Iodine-129 concentration in seawater near Fukushima before and after the accident at the Fukushima Daiichi Nuclear Power Plant

T. Suzuki 1,\*, S. Otosaka 1, J. Kuwabara 2, H. Kawamura1, T. Kobayashi1

[1] Japan Atomic Energy Agency, Ibaraki, Japan

[2] Japan Atomic Energy Agency, Aomori, Japan

Correspondence to: T. Suzuki (suzuki.takashi58@jaea.go.jp)

Abstract

Anthropogenic radionuclides were released into the environment in large quantities by the Fukushima Daiichi Nuclear Power Plant (1FNPP) accident. To evaluate accident-derived 129I, the 129I concentrations in seawater before and after the accident were compared.

Before the accident (2008–2009), the 129I concentrations in the western margin of the North Pacific between 32 °N and 44 °N showed a latitudinal gradient that was expressed as a linear function of latitude. The highest and average 129I concentrations after the accident were 73 times and approximately 8 times, respectively, higher than those before the accident in this study area. Considering the distribution of 129I in surface seawater, the accident-derived 129I in the southern and northern stations of the 1FNPP was predominantly supplied by seawater advection and atmospheric deposition including microbial volatilization, respectively.

As of October 2011, depth profiles of 129I revealed that 129I originating from the 1FNPP existed mainly in the upper 100 m depth. From the depth profiles, the cumulative inventories of accident-derived 129I were estimated to be (1.6–9.6) × 1012 atoms/m2 in this study area.

On the basis of the 129I data in the seawater near Fukushima, the effective dose of 129I from seafood ingestion was much smaller than the annual dose limit.

# Introduction

Significant fission products such as 134Cs (half-life: 2.06 years), 137Cs (half-life: 30.2 years), and 131I (half-life: 8.02 days) were released into the environment by the Fukushima Daiichi Nuclear Power Plant (1FNPP) accident caused by the Great East Japan earthquake and tsunami on Mar 11, 2011 (Honda et al., 2012; Momoshima et al., 2012). From the viewpoint of environmental safety with respect to nuclear accidents, 134Cs, 137Cs, and 131I are regarded as important radionuclides because of their high fission yield and high γ-ray energy.

Radioiodine can enter the human body via food and drinking water and then selectively accumulates in the thyroid, creating an irradiation risk. A significantly higher incidence of thyroid cancer in children living around the Chernobyl accident site has been reported (Baverstock et al., 1992). Although thyroid cancer related to 131I exposure as a result of nuclear accidents has been of considerable concern, less attention has been paid to the increasing inventory of 129I (half-life: 1.57 × 107 years) from the 1FNPP accident. Because 129I has a long half-life, the amount and behavior of accident-derived 129I in the environment should be investigated to address concerns about the radiological impacts to future generations. Measuring 129I in environmental samples also offers an opportunity to retrospectively analyze the migration of accident-derived 131I, which has decayed and become undetectable from the passage of post-accident time.

Anthropogenic 129I is released into the environment from nuclear fuel reprocessing plants and is then transported mainly to the northern hemisphere (Suzuki et al., 2010; Snyder et al., 2010; Toyama et al., 2012). Thus, the 129I level in the environment is already increased from its natural level. Soil and seawater play a role as a reservoir of 129I (Muramatsu et al., 2004). The impact of 129I on land from the 1FNPP accident on land has been reported (Miyake et al., 2012), but not the impact on the ocean. This study focuses on the impact of 129I on seawater from the 1FNPP accident. To precisely evaluate the increase of 129I caused by the accident, information about the 129I before the accident in the western North Pacific Ocean is crucial. Many studies about 129I in seawater have been conducted around nuclear fuel reprocessing plants (Keogh et al., 2007; Raisbeck and Yiou, 1999; Alfimov et al., 2004; Michel et al., 2012; Hou et al., 2001), but only a few studies have been undertaken in the Pacific Ocean (Suzuki et al., 2010; Povinec et al., 2010). Therefore, the background level of 129I in the western North Pacific Ocean had not previously been determined.

Prior to the 1FNPP accident, a new nuclear fuel reprocessing plant was tested for routine operation at Rokkasho, Japan. Anticipating the release of 129I from the operation of the reprocessing plant, seawater samples in the western North Pacific Ocean were collected to clearly define the background level before plant operations commenced. After the 1FNPP accident, seawater samples collected before the accident have proven useful for evaluating the background level of 129I in the western North Pacific Ocean. In this paper, we report the distributions of 129I in the western North Pacific before and after the 1FNPP accident and evaluate the accident-derived 129I in seawater. In addition, we infer the migration of 129I off Fukushima after the accident and assess the internal dose of accident-related 129I as a result of seafood consumption.

# Experimental

## Seawater sampling

Seawater sampling before the 1FNPP accident were conducted at three stations: KNOT (154° 58′ E, 43° 58′ N) on May 10, 2008 by the T/S Oshoro Maru; off Kii (135° 31′ E, 32° 28′ N) on Aug 5, 2008 by the R/V Soyo Maru; Joban C (142° 13′ E, 36° 48′ N) on Jul 18, 2009 by the R/V Soyo Maru; and Miyako (145° 00′ E, 40° 00′ N) on Jul 20, 2009 by the R/V Soyo Maru, as shown in Fig. 1 a). Fig. 1 a) includes another station that can provide a background level for 1FNPP in the western North Pacific Ocean (Suzuki et al., 2010).

Seawater sampling after the 1FNPP accident were undertaken by four expeditions: from Apr 27 to May 1, 2011 by the R/V Tansei-Maru, cruise KT-11-06 (Fig. 1 b); from Jun 12 to 13, 2011 by the Bosei-Maru, cruise BO-11-05 (Fig. 1 c); from Aug 2 to 3, 2011 by the Hakuhou-Maru, cruise KH-11-07 (Fig. 1 d); and on Oct 29, 2011 by the Tansei-Maru, cruise KT-11-27 (Fig. 1 e). Seawater samples at eight stations were collected from surface to deep layers at stations A, B, and C by cruise KT-11-06; at stations FS1, ES2, and FS2 by cruise KH-11-07; and at stations ST08 and K8 by cruise KT-11-27. Fourteen surface seawater samples were collected at other stations.

## Analytical procedure

Iodine was extracted from seawater samples by the solvent extraction technique (Suzuki et al., 2008). Ascorbic acid with hydrochloric acid was added to a seawater sample to reduce iodate to iodide after passing through a 0.45μm filter. Iodide was oxidized to molecular iodine by the addition of sodium nitrite for the extraction to chloroform. Back extraction was carried out from chloroform using sodium sulfite by reducing molecular iodine to iodide. The extracted iodide was precipitated as silver iodide by silver nitrate. The silver iodide sample was washed by nitric acid and pure water and then loaded to a copper target holder.

Iodine isotopic ratios were measured by accelerator mass spectrometry at the Aomori Research and Development Center of the Japan Atomic Energy Agency. All measured data was normalized to the standard reference material having 129I/127I = (9.85 ± 0.12) × 10−13, which was obtained from the National Institute of Standards and Technology (NIST SRM 3230) (Suzuki et al., 2006).

Total iodine (iodate + iodide) concentrations in seawater samples were measured by cathodic stripping square wave voltammetry or quadrupole inductively coupled plasma mass spectrometry (Campos, 1997).

# Results and discussion

## Distribution of 129I in surface seawater

### Before the 1FNPP accident

With regard to the global distribution of 129I in surface reservoirs, the latitudinal distribution provides us with information useful for understanding the fate of 129I (Snyder et al., 2010). Thus, the 129I concentrations in surface seawater before and after the 1FNPP accident with another published data (Hou et al., 2013) are plotted in Fig. 2 against the latitude of the sampling location. The surface 129I concentrations are also plotted at the sampling locations in Fig. 1.

The concentration of 129I before the 1FNPP accident was in the 0.94 (off Kii) – 1.83 (offshore of Kushiro: Suzuki et al., 2010) × 107 atoms/L range (Table 1). The 129I in surface seawater between 32 °N and 44 °N in the western North Pacific Ocean before the 1FNPP accident was high in the north and decreased with decreasing latitude. These concentrations correspond to the locations of nuclear fuel reprocessing plants, which is a primary source of 129I in the environment. Since major reprocessing plants such as Sellafield (54°00′ N) in the United Kingdom, La Hague (49°30′ N) in France, and Hanford (46°37′ N) in the United States are located in the middle- to high-latitude regions of the Northern Hemisphere, 129I originating from nuclear fuel reprocessing plants was mainly distributed over the Northern Hemisphere (Snyder et al., 2010; Suzuki et al., 2010; Moran et al., 1999). The latitudinal distribution of 129I in surface seawater before the 1FNPP accident is attributed to the atmospheric deposition of 129I originating from the major reprocessing plants and can be expressed as a linear function of latitude (Fig. 2).

*129CBG* = 0.080 × *L* − 1.7, (32 < L < 44)

where *129CBG* (107 atoms/L) is the 129I concentration in surface seawater before the 1FNPP accident and *L* (°N) is the latitude.

### After the 1FNPP accident

In this study area, the 129I concentrations in the surface seawater after the 1FNPP accident were in the 1.08–89.8 × 107 atoms/L (corresponding to 129I/127I = 4.47–362 × 10-11) range (Figs. 1 and 2). The highest 129I concentration (129I = 89.8 × 107 atoms/L, 129I/127I = 362 × 10-11) was observed at station UW2 on Jun 12, 2011 during cruise BO-11-05, this value was 73 times higher than that before the accident, and this value was higher than that of the previous study (Hou et al., 2013). The average 129I concentration in surface seawater after the 1FNPP accident was 9.3 × 107 atoms/L, a value approximately 8 times higher than that before the accident. Since routine operation of the Tokai reprocessing plant and test operation of the Rokkasho reprocessing plant had both ceased in 2008, the elevated 129I can be considered to originate from 1FNPP. Accident-derived 129I was detected at most stations after April 2011 except for two stations: station 8 during cruise KT-11-06 (28 Apr 2011) and station UW1 during cruise BO-11-05 (12 Jun 2011). Station 8 (141°50′ E, 36°31′N) is located near station UW1 (141°53′ E, 36°00′ N). In the coastal regions, several water masses, such as the Kuroshio, meso-scale eddies associated with the Kuroshio, and fresh water from the land, coexist in a complex fashion. The satellite images (Ibaraki Prefectural Fisheries Experimental Station, 2012) of sea surface temperature indicate that the surface of station 8 at the time of observation was dominated by meso-scale eddies associated with the Kuroshio that were coming from low contamination regions (Buesseler et al., 2012). The lack of elevated 129I concentrations at these two stations is considered to be caused by the complex seawater currents in the area near Fukushima.

The distribution patterns of surface 131I and 137Cs simulated numerically (Kawamura et al., 2011; Tsumune et al., 2012) showed that the accident-derived radionuclides supplied directly into the ocean were initially advected southward along the coast and then flowed eastward with the Kuroshio and its extension. The numerical simulation results from Mar 25 to May 1, 2011 also showed that the radionuclides were minimally advected northward near Fukushima (Kawamura et al., 2011). On the other hand, atmospheric dispersion simulations indicated that 131I and 137Cs were transported to the northeast and south of the 1FNPP and deposited on the surface of the western North Pacific (Kawamura et al., 2011; Terada et al., 2012). Thus, the observed accident-derived 129I at northern stations can be inferred to be supplied to the ocean via atmospheric input. Another possibility mechanism to explain the high concentration at northern stations is the microbial volatilization at the southern stations. Released 129I to the ocean transported to southward with seawater current and then volatilized by microbial activity and then moved to northward by wind and then deposited.

Considering the 129I results, seawater current, and simulation results, we conclude that 129I in the southern part from the 1FNPP was predominantly transported by seawater advection and that 129I in the northern part of the 1FNPP was predominantly transported via the atmosphere including microbial volatilization.

## Depth profile of 129I

The 129I concentrations in the seawater before and after the 1FNPP accident are plotted in Fig. 3 as a function of water depth. The all-depth profiles both before and after the 1FNPP accident show that the 129I concentration had its maximum in the surface-mixed layer and decreased with depth below the layer. Because there is no 129I source in the Pacific Ocean, the differences in the 129I concentrations before and after the accident indicate the accident-derived 129I in the water column. The 129I concentrations after the accident at 1000 m depth were at levels similar to those before the accident. The results indicate that, as of October 2011, the accident-derived 129I spread mainly in the upper 100 m depth of this study area and that only a small amount was transported to deep layers. Inventories of 129I obtained in this study in the water column between the surface and 1000 m are summarized in Table 2.

Integrated depth profiles show the inventory of 129I including the background and the accident-derived 129I. The ocean inventories of 129I depended on the distance from major reprocessing plants as well as of water mass structures in the water column (Alfimov et al., 2004; Schink et al., 1995; Suzuki et al., 2010). Therefore, background inventories in the western margin of the North Pacific Ocean must be applied to evaluate the amount of 129I derived from the 1FNPP accident. Since accident-derived 129I was not detected below 1000 m depth, the inventories for all profiles up to 1000 m are tabulated in Table 2 to evaluate the influence of the 1FNPP accident. Before the 1FNPP accident, the inventories in the upper 1000 m were in the 6.3–8.4 × 1012 atoms/m2 range and averaged inventoried 7.3 × 1012 atoms/m2. The inventories after the 1FNPP accident in the upper 1000 m were varied, (8.9–16.9) × 1012 atoms/m2. Inventories after the 1FNPP accident increased at all stations observed in this study. As a result of the accident, the increase in 129I across the study area ranged from 1.6–9.6 × 1012 atoms/m2.

## Dose estimation from 129I

This study shows that the 129I concentration has increased near Fukushima on account of the 1FNPP accident. Because iodine is a biophilic element, 129I enters the food chain and bio-accumulates in seafood. Because of its long half-life, an internal dose may be a serious concern for many generations. To evaluate the internal dose, an effective dose was roughly estimated based on ingestion of contaminated marine food. The effective dose can be obtained from the 129I concentration in seawater (Bq/L), the concentration factor for each marine food (L/kg), the total amount of marine food consumed in a year, and the effective dose coefficient (129I: 1.1 × 10−7 Sv/Bq for adults) (ICRP, 1995).

Radioiodine is accumulated in marine foods such as fish, crustaceans, mollusks, and macroalgae at concentration factors of 9, 3, 10, and 10,000 L/kg, respectively (IAEA, 2004). The latest national survey revealed that the ordinary Japanese individual ingests 32.6 g fish, 4.4 g crustaceans, 2.9 g mollusks, and 11.0 g macroalgae in a day (MHLW, 2012). By assuming that the 129I concentrations in surface seawater after the 1FNPP accident continue to have the value of 1.08–89.8 × 107 atoms/L (corresponding to 15.2–1255 nBq/L) for a year, the effective dose was estimated to be 6.7–550 × 10−11 Sv/year. It was quite lower than annual dose limit of 1.0 × 10−3 Sv/year. Since the 129I concentration in seawater near Fukushima is expected to decrease by dilution in the ocean, the actual effective dose would be much lower than the estimated one. Therefore, we concluded that the internal dose from the ingestion of seafood is negligibly small.

# Conclusions

This study focused on 129I, a long-lived radionuclide, derived from the 1FNPP accident. 129I concentrations in surface seawater before the 1FNPP accident, which can be used as the background level, showed a latitudinal gradient in horizontal distribution that could be expressed as a linear function of latitude between 32 °N and 44 °N. In the western margin of the North Pacific, the highest and average 129I concentrations measured after the 1FNPP accident were 73 times and approximately 8 times, respectively, higher than those before the accident in this study area. The inventory of 129I originating from the accident was 1.6–9.6 × 1012 atoms/m2 in this study area.

Based on the conservative estimation from measurement data of 129I, the internal dose from the ingestion of seafood is negligibly small.

Because 129I is one of the long-lived radionuclides, the data obtained in this study could be applied to the study of radionuclide migration from the 1FNPP accident. To investigate the impact of the 1FNPP accident more in detail, further investigation is required.

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Table 1. Concentrations of 129I in seawater samples

|  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Cruise | Station | Date |  |  | Depth | Salinity | Potential Temp. | Density | 129I/127I | 129I | |
| E | N | (m) | PSU | °C | σθ | ×10-11 | ×107 atoms/L | nBq/L |
| Before the 1FNPP accident | | | | | | | | | | | |
| OS08 | KNOT | 2008/5/10 | 154°58’ | 43°58’ | 4 | 33.178 | 5.477 | 26.176 | 5.85±0.21 | 1.78±0.07 | 24.9±1.0 |
|  |  |  |  |  | 10 | 33.178 | 5.474 | 26.176 | 6.22±0.22 | 1.83±0.08 | 25.6±1.1 |
|  |  |  |  |  | 20 | 33.217 | 5.339 | 26.223 | 6.25±0.19 | 1.78±0.06 | 24.9±0.8 |
|  |  |  |  |  | 30 | 33.289 | 5.084 | 26.309 | 5.53±0.25 | 1.56±0.08 | 21.8±1.2 |
|  |  |  |  |  | 39 | 33.400 | 4.717 | 26.438 | 6.38±0.23 | 1.72±0.07 | 24.1±1.0 |
|  |  |  |  |  | 48 | 33.428 | 4.661 | 26.467 | 5.70±0.24 | 1.63±0.09 | 22.7±1.2 |
|  |  |  |  |  | 74 | 33.572 | 5.079 | 26.534 | 4.79±0.21 | 1.53±0.08 | 21.4±1.0 |
|  |  |  |  |  | 99 | 33.563 | 4.544 | 26.586 | 5.70±0.20 | 1.63±0.07 | 22.8±0.9 |
|  |  |  |  |  | 122 | 33.531 | 3.930 | 26.625 | 5.31±0.18 | 1.47±0.05 | 20.5±0.7 |
|  |  |  |  |  | 149 | 33.512 | 3.116 | 26.687 | 4.42±0.23 | 1.32±0.08 | 18.4±1.1 |
|  |  |  |  |  | 197 | 33.637 | 2.859 | 26.810 | 3.98±0.17 | 1.18±0.06 | 16.5±0.8 |
|  |  |  |  |  | 299 | 33.869 | 3.191 | 26.965 | 2.65±0.12 | 0.83±0.04 | 11.7±0.6 |
|  |  |  |  |  | 396 | 33.991 | 3.181 | 27.063 | 2.06±0.10 | 0.66±0.04 | 9.2±0.5 |
|  |  |  |  |  | 495 | 34.118 | 3.279 | 27.155 | 1.45±0.09 | 0.44±0.03 | 6.2±0.4 |
|  |  |  |  |  | 743 | 34.309 | 2.842 | 27.348 | 0.56±0.06 | 0.17±0.02 | 2.3±0.3 |
|  |  |  |  |  | 990 | 34.414 | 2.528 | 27.459 | 0.22±0.05 | 0.07±0.01 | 1.0±0.2 |
|  |  |  |  |  | 1235 | 34.462 | 2.271 | 27.519 | 0.19±0.04 | 0.06±0.01 | 0.8±0.2 |
|  |  |  |  |  | 1483 | 34.510 | 2.079 | 27.573 | 0.07±0.05 | 0.02±0.02 | 0.3±0.2 |
|  |  |  |  |  | 1727 | 34.559 | 1.895 | 27.626 | 0.03±0.05 | 0.01±0.01 | 0.1±0.2 |
|  |  |  |  |  | 1974 | 34.592 | 1.722 | 27.666 | 0.03±0.04 | 0.01±0.01 | 0.1±0.2 |
|  |  |  |  |  | 2708 | 34.642 | 1.390 | 27.730 | 0.03±0.04 | 0.01±0.01 | 0.1±0.2 |
|  |  |  |  |  | 2955 | 34.651 | 1.320 | 27.743 | 0.01±0.04 | 0.01±0.01 | 0.1±0.2 |
| SY08 | off Kii | 2008/8/5 | 135°31’ | 32°28’ | 6 | 34.081 | 29.931 | 21.061 | 3.13±0.12 | 0.94±0.04 | 13.2±0.6 |
|  |  |  |  |  | 50 | 34.309 | 25.889 | 22.547 | 2.84±0.11 | 0.86±0.04 | 12.0±0.5 |
|  |  |  |  |  | 101 | 34.796 | 21.571 | 24.133 | 3.74±0.15 | 1.12±0.05 | 15.7±0.8 |
|  |  |  |  |  | 151 | 34.828 | 19.726 | 24.703 | 3.09±0.12 | 0.92±0.04 | 12.9±0.6 |
|  |  |  |  |  | 201 | 34.810 | 18.907 | 24.900 | 3.42±0.11 | 1.08±0.04 | 15.1±0.6 |
|  |  |  |  |  | 251 | 34.800 | 18.379 | 25.025 | 3.84±0.14 | 1.17±0.05 | 16.3±0.7 |
|  |  |  |  |  | 498 | 34.435 | 12.659 | 26.023 | 3.88±0.12 | 1.23±0.05 | 17.2±0.7 |
|  |  |  |  |  | 750 | 34.255 | 6.187 | 26.940 | 1.59±0.13 | 0.52±0.05 | 7.3±0.7 |
|  |  |  |  |  | 999 | 34.388 | 3.752 | 27.324 | 0.85±0.07 | 0.27±0.02 | 3.7±0.3 |
|  |  |  |  |  | 1248 | 34.460 | 2.931 | 27.461 | 0.60±0.06 | 0.20±0.02 | 2.8±0.3 |
|  |  |  |  |  | 1499 | 34.522 | 2.480 | 27.550 | 0.53±0.05 | 0.17±0.02 | 2.4±0.2 |
|  |  |  |  |  | 2002 | 34.599 | 1.883 | 27.660 | 0.84±0.08 | 0.27±0.03 | 3.7±0.4 |
| SY09 | Joban C | 2009/7/18 | 142°13’ | 36°48’ | 5 | 34.110 | 21.625 | 23.648 | 4.75±0.28 | 1.29±0.08 | 18.1±1.1 |
|  |  |  |  |  | 10 | 34.201 | 20.819 | 23.937 | 4.30±0.25 | 1.24±0.07 | 17.3±1.1 |
|  |  |  |  |  | 20 | 34.142 | 16.369 | 25.005 | 4.35±0.25 | 1.19±0.07 | 16.6±1.0 |
|  |  |  |  |  | 29 | 34.319 | 13.446 | 25.777 | 4.10±0.26 | 1.20±0.08 | 16.7±1.1 |
|  |  |  |  |  | 39 | 34.267 | 12.366 | 25.951 | 4.47±0.26 | 1.29±0.08 | 18.1±1.1 |
|  |  |  |  |  | 50 | 34.243 | 11.561 | 26.085 | 3.97±0.25 | 1.13±0.07 | 15.8±1.0 |
|  |  |  |  |  | 69 | 34.135 | 10.302 | 26.227 | 5.22±0.29 | 1.47±0.08 | 20.5±1.1 |
|  |  |  |  |  | 100 | 33.979 | 8.711 | 26.366 | 4.73±0.28 | 1.42±0.09 | 19.8±1.2 |
|  |  |  |  |  | 124 | 34.003 | 8.420 | 26.430 | 4.66±0.27 | 1.32±0.08 | 18.5±1.1 |
|  |  |  |  |  | 149 | 33.842 | 7.167 | 26.486 | 5.19±0.24 | 1.48±0.07 | 20.7±1.0 |
|  |  |  |  |  | 198 | 33.685 | 5.027 | 26.631 | 5.42±0.28 | 1.49±0.08 | 20.9±1.1 |
|  |  |  |  |  | 298 | 33.718 | 3.908 | 26.778 | 4.33±0.25 | 1.31±0.08 | 18.3±1.1 |
|  |  |  |  |  | 397 | 33.954 | 4.230 | 26.934 | 2.68±0.20 | 0.83±0.06 | 11.5±0.9 |
|  |  |  |  |  | 496 | 34.093 | 4.095 | 27.059 | 1.65±0.16 | 0.51±0.05 | 7.1±0.7 |
|  |  |  |  |  | 743 | 34.229 | 3.373 | 27.296 | 0.70±0.14 | 0.21±0.04 | 3.0±0.6 |
|  |  |  |  |  | 990 | 34.410 | 2.787 | 27.439 | 0.09±0.11 | 0.03±0.04 | 0.4±0.5 |
|  |  |  |  |  | 1236 | 34.476 | 2.397 | 27.527 | 0.04±0.10 | 0.01±0.03 | 0.2±0.4 |
|  |  |  |  |  | 1730 | 34.556 | 1.992 | 27.625 | 0.18±0.11 | 0.06±0.03 | 0.8±0.5 |
|  |  |  |  |  | 1976 | 34.586 | 1.814 | 27.664 | 0.03±0.10 | 0.01±0.03 | 0.1±0.4 |
|  |  |  |  |  | 2221 | 34.608 | 1.674 | 27.694 | 0.02±0.10 | 0.01±0.03 | 0.1±0.5 |
|  | Miyako | 2009/7/20 | 145°00’ | 40°00’ | 5 | 33.817 | 17.583 | 24.469 | 5.01±0.25 | 1.43±0.07 | 19.9±1.0 |
|  |  |  |  |  | 10 | 33.815 | 17.586 | 24.467 | 4.52±0.24 | 1.28±0.07 | 17.9±1.0 |
|  |  |  |  |  | 20 | 33.863 | 15.008 | 25.095 | 5.12±0.28 | 1.36±0.08 | 19.0±1.1 |
|  |  |  |  |  | 30 | 34.047 | 13.912 | 25.471 | 5.41±0.27 | 1.53±0.08 | 21.4±1.1 |
|  |  |  |  |  | 39 | 34.166 | 14.011 | 25.542 | 4.48±0.24 | 1.25±0.07 | 17.5±1.0 |
|  |  |  |  |  | 50 | 34.273 | 13.041 | 25.823 | 4.73±0.26 | 1.34±0.08 | 18.8±1.1 |
|  |  |  |  |  | 74 | 34.188 | 11.315 | 26.088 | 5.13±0.28 | 1.46±0.08 | 20.4±1.1 |
|  |  |  |  |  | 99 | 33.785 | 8.127 | 26.303 | 5.42±0.30 | 1.46±0.08 | 20.5±1.1 |
|  |  |  |  |  | 123 | 33.611 | 6.232 | 26.427 | 4.38±0.24 | 1.27±0.07 | 17.8±1.0 |
|  |  |  |  |  | 148 | 33.612 | 5.608 | 26.505 | 6.19±0.30 | 1.72±0.09 | 24.1±1.2 |
|  |  |  |  |  | 199 | 33.878 | 6.483 | 26.607 | 4.27±0.24 | 1.21±0.07 | 17.0±1.0 |
|  |  |  |  |  | 298 | 33.847 | 4.663 | 26.802 | 3.81±0.20 | 1.09±0.06 | 15.2±0.8 |
|  |  |  |  |  | 397 | 33.907 | 4.330 | 26.885 | 3.03±0.21 | 0.08±0.06 | 12.3±0.9 |
|  |  |  |  |  | 742 | 34.285 | 3.421 | 27.280 | 0.97±0.14 | 0.30±0.04 | 4.2±0.6 |
|  |  |  |  |  | 1482 | 34.518 | 2.177 | 27.580 | 0.15±0.10 | 0.05±0.03 | 0.7±0.4 |
|  |  |  |  |  | 1728 | 34.561 | 1.955 | 27.632 | 0.07±0.10 | 0.02±0.03 | 0.3±0.4 |
| After the 1FNPP accident | | | | | | | | | | | |
| KT11-06 | 6 | 2011/4/27 | 140°50’ | 36°00’ | 0 |  |  |  | 11.49±0.45 | 2.93±0.11 | 41.0±1.6 |
|  | 7 | 2011/4/28 | 141°40’ | 36°03’ | 0 |  |  |  | 18.88±0.51 | 4.65±0.13 | 65.1±1.8 |
|  | 8 | 2011/4/28 | 141°50’ | 36°31’ | 0 |  |  |  | 4.70±0.29 | 1.19±0.07 | 16.7±1.0 |
|  | 9 | 2011/4/29 | 142°40’ | 37°10’ | 0 |  |  |  | 22.70±0.85 | 5.60±0.21 | 78.3±2.9 |
|  | 10 | 2011/4/29 | 143°02’ | 38°09’ | 0 |  |  |  | 42.01±0.92 | 10.37±0.23 | 145.0±3.2 |
|  | 12 | 2011/4/29 | 143°40’ | 38°15’ | 0 |  |  |  | 13.14±0.60 | 3.29±0.15 | 46.1±2.1 |
|  | 14 | 2011/4/30 | 143°47’ | 38°13’ | 0 |  |  |  | 32.49±0.81 | 8.15±0.20 | 114.0±2.8 |
|  | A | 2011/4/29 | 142°50’ | 38°24’ | 9 | 33.988 | 11.262 | 25.941 | 50.78±0.96 | 12.97±0.25 | 181.4±3.4 |
|  |  |  |  |  | 29 | 33.936 | 10.511 | 26.034 | 70.37±1.53 | 17.65±0.38 | 246.9±5.4 |
|  |  |  |  |  | 49 | 34.252 | 10.403 | 26.229 | 30.92±0.88 | 8.05±0.23 | 112.6±3.2 |
|  |  |  |  |  | 98 | 33.741 | 6.928 | 26.437 | 12.59±0.51 | 3.18±0.13 | 44.4±1.8 |
|  |  |  |  |  | 198 | 33.701 | 5.484 | 26.589 | 5.89±0.32 | 1.50±0.08 | 21.0±1.1 |
|  |  |  |  |  | 594 | 34.192 | 4.269 | 27.116 | 1.29±0.25 | 0.34±0.07 | 4.8±0.9 |
|  |  |  |  |  | 1421 | 34.508 | 2.257 | 27.557 | 0.37±0.15 | 0.10±0.04 | 1.4±0.6 |
|  | B | 2011/4/30 | 143°28’ | 38°17’ | 9 | 34.273 | 12.759 | 25.657 | 27.73±0.78 | 6.83±0.20 | 95.5±2.7 |
|  |  |  |  |  | 50 | 34.274 | 11.513 | 25.894 | 11.64±0.56 | 2.84±0.14 | 39.7±1.9 |
|  |  |  |  |  | 99 | 34.212 | 10.396 | 26.094 | 8.82±0.43 | 2.22±0.11 | 31.1±1.5 |
|  |  |  |  |  | 198 | 33.987 | 8.294 | 26.435 | 5.66±0.41 | 1.45±0.10 | 20.2±1.5 |
|  |  |  |  |  | 497 | 34.070 | 4.073 | 27.039 | 2.53±0.25 | 0.69±0.07 | 9.6±0.9 |
|  |  |  |  |  | 989 | 34.375 | 2.932 | 27.392 | 0.91±0.23 | 0.25±0.06 | 3.5±0.9 |
|  | C | 2011/5/1 | 143°54’ | 39°23’ | 10 | 33.957 | 9.981 | 26.165 | 11.63±0.69 | 2.93±0.17 | 40.9±2.4 |
|  |  |  |  |  | 50 | 33.981 | 9.540 | 26.238 | 10.81±0.50 | 2.73±0.13 | 38.3±1.8 |
|  |  |  |  |  | 99 | 33.985 | 9.199 | 26.294 | 11.70±0.57 | 3.02±0.15 | 42.2±2.1 |
|  | 15 | 2011/5/1 | 143°53’ | 38°57’ | 0 |  |  |  | 12.84±0.61 | 3.19±0.15 | 44.7±2.1 |
|  | 16 | 2011/5/1 | 143°59’ | 39°45’ | 0 |  |  |  | 15.84±0.54 | 3.91±0.13 | 54.7±1.9 |
| BO-11-05 | UW1 | 2011/6/12 | 141°59’ | 36°00’ | 0 | 34.379 | 21.299 | 23.643 | 4.47±0.27 | 1.08±0.06 | 15.2±0.9 |
|  | UW2 | 2011/6/12 | 142°19’ | 36°31’ | 0 | 33.426 | 17.677 | 24.576 | 362.25±4.87 | 89.76±1.21 | 1255±16.9 |
|  | UW3 | 2011/6/12 | 142°31’ | 37°01’ | 0 | 34.011 | 18.224 | 24.443 | 58.99±1.22 | 14.36±0.30 | 200.8±4.2 |
|  | UW4 | 2011/6/13 | 142°45’ | 37°30’ | 0 | 34.108 | 18.947 | 24.262 | 36.36±0.75 | 8.98±0.18 | 125.6±2.6 |
| KH-11-07 | FS1 | 2011/8/2 | 142°10’ | 37°20’ | 0 |  |  |  | 23.29±0.60 | 5.56±0.14 | 77.8±2.0 |
|  |  |  |  |  | 101 | 33.772 | 8.277 | 26.437 | 7.33±0.33 | 2.00±0.09 | 28.0±1.3 |
|  |  |  |  |  | 198 | 33.573 | 4.948 | 26.878 | 6.76±0.37 | 1.85±0.10 | 25.9±1.4 |
|  |  |  |  |  | 297 | 33.658 | 3.589 | 27.022 | 5.43±0.36 | 1.53±0.10 | 21.3±1.4 |
|  |  |  |  |  | 496 | 34.011 | 3.703 | 27.011 | 2.32±0.28 | 0.96±0.08 | 13.4±1.1 |
|  |  |  |  |  | 692 | 34.247 | 3.506 | 27.030 | 2.61±0.25 | 0.78±0.08 | 10.9±1.1 |
|  | ES2 | 2011/8/2 | 142°15’ | 37°04’ | 0 |  |  |  | 11.02±0.48 | 2.52±0.11 | 35.2±1.5 |
|  |  |  |  |  | 100 | 33.969 | 9.217 | 26.291 | 9.10±0.41 | 2.44±0.11 | 34.1±1.5 |
|  |  |  |  |  | 199 | 34.095 | 7.752 | 26.515 | 4.39±0.24 | 1.21±0.07 | 16.9±0.9 |
|  |  |  |  |  | 297 | 33.906 | 5.286 | 26.839 | 4.58±0.29 | 1.28±0.08 | 17.9±1.1 |
|  |  |  |  |  | 496 | 34.148 | 4.571 | 26.920 | 2.86±0.19 | 0.83±0.06 | 11.6±0.8 |
|  |  |  |  |  | 692 | 34.246 | 3.603 | 27.020 | 2.02±0.27 | 0.59±0.08 | 8.3±1.1 |
|  |  |  |  |  | 989 | 34.400 | 2.864 | 27.089 | 1.94±0.26 | 0.57±0.08 | 8.0±1.1 |
|  |  |  |  |  | 1978 | 34.591 | 1.862 | 27.171 | 0.48±0.15 | 0.14±0.05 | 2.0±0.6 |
|  | FS2 | 2011/8/2 | 142°00’ | 37°00’ | 0 |  |  |  | 11.02±0.48 | 2.65±0.12 | 37.0±1.6 |
|  |  |  |  |  | 101 | 34.025 | 9.732 | 26.207 | 14.38±0.52 | 2.91±0.10 | 40.6±1.5 |
|  |  |  |  |  | 197 | 33.770 | 6.585 | 26.677 | 7.62±0.45 | 2.03±0.12 | 28.4±1.7 |
|  |  |  |  |  | 299 | 33.810 | 5.256 | 26.843 | 7.96±0.38 | 2.18±0.10 | 30.4±1.4 |
|  |  |  |  |  | 497 | 33.985 | 3.523 | 27.028 | 4.77±0.22 | 1.39±0.06 | 19.4±0.9 |
|  |  |  |  |  | 693 | 34.221 | 3.575 | 27.023 | 3.36±0.38 | 0.98±0.11 | 13.6±1.5 |
|  |  |  |  |  | 990 | 34.372 | 3.017 | 27.075 | 1.38±0.18 | 0.41±0.05 | 5.7±0.7 |
|  |  |  |  |  | 1307 | 34.480 | 2.420 | 27.127 | 1.09±0.19 | 0.33±0.06 | 4.6±0.8 |
|  | FS5 | 2011/8/3 | 141°20’ | 36°00’ | 0 |  |  |  | 99.62±2.12 | 23.66±0.50 | 330.9±7.0 |
| KT-11-27 | ST08 | 2011/10/29 | 142°00’ | 37°00’ | 0 |  |  |  | 15.84±0.63 | 4.33±0.17 | 60.6±2.4 |
|  |  |  |  |  | 49 | 33.881 | 17.928 | 24.515 | 11.62±0.42 | 3.18±0.12 | 44.5±1.6 |
|  |  |  |  |  | 100 | 34.135 | 11.199 | 25.951 | 10.83±0.50 | 2.94±0.14 | 41.2±1.9 |
|  |  |  |  |  | 201 | 33.858 | 7.023 | 26.618 | 5.34±0.35 | 1.50±0.10 | 20.9±1.4 |
|  |  |  |  |  | 300 | 33.773 | 4.542 | 26.923 | 4.09±0.27 | 1.15±0.08 | 16.1±1.1 |
|  |  |  |  |  | 500 | 34.031 | 3.944 | 26.986 | 2.08±0.17 | 0.60±0.05 | 8.4±0.7 |
|  |  |  |  |  | 1000 | 34.365 | 2.901 | 27.086 | 0.62±0.15 | 0.19±0.05 | 2.6±0.6 |
|  | K8 | 2011/10/29 | 142°11’ | 37°20’ | 0 |  |  |  | 13.32±0.46 | 3.61±0.12 | 50.5±1.7 |
|  |  |  |  |  | 49 | 34.194 | 17.051 | 24.726 | 9.38±0.53 | 2.50±0.14 | 35.0±2.0 |
|  |  |  |  |  | 99 | 34.082 | 10.493 | 26.077 | 9.99±0.38 | 2.75±0.11 | 38.4±1.5 |
|  |  |  |  |  | 201 | 33.706 | 5.621 | 26.799 | 4.41±0.24 | 1.23±0.07 | 17.3±0.9 |
|  |  |  |  |  | 299 | 33.927 | 5.463 | 26.818 | 3.25±0.22 | 0.92±0.06 | 12.8±0.9 |
|  |  |  |  |  | 500 | 34.093 | 4.278 | 26.952 | 1.71±0.15 | 0.50±0.04 | 7.0±0.6 |
|  |  |  |  |  | 700 | 34.231 | 3.469 | 27.033 | 0.92±0.13 | 0.27±0.04 | 3.8±0.6 |
|  |  |  |  |  | 1020 | 34.394 | 2.719 | 27.101 | 0.22±0.15 | 0.07±0.04 | 0.9±0.6 |

Table 2 Inventory of 129I before and after the 1FNPP accident and its influence

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Cruise | Station | Sampling date | Inventory  (× 1012 atoms/m2) | Increase after the accident  (× 1012 atoms/m2) | Reference |
| OS08 | KNOT | 2008/5/10 | 6.3 |  | This study |
| SY08 | off Kii | 2008/8/5 | 8.4 |  | This study |
| SY09 | Joban C | 2009/7/18 | 7.0 |  | This study |
|  | Miyako | 2009/7/20 | 7.7 |  | This study |
| SY07 | Offshore of Kushiro | 2007/8/1 | 7.2 |  | Suzuki et al., 2010 |
| Average before the accident | | | 7.3 |  |  |
| KT-11-06 | A | 2011/4/29 | 16.9 | 9.6 | This study |
|  | B | 2011/4/30 | 11.2 | 3.9 | This study |
| KH-11-07 | FS1 | 2011/8/2 | 13.9 | 6.6 | This study |
|  | ES2 | 2011/8/2 | 10.8 | 3.5 | This study |
|  | FS2 | 2011/8/2 | 15.3 | 8.0 | This study |
| KT-11-27 | ST08 | 2011/10/29 | 10.7 | 3.4 | This study |
|  | K8 | 2011/10/29 | 8.9 | 1.6 | This study |

Fig. 1 Map of sampling locations and the result of surface 129I concentrations before the 1FNPP accident (a) and afterwards (b–e). After the accident, seawater sampling was undertaken during four cruises: b) KT-11-06, c) BO-11-05, d) KH-11-07, and e) KT-11-27. In Fig. 1 c), circle symbols were cited from Hou et al., 2013. Red at the sampling locations indicates that the 129I concentration is higher than 10 × 107 atoms/L.

Fig. 2 129I concentrations in surface seawater before and after the 1FNPP accident as a function of latitude. The dark green, light green, dark orange, and light orange symbols indicate cruises KT-11-06, BO-11-05, KH11-07, and KT-11-27, respectively, after the 1FNPP accident. The purple symbols were cited from Hou et al., 2013. The white symbols indicate a cruise before the Fukushima NPP accident.

Fig. 3 Depth profiles of 129I before and after the 1FNPP accident. The dark green, dark orange, and light orange symbols indicate cruises KT-11-06, KH11-07, and KT-11-27, respectively, after the 1FNPP accident. The white symbols indicate a cruise before the 1FNPP accident.