the north of the band and 7 cm/s to the south of the band [Yamaoka, 2008]. The velocity of the Kuroshio Current is much faster than that of the North Equatorial Current, at about 50–100 cm/s. If an average velocity of about 20 cm/s is assumed from these numbers, it can be estimated that it would take about 1 year for the surface current to travel over the distance from the PPG experimental site to Iki Island, about 7,000 km, in the Northwest Pacific.

The third small peak for the  $^{236}\text{U}/^{238}\text{U}$  isotope ratio occurred in 1963. This peak has been recognized as representing "global

**Table 2.** List of Nuclear Weapons Tests of the U.S. at Marshall Islands, PPG

Age	Operation Series	Number	Mt <sup>a</sup>		
			Fission	Fusion	Total
1946	Crossroads	2	0.042	0	0.042
1948	Sandstone	3	0.104	0	0.104
1951	Greenhouse	4	0.323	0.075	0.398
1952	Ivy	2	5.9	4.9	10.8
1954	Castle	6	30.8	17.3	48.1
1956	Redwing	17	8.8	12	20.8
1958	Hardtack I	31	11.7	16.3	28.0

<sup>a</sup>TNT equivalent (1 kiloton of TNT = 4.184 TJ).

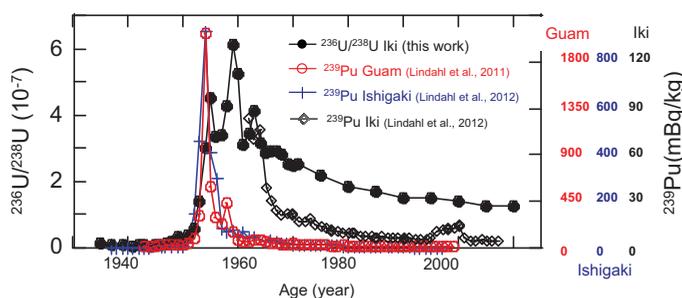
$10^{-9}$ ) as that obtained for Japan Sea surface seawater sampled in 2010 [Sakaguchi *et al.*, 2012]. This supports the assumption that our coral core has preserved information on the U isotopes in the surface seawater.

### 3.2. Input of Uranium and Plutonium Isotopes to the North Pacific Ocean

The U variation in our coral core sample obtained from Iki Island can be compared with the refractory actinide, plutonium (Pu), in coral cores collected from the North Equatorial and Kuroshio Current areas in the North Pacific: Guam, Ishigaki and Iki Islands [Lindahl *et al.*, 2011, 2012].

Figure 4 shows a comparison between U and Pu isotopic variations in coral core samples through the observation period. The  $^{239}\text{Pu}$  concentration decreased with distance from the PPG site, and all cores (except one from Iki Island which starts only 1960 [Lindahl *et al.*, 2012]), showed an abrupt increase in the early 1950s. However, the U and Pu isotopes show markedly different time trends.  $^{236}\text{U}$  in the Iki coral core reached its maximum value in 1959 (the middle of 1958 to the middle of 1959), while  $^{239}\text{Pu}$  in the Guam and Ishigaki cores reached its maximum values in 1954; this is the only one prominent Pu peak throughout the observation period. The different peaks for U and Pu can be explained by that (1) different explosive devices were used between the early and late 1950s, and (2) by the influence from other ocean surface currents and/or discharges from large rivers. Regarding (1), it is said [Sublette, 2007] that the tampers for the thermonuclear devices were changed from natural U to enriched U during this period. Thermonuclear explosions produce  $^{236}\text{U}$  by high energetic (14 MeV) neutron reactions on natural U via the nuclear reaction  $^{238}\text{U}(n,3n)^{236}\text{U}$  ( $\sigma=405$  mb) and  $^{239}\text{Pu}$  by fast and epithermal neutrons via  $^{238}\text{U}(n, \gamma)^{239}\text{U} \rightarrow ^{239}\text{Np} \rightarrow ^{239}\text{Pu}$ . However, if enriched U had been used as tampers,  $^{239}\text{Pu}$  would not have been produced efficiently, leading to a dominant production of  $^{236}\text{U}$  through the  $^{235}\text{U}(n, \gamma)^{236}\text{U}$  reaction. Our data thus supports Sublette [2007]. A possible effect in the context of (2) may be the inflow of the Chang Jiang River. It has been reported that discharges from this river affect the Tsushima Current [e.g., Matsui and Senjyu, 2009; Lindahl *et al.*, 2012]. Actually, there was a severe flood of the Chang Jiang River in 1954, which may have diluted the  $^{236}\text{U}/^{238}\text{U}$  ratio with colloidal natural U that originated from the mud flow from the river and/or from the resuspension of sea sediments in the estuary system. Additionally,  $^{236}\text{U}$  fallout in the catchment area of the river may have been remobilized and transported into the river quickly, while Pu is expected to be strongly bound to soil. Another possible effect was mixing of water from the surface currents introduced from the South China

Sea, although this has not been clarified.



**Figure 4.** Variations of  $^{236}\text{U}/^{238}\text{U}$  atom ratios (this work) and  $^{239}\text{Pu}$  concentrations [Lindahl *et al.*, 2011, 2012] reconstructed from the annual rings in the coral cores from Guam, Ishigaki and Iki Islands.

Following the prominent peaks in the 1950s, the Pu concentration decreases quickly, while  $^{236}\text{U}$  stays elevated much longer and show only a gradually decreased with time. This reflects the different behavior of the elements in seawater, as Pu binds to particles and is quickly scavenged to deeper water layers and bottom sediments, and thus unavailable for corals. Uranium, on the other hand, stays in solution.

A difference between the elements was also found in the 1995–2000 coral bands. The concentrations of  $^{239}\text{Pu}$  increased during this period, while no such increase was observed for  $^{236}\text{U}$ . *Lindahl et al.* [2012] suggested the possibility of a contribution from the Taiwan Warm Current, which was influenced by the Chang Jiang River. Assuming that this hypothesis is correct, resuspension of offshore bottom sediments with the outflow from this river could be conceivable. These bottom sediments in the East China Sea should contain Pu predominantly after starting the nuclear tests [*Liu et al.*, 2011].

The strong fingerprint of the PPG tests observed in the Pu in coral cores suggests that they were responsible for the larger part of the Pu input to the Northwest Pacific area. This is, however, in contradiction to data from the sea bottom sediments analyzed by *Zheng et al.* [2005], which show that the contribution of PPG derived Pu to Japans Sea sediments was only about 20%, while the other 80% came from global fallout. As *Buesseler* [1997] reported, the solubility of nuclides is also related to their physical and chemical form as local, regional, or global fallout aerosols or particles. Thus, it is necessary to be careful to use anthropogenic radionuclides as tracers for environmental dynamics, especially for particle-reactive nuclides.

### 3.3. The Apparent Half Residence Time of $^{236}\text{U}$ in Surface Water Since 1970

A gradual decrease in the  $^{236}\text{U}/^{238}\text{U}$  ratio has been observed since the early 1970s in the coral core samples from Iki Island. A likely explanation for this is that diffusion of  $^{236}\text{U}$  to the deeper waters of the ocean has occurred. Applying the equation from *Miyao et al.* [1998] for conservative radionuclides injected into surface sea water, our data yield an apparent half-residence time ( $T_{\text{app}}$ ) of  $^{236}\text{U}$  in surface water from the early 1970s to present ( $\lambda_{236} + \lambda_{\text{app}} = 0.0167$ ,  $T_{\text{app}} = 40.3$  years). This value is less than half of that obtained for the Caribbean Sea, which has been estimated to be about 100 years, using reconstructed  $^{236}\text{U}$  values by *Winkler et al.* [2012]. Several reasons may be responsible for the difference in the apparent half-residence time: (1) a difference in the vertical mixing of surface waters, which can be reworded as a difference in the diffusion coefficients, (2) horizontal mixing with surface currents originating from different regions, and (3) a continuous supply from a source such as erosion/weathering of contaminated surface soil.

The half-residence time of 40 years for  $^{236}\text{U}$  in the surface water of the Japan Sea is about 2.5 times longer than that for the Pu isotopes (16 years), which was calculated using the  $^{239}\text{Pu}$  concentration, except for an anomalous signal from 1995 to 2000, which was taken from a coral core obtained from the same island [*Lindahl et al.*, 2012]. Furthermore, the apparent half-residence time of  $^{236}\text{U}$  was also longer than that of  $^{137}\text{Cs}$  which was estimated to be 16 years, excluding the Chernobyl effect, by *Miyao et al.* [1998], despite  $^{137}\text{Cs}$  is generally recognized as a conservative element, which gives good service as an oceanic circulation tracer. Cs is rather more particle reactive than U. Actually, the scavenging ratio of  $^{137}\text{Cs}$ , which was defined as the inventory ratio of  $^{137}\text{Cs}$  between sediments and water column, was larger (1/40) than that of  $^{236}\text{U}$  (1/100) [*Sakaguchi et al.*, 2012]. Alternatively, it is possible to specify a different half-residence time due to a difference in the dominant source of each nuclide. The maximum input of  $^{137}\text{Cs}$  to the surface land of Japan occurred in 1963 based on inspection of monthly precipitation data for this nuclide [*Meteorological Research Institute (MRI)*, 2011]. *Zheng et al.* [2004] also showed a maximum of  $^{137}\text{Cs}$  in 1963 in sediment cores obtained from the Northwest Pacific, in the vicinity of Japan. Thus, it may be the case that the history of  $^{137}\text{Cs}$  input to surface seawater is the same as that for atmospheric deposition, and the depth profiles and apparent half-residence time of this nuclide in surface water may be different from  $^{236}\text{U}$ . In any case, these three isotopes (U, Pu, Cs) show significant differences in their behavior and origin.

### 3.4. Possible Application of $^{236}\text{U}$ in Coral Samples

In oceanography, U has been recognized as a conservative element. The  $^{238}\text{U}$  concentration in seawater is relatively constant throughout the world, and is about 3 ppb [e.g., *Ku et al.*, 1977; *Nozaki*, 2001]. Using this value for  $^{238}\text{U}$ , the concentration of  $^{236}\text{U}$  (atoms/kg) in surface water in the Japan Sea may be estimated for the period 1935–2010. The calculated concentration of  $^{236}\text{U}$  (atom/kg) in surface water is shown in Table 1.

As shown in section 3.1 for the reconstructed  $^{236}\text{U}/^{238}\text{U}$  ratios ( $^{236}\text{U}$  concentration in surface water), the origin of  $^{236}\text{U}$  in the Japan Sea is two-fold, namely inflow from the PPG and deposition as global fallout. Assuming that the  $^{236}\text{U}$  supplied from these sources diffuses downward gradually to greater depths, the ideal depth profile for  $^{236}\text{U}$  in the Japan Sea can be calculated using the finite volume method (FVM). Generally, the vertical distribution of a conservative nuclide in the ocean can be simulated by diffusion in a first approximation, because vertical advection is a negligibly small phenomenon compared with horizontal

advection. We employ a simple FVM model, because the system is relatively simple and the physical parameters can be retained. The seawater depth from surface to the bottom was divided into 150 finite volumes (FVs) and into 30 FVs for site CR34 that bottom depth is less than 1000 m. Considering the actual data resolution (about a few tens meters) for surface seawater and an average of bottom depths (3000–4000 m) among the water sampling stations, this number of FVs is appropriate for our simulation although it is three times larger than that of the modelling from *Tsumune et al.* [2001]. The concentration of  $^{236}\text{U}$  in the  $j$ -th FV at time  $t + \delta t$  was calculated as:

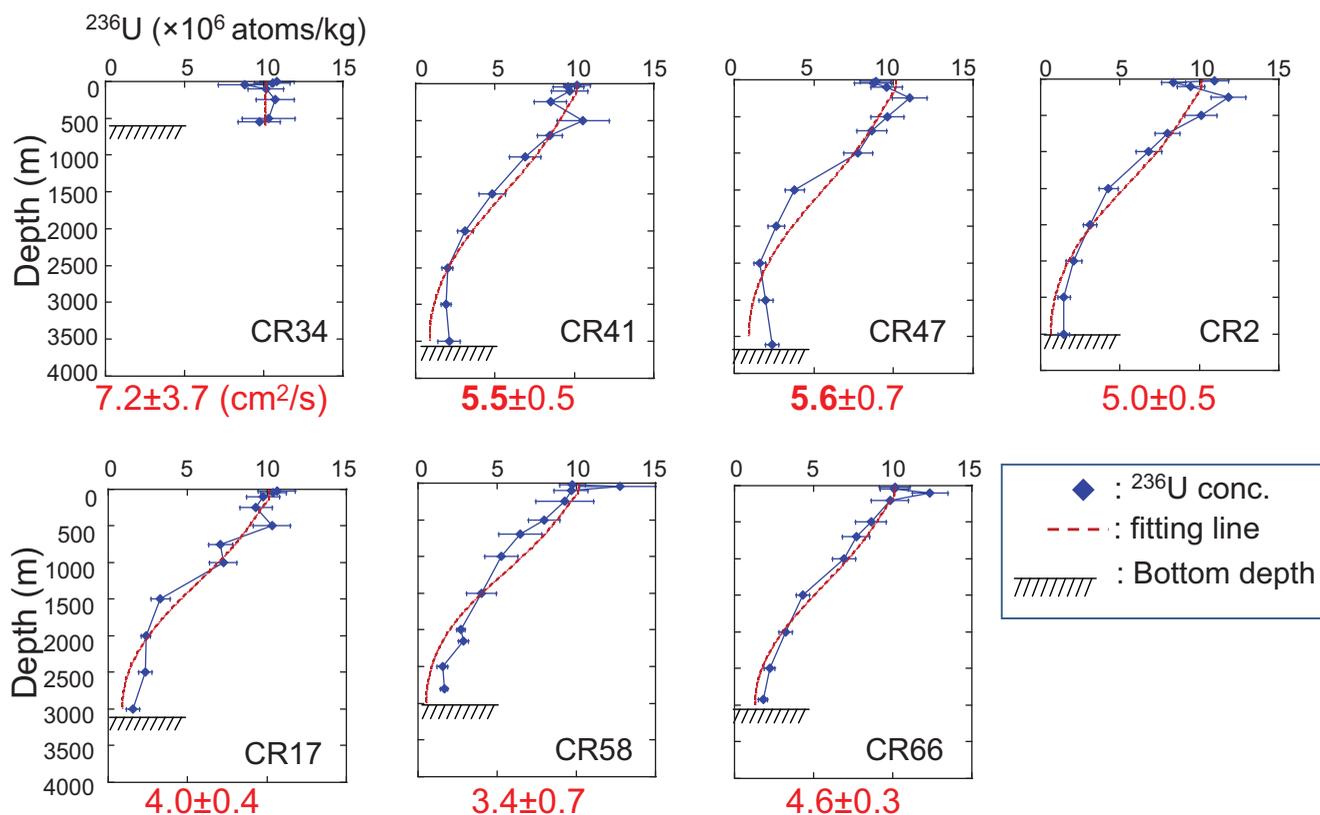
$$C_j(t + \delta t) = C_j(t) + \delta t \left[ \left\{ \kappa_{j+1/2} (C_{j+1}(t) - C_j(t)) - \kappa_{j-1/2} (C_j(t) - C_{j-1}(t)) \right\} / \delta z^2 - \lambda_{236} C_j(t) \right] \quad (1)$$

where  $C_j(t)$  is the concentration of  $^{236}\text{U}$  (atoms/kg) in the  $j$ -th FV ( $j = 1-150$ ) at the time  $t$ ,  $\delta t$  is the time-step interval (s),  $\kappa_{j+1/2}$  is the vertical diffusion coefficient ( $\text{cm}^2/\text{s}$ ) for turbulent flow between the  $j$ - and  $(j+1)$ -th FVs, and  $\lambda_{236}$  is the decay constant for  $^{236}\text{U}$  (1/s). The decay term is negligible because of the small decay constant of  $^{236}\text{U}$  compared with the observation time. The surface layer was modeled as a 150 m-deep section in which is  $^{236}\text{U}$  mixed well, assuming a constant concentration obtained from the  $^{236}\text{U}$  data (Table 1 and Figure 3) due to immediate diffusion. The value in each time step was estimated from the quadratic interpolation of successive measurement points in the coral core. The bottom boundary was set to be impermeable ( $dC/dz = 0$ ) as  $^{236}\text{U}$  did not diffuse into the sediments. Sixteen thousand steps encompassing 75 years (1935–2010) were calculated according to equation (1) assuming that the initial  $^{236}\text{U}$  concentration was zero. On the assumption that the diffusion coefficient ( $\kappa$ ) is constant with depth, the diffusion coefficients were automatically determined to fit the computed depth profiles of  $^{236}\text{U}$  concentration for the measured  $^{236}\text{U}$  values at each sampling station (Figure 1) using a least squares method. For this fitting, the concentrations of  $^{236}\text{U}$  at the sampling depth were calculated using a linear interpolation between two nearest centre points of the FV.

To evaluate propagation of the measurement error to the value of  $\kappa$ , we performed the least-square fitting for 100 synthesized data sets including random noise of normal distribution for each sampling sites. The  $\kappa$  values obtained from the fitting embrace about 10% standard deviation and the averages converge within 1%. This indicates that the errors in the measurements does not significantly influence the value of  $\kappa$ .

The depth profiles together with diffusion coefficients at each location for  $^{236}\text{U}$  in the Japan Sea were constructed and shown in Figure 5. The constructed profiles were found to fit well to the vertical distributions of  $^{236}\text{U}$  measured in each water column. This means, the distribution has been basically controlled by diffusion in the Japan Sea. In other words, the Japan Sea proper water is quite stable, and a prominent subduction of surface water was not found. The diffusion coefficients of  $^{236}\text{U}$  for the Japan Sea ranged from 3.4 to 5.6  $\text{cm}^2/\text{s}$  except for location CR34 which was close to the Tatar Channel. These values were in the same range as those obtained using Ra isotope data (6  $\text{cm}^2/\text{s}$ ) [*Tanaka et al.*, 2006] and  $^{137}\text{Cs}$  isotope data (1–10  $\text{cm}^2/\text{s}$ ) [*Tsumune et al.*, 1999, 2001]. However, the values are one order of magnitude larger than the diffusion coefficient, which was calculated using  $^{236}\text{U}$  values measured by *Winkler et al.* [2012] in a coral core, at the Caribbean Sea. Diffusion coefficients of  $^{236}\text{U}$  are closely related to the half-residence time of this nuclide in surface water, as shown in the first section of 3.3. In order to clarify the reason for difference in diffusion coefficients between the two seas, the  $^{236}\text{U}$  vertical and horizontal profiles in the Caribbean Sea would be needed.

The diffusion coefficients in the northern region (CR 41 and 47) of the Japan Sea gave large values in comparison to those for the southern stations. In addition, the simulated depth profiles in the northern section did not fit as well as that of the southern station (CR66). These phenomena have also been noted for  $^{137}\text{Cs}$  [*Sakaguchi et al.*, 2014]. This might be simply explained by active vertical mixing and/or deep water formation with subduction of surface water to deeper layers, though  $^{236}\text{U}$  depth profiles can be basically explained by diffusion as mentioned above. As for the possibility of active vertical mixing of water in the Japan Sea, *Talley et al.* [2006] reported that the dissolved oxygen concentration in deep water of the Japan Sea is much higher than anywhere else in the Pacific, and concluded that the Japan Sea stands out as a region of high ventilation/mixing rate. If our results also reflected the effect of active vertical mixing, the simulation must be modified including modification parameters for advection and/or turbulent flow for the vertical direction. The circulation of seawater of the Japan Sea will be discussed in more detail in a later publication using  $^{137}\text{Cs}$  and  $^{236}\text{U}$  as water circulation tracers, considering also turbulent flow.



**Figure 5.** Concentrations of dissolved  $^{236}\text{U}$  (atom/kg) in the water column as a function of depth. Error bars are one standard deviation. Dashed line shows the simulated profile with FVM using reconstructed  $^{236}\text{U}$  values as the input parameter for the surface water. Diffusion coefficient is also given for each station.

#### 4. Conclusion

Uranium isotopes ( $^{238}\text{U}$  and  $^{236}\text{U}$ ) in the annual rings of a coral sample from Iki Island were measured with AMS after appropriate sample pretreatment. The analyzed values reflected the recent history (ca. 1935–2010) of isotopic composition for U in surface waters of the Japan Sea and the Northwest Pacific Ocean. The specific features of the variation of  $^{236}\text{U}/^{238}\text{U}$  obtained for the coral core were: (1) the ratio increased abruptly from the early 1950s (1953), (2) three prominent peaks were observed over the decade beginning in the early 1950s, and (3) the isotope ratio decreased gradually following the third peak in 1963. The recorded variations in isotope ratio reflect and elucidate the nuclear testing at the PPG, and include global stratospheric fallout. The  $^{236}\text{U}$  variation is different from that of Pu which has been reported for the Northwest Pacific by Lindahl *et al.* [2011, 2012]. The depth profiles of  $^{236}\text{U}$  in the water column of the Japan Sea could be simulated using the reconstructed values for  $^{236}\text{U}$  in the surface seawater. However, the depth profiles for  $^{236}\text{U}$  concentrations obtained from the northern area of the Japan Sea were not in complete agreement with those obtained by the simulation. Furthermore, the diffusion coefficients obtained from the northern stations were greater than that of the southern station and this may be due to the active vertical mixing of seawater and/or enhancement of the  $^{236}\text{U}$  concentration in the bottom water by the inflow of surface water with deep water formation.

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### Erratum

In the originally published version of this article, the fourth author's name was presented incorrectly as Robin Golser. This has been corrected to read Robin Golser, and this version may be considered the authoritative version of record.