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Input of ^{129}I into the western Pacific Ocean resulting from the Fukushima nuclear event

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Abstract We present an initial characterization of the input of ^{129}I into the Pacific Ocean resulting from the 2011 Fukushima nuclear accident. This characterization is based primarily on ^{129}I measurements on samples collected from a research cruise conducted in waters off the eastern coast of Japan in June 2011. These measurements were compared with samples intended to reflect pre-Fukushima background that were collected during a May 2011 transect of the Pacific by a commercial container vessel. In surface waters, we observed peak ^{129}I concentrations of $\sim$300 μBq/m^3 which represents an elevation of nearly three orders of magnitude compared to pre-Fukushima backgrounds. We coupled our ^{129}I results with ^{137}Cs measurements from the same cruise and derived an average ^{129}I/^{137}Cs activity ratio of 0.442×10^{-6} for the effluent from Fukushima. Finally, we present ^{129}I depth profiles from five stations from this cruise which form the basis for future studies of ocean transport and mixing process as well as estimations of the total budget of ^{129}I released into the Pacific.

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Introduction

The tsunami resulting from the March 11, 2011 Tōhoku earthquake triggered a catastrophic accident at the Fukushima Daiichi nuclear power plant that caused the release of radioactive elements into the environment [1–5]. Some of these radioactive species (e.g., ^{131}I, ^{134}Cs, ^{137}Cs) are of major concern because they present a hazard associated with radiological dose to surrounding plants, animals, and human populations. Significant effort has been directed to assess the magnitude of the release of radioactivity to the surrounding environment and incorporation into plant and animal food supplies. Such studies are important for estimating the radiological dose exposure to local human populations. The primary input of radioactivity into the Pacific Ocean was a combination of direct discharge from the power plants and deposition resulting from atmospheric releases caused by explosions during the nuclear disaster [5]. Determining the relative proportion of these two release routes is important for accurate dose assessments.

^{129}Iodine is another radioactive species that was released by the Fukushima nuclear accident. Owing to an extremely long radioactive half-life (15.7 Ma), ^{129}I presents virtually no radiological hazard so it has understandably been given less focus thus far. It is nonetheless valuable to assess the environmental release of ^{129}I for a number of reasons.

First, ^{129}I is a powerful tool for studying ocean transport and mixing processes [6–12]. In addition to the long radioactive half-life of ^{129}I, the long residence time ($\sim$245 ka) and relatively low bioavailability of iodine

enable ^{129}I to behave as a nearly conservative tracer in the ocean. The utility of ^{129}I has been extensively demonstrated in many studies in which releases from the Sellafield and La Hague reprocessing plants were traced into the surrounding seas (e.g., Norwegian, Baltic, etc.) and further the North Atlantic Ocean. The Fukushima releases represent a similar, albeit less intense and more discrete, point source for tracing water mass movement in the Western Pacific Ocean.

Another valuable reason for investigating the release of ^{129}I from Fukushima is its potential for differentiating between atmospherically deposited and directly discharged radioactivity in the Pacific Ocean. It has been demonstrated that atmospherically-released radioiodine and radiocesium from the 1986 Chernobyl nuclear accident were chemically fractionated because of differences in volatility [13, 14]. A similar fractionation can be expected in the atmospheric release from Fukushima, while the direct discharge should more closely reflect the relative ratio of radioiodine and radiocesium in the damaged fuel rods. If the degree of fractionation is sufficiently high, then it should be possible to assess the relative contributions of atmospherically deposited and directly discharged radioactivity from Fukushima in the Pacific Ocean. ^{129}Iodine is the only radioactive isotope of iodine with a sufficiently long half-life to allow this type of investigation.

The purpose of the present work is to present an initial assessment of the input of Fukushima-derived ^{129}I into the Western Pacific Ocean. This assessment is primarily based on measurements of water samples collected during a research cruise conducted off the eastern coast of Japan during June 2011. In order to put these measurements into proper context, we establish a pre-Fukushima background of ^{129}I through measurements of water samples collected from a container ship that crossed the Pacific Ocean in May 2011. We also present an initial estimate of the ^{129}I/^{137}Cs activity ratio from the direct discharge of the Fukushima effluent and compare this to values from Chernobyl.

Experimental

To establish a pre-Fukushima background of ^{129}I in the Pacific Ocean, we collected water samples from a container ship (OOCL Tokyo) during a May 2011 crossing of the Pacific from the Port of Hong Kong (China) to the Port of Long Beach (USA). The exact coordinates and time of collections are listed in Table 1. In brief, the OOCL Tokyo's course took her from the South China Sea, through the Taiwan Straight into the East China Sea, then into the Philippine Sea just south of Kyushu, where she began a great circle route to a latitude of $\sim$43°N. Samples were collected using a continuous flow surface seawater line.

To assess the input of Fukushima-derived ^{129}I into the Pacific Ocean, water samples were collected during a research cruise conducted on board the R/V Kàimikai-*o*-Kanaloa (KOK) from June 4 to June 18, 2011. The cruise plan has been previously published [15], but in short, consisted of 32 major sampling stations ranging from 30 to 600 km from the Fukushima Daiichi nuclear power plant. Stations were selected to focus on the area north of the Kuroshio current, but a few stations were located within the current. A standard Rosette water sampler fitted with Niskin bottles and conductivity, temperature, and depth (CTD) sensors was deployed at each station. Water samples were collected at discrete depths ranging from 1,000 m up to near the sea surface (5–20 m).

Samples from both the OOCL Tokyo and the KOK were collected in a similar manner. Prior to collection, the supply tap (from either the continuous flow surface sampler or a Niskin bottle) was opened and allowed to flush for a period of time to ensure samples were not contaminated with exterior residual water. Bottles (0.5-L, HDPE, acid-cleaned with 2 % nitric acid) were rinsed several times before being filled. Bottles were sealed, their caps taped, and stored in the dark until sampled for ^{129}I analysis.

Samples were prepared for ^{129}I analysis using an adapted version of a commonly-used solvent extraction procedure [16–18]. Briefly, 0.5 mg of iodine carrier (Woodward Iodine Corporation) with a very low ^{129}I content (^{129}I/^{127}I $\sim 2 \times 10^{-14}$) was added to a 250-mL aliquot of each sample. Dissolved inorganic iodine was reduced to iodide by addition of sodium sulfite and hydroxylamine hydrochloride and then oxidized to molecular iodine by addition of nitric acid and sodium nitrite. The molecular iodine was extracted into chloroform and then back-extracted into a reducing solution of aqueous sodium sulfite and potassium hydroxide. The measured recovery rate of this method is typically $\sim$50–60 %. Silver iodide was precipitated by addition of silver nitrate and the precipitate was rinsed and dried before being mechanically mixed with niobium powder and loaded into a target holder for analysis. The ^{129}I/^{127}I isotope ratios of the prepared targets were measured by accelerator mass spectrometry (AMS) at the Lawrence Livermore National Laboratory Center for AMS [19].

Results and discussion

The results of ^{129}I/^{127}I measurement of the surface water samples collected during the May 2011 crossing of the Pacific by the OOCL Tokyo are presented in Table 1. A consistently increasing trend from west to east is observed, although the total range in measured values is only a factor of two. All of the measured values fall within

Table 1 $^{129}I/^{127}I$ ratios measured in surface water samples collected during the May 2011 crossing of the Pacific Ocean by the OOCL Tokyo

Station	Coordinates	UTC date/time	$^{129}I/^{127}I$ ($\times 10^{-11}$)
1	22°22′N, 115°57′E	5/16/11 11:56	2.68 (0.11)
3	24°19′N, 118°58′E	5/16/11 22:15	2.50 (0.09)
6	26°25′N, 122°45′E	5/17/11 10:00	2.61 (0.10)
9	30°35′N, 130°07′E	5/18/11 07:37	2.67 (0.10)
12	32°49′N, 137°31′E	5/19/11 02:44	2.87 (0.10)
15	35°30′N, 143°54′E	5/19/11 19:17	2.84 (0.11)
18	38°24′N, 150°27′E	5/20/11 11:55	3.96 (0.14)
21	41°25′N, 159°12′E	5/21/11 09:09	2.16 (0.09)
24	43°19′N, 170°11′E	5/22/11 09:32	2.97 (0.11)
27	43°16′N, 178°47′W	5/23/11 08:41	3.14 (0.12)
30	43°08′N, 169°03′W	5/24/11 07:45	2.92 (0.10)
33	42°59′N, 159°45′W	5/25/11 08:03	3.44 (0.13)
36	42°10′N, 150°41′W	5/26/11 08:20	3.48 (0.12)
39	40°45′N, 142°25′W	5/27/11 06:55	4.31 (0.15)
42	38°39′N, 134°08′W	5/28/11 07:34	4.86 (0.17)
45	36°18′N, 127°10′W	5/29/11 05:35	5.16 (0.17)

Values in parentheses represent 1-sigma uncertainty associated with the analytical measurements

the lower end of the range given for shallow seawater by Snyder [20]. The reason for the increasing trend is not completely understood. As these samples were collected 2 months following the Fukushima accident, there is likely some component of ^{129}I resulting from atmospheric deposition. However, if this were the dominant recent input of ^{129}I, then a peak would be expected closer to the point source. Indeed, a local maxima is observed at Station 18, which is geographically the closest station to Fukushima. The fact that the highest values of $^{129}I/^{127}I$ were observed at the greatest distance (Stations 39, 42, 45) from Fukushima implies that there is another input of ^{129}I into the Pacific.

The results of ^{129}I measurements of surface samples from the June 2011 KOK cruise are presented in Fig. 1. For comparison to corresponding ^{137}Cs measurements from [15], the ^{129}I data in this figure are expressed in units of $\mu Bq/m^3$ by converting the measured $^{129}I/^{127}I$ ratio using the literature value of iodine concentration in seawater of 0.47 nM [21]. The ^{129}I data show the expected general trend of increasing concentration with proximity to the Fukushima nuclear facility. As discussed in [15], the confluence of the Kuroshio and Oyashiro currents create an eddy that effectively constrained a bulk of the ^{129}I from migrating south into the Kuroshiro. There is general agreement between the measured ^{129}I concentrations and the ^{137}Cs concentrations presented in [15], however the $^{129}I/^{137}Cs$ ratio is not constant for all stations.

Figure 2 shows a plot of the $^{129}I/^{137}Cs$ activity ratio as a function of the total ^{137}Cs concentration measured in the surface waters from the June 2011 KOK cruise. At low ^{137}Cs concentrations, the $^{129}I/^{137}Cs$ ratio asymptotically approaches a value reflecting pre-Fukushima value of

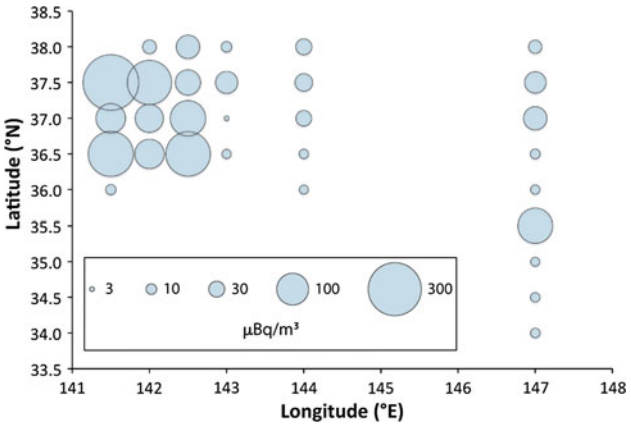

Fig. 1 Measured concentration of ^{129}I in surface waters off the eastern coast of Japan. Samples were collected during June of 2011 from the R/V Kàimikai-*o*-Kanaloa

fallout in the Pacific. As the ^{137}Cs concentration increases, this ratio approaches a value that reflects the average ratio in Fukushima effluent. Based on this observation, the data were fit to a two end-member mixing equation:

$$\left[\frac{^{129}I}{^{137}Cs}\right]_{Meas} = \left(\frac{[^{137}Cs]_{Meas}}{[^{137}Cs]_{Meas}+[^{137}Cs]_{Bkg}}\right) \cdot \left[\frac{^{129}I}{^{137}Cs}\right]_{Fuk} + \left(\frac{[^{137}Cs]_{Bkg}}{[^{137}Cs]_{Meas}+[^{137}Cs]_{Bkg}}\right) \cdot \left[\frac{^{129}I}{^{137}Cs}\right]_{Bkg} \quad (1)$$

where $\left[\frac{^{129}I}{^{137}Cs}\right]$ represents the $^{129}I/^{137}Cs$ activity ratio, $[^{137}Cs]$ represents the ^{137}Cs concentration, and the subscripts: Meas, Fuk, Bkg refer to the values measured from

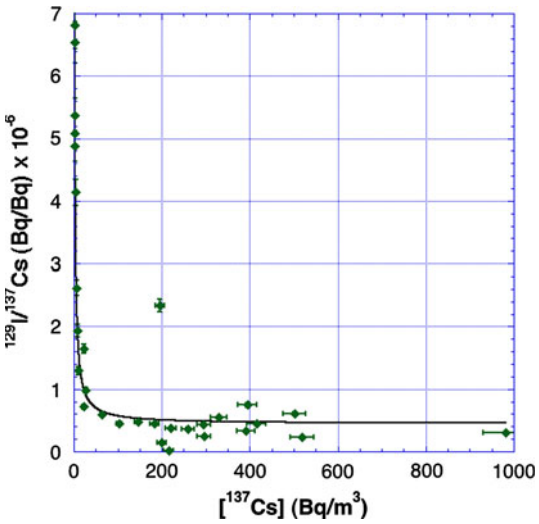

Fig. 2 Plot showing the $^{129}I/^{137}Cs$ activity ratio as a function of ^{137}Cs concentration measured in surface water samples collected from the June 2011 cruise of the R/V Kàimikai-*o*-Kanaloa. The data were fitted to a two end-member mixing function given in Eq. 1

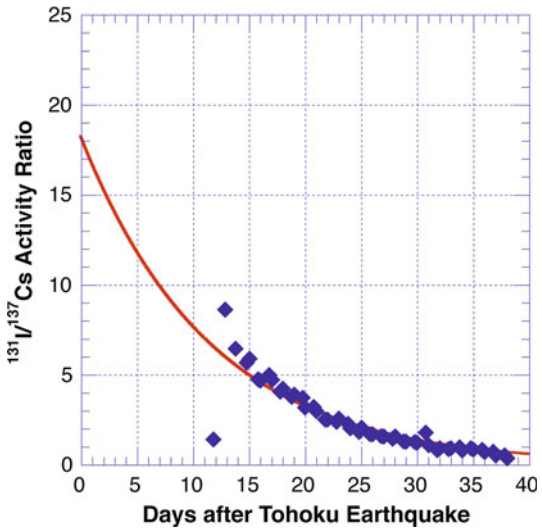

Fig. 3 Plot of the measured $^{131}I/^{137}Cs$ activity ratio measured at a sampling station located approximately 30 m north of the discharge canal of units 5 and 6 of the Fukushima Daiichi nuclear power plant. Data were taken from press releases issues by TEPCO [22]. Line shows a fit of the data to an exponential function corresponding to radioactive decay extrapolated back to the date of the Tōhoku earthquake

the June 2011 KOK cruise, the derived average value in Fukushima effluent, and the derived average value in pre-Fukushima Pacific Ocean seawater respectively. From this fit we derive an average $^{129}I/^{137}Cs$ activity ratio of 0.443×10^{-6} in Fukushima effluent and a ratio of 58.9×10^{-6} for pre-Fukushima Pacific Ocean water.

It is instructive to compare the $^{129}I/^{137}Cs$ activity ratio derived from this data set to the comparable ratio from Chernobyl. For example, [13] reports a value of 0.275×10^{-6} for the $^{129}I/^{137}Cs$ activity ratio calculated in the active zone of the 4th block of Chernobyl nuclear power plant at the time of that accident. This value is 38 % lower than our derived value for Fukushima effluent. For further comparison, we examined gamma spectroscopy measurements of ^{131}I and ^{137}Cs in near-shore seawater samples as reported by the Tokyo Electric Power Company (TEPCO) beginning on March 23, 2011 [22]. Figure 3 shows the measured $^{131}I/^{137}Cs$ activity ratio as a function of days after the Tōhoku earthquake. Fitting these data to an exponential decay function yields an initial (i.e., at the time of the earthquake) ratio of 18.2 which is close to the estimates of 20–60 reported in [4, 5]. The corresponding value from [13] is 10.4 which is 43 % lower than our derived value for Fukushima effluent. This is consistent with the relative difference between the $^{129}I/^{137}Cs$ ratios discussed above.

The results of ^{129}I measurements in depth profile samples collected from five stations during the June 2011 KOK cruise are shown in Fig. 4 along with corresponding basic hydrographic data (salinity and temperature). As expected, the three near-field stations (21, 27, 29) show the highest concentration of ^{129}I throughout the depth profile. Stations 21 and 27 show a peak in the ^{129}I concentration at the surface with an exponential decrease with depth. In contrast, stations 14 and 29 show a peak in ^{129}I concentration below the surface, but still within the mixed layer. The ^{129}I concentration within the mixed layer of station 5 seems to be fairly well homogenized. When augmented with depth profile data from the rest of the sampling stations, these data will form the basis for estimating the total release of ^{129}I activity from Fukushima as well lay the foundation for interpreting hydrodynamic transport and mixing processes.

Conclusion

Our work provides a characterization of the ^{129}I that was released into the Pacific Ocean resulting from the Fukushima nuclear accident. Observed concentrations of ^{129}I in near-shore surface waters were nearly three order of magnitude above the pre-Fukushima background based on our measurements. This input signal should be sufficiently high to enable future studies that exploit ^{129}I as a tracer for ocean movement and mixing processes. Furthermore, we provide an initial estimate of the average $^{129}I/^{137}Cs$ activity ratio in Fukushima effluent. Deviations from this average ratio were observed that may reflect inputs of both atmospherically-deposited and directly-discharged radioactivity in the Pacific Ocean. Further studies of the $^{129}I/^{137}Cs$ ratio in a broader range of samples are required to estimate the

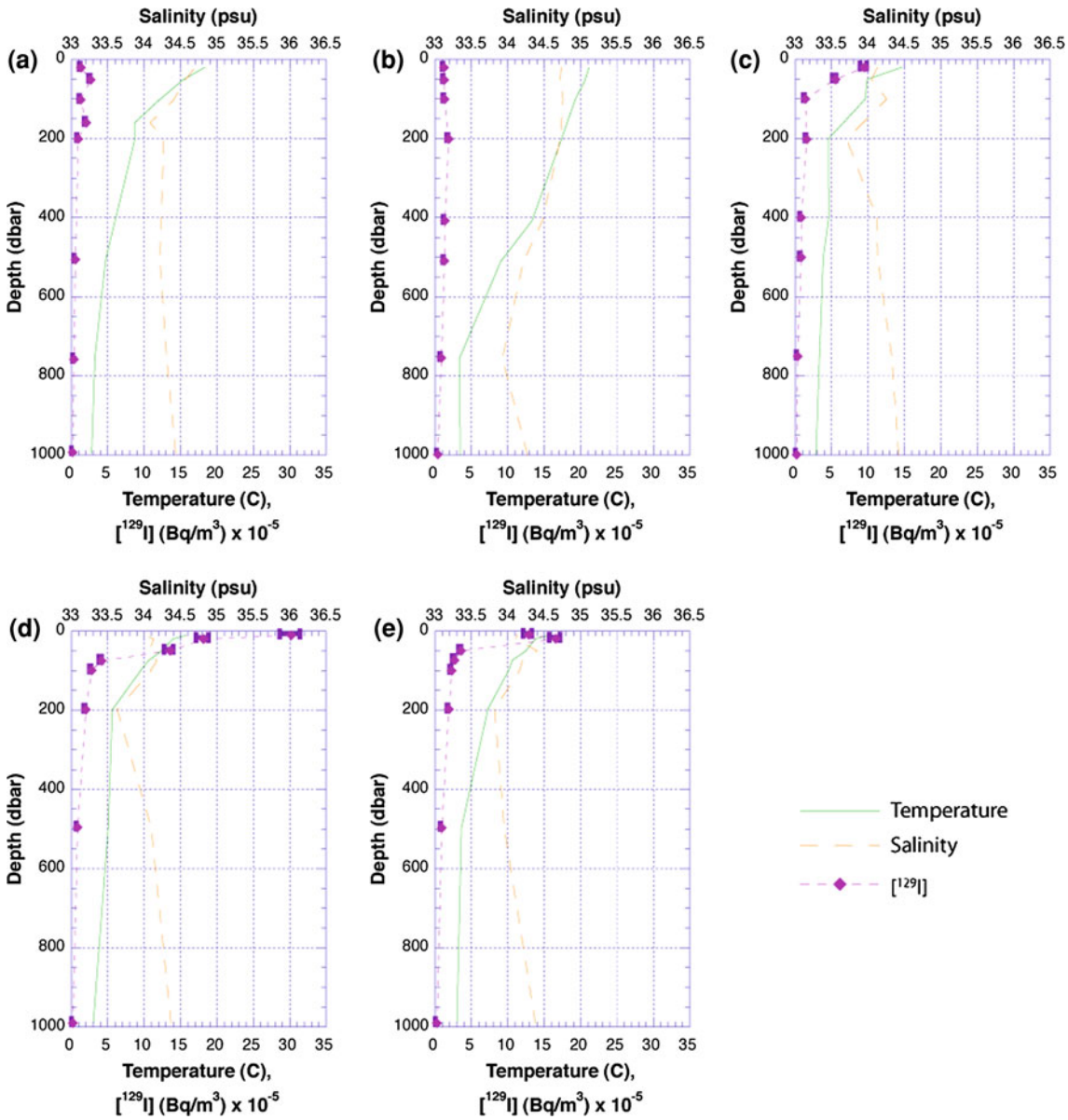

Fig. 4 Plot of salinity, temperature, and ^{129}I concentration as a function of depth at five stations from the June 2011 cruise of the R/V Kàimikai-*o*-Kanaloa: **a** Station 5 [36°N, 147°E]; **b** Station 14 [36°N, 144°E]; **c** Station 21 [37.5°N, 142.5°E]; **d** Station 27 [36.5°N, 141.5°E]; Station 29 [36.5°N, 142°E]

relative contribution of these two inputs. Finally, the observed depth profiles of ^{129}I are mainly consistent with ^{137}Cs, however some anomalies were observed that warrant additional investigation.

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