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Measurements of ^{129}I in the Pacific Ocean at Scripps Pier and Pacific Northwest sites: A search for effects from the 2011 Fukushima Daiichi Nuclear Power Plant accident and Hanford

Ching-Chih Chang [a,b], George S. Burr [a,c], A.J. Timothy Jull [a,b,e,*], Joellen Russell [b], Antra Priyadarshi [d], Mang Lin [d,1], Mark Thiemens [d], Dana Biddulph [a]

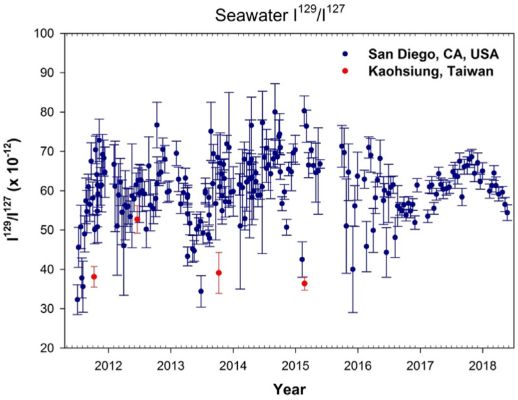
[a] *University of Arizona AMS Laboratory, University of Arizona, Tucson, AZ, USA*
[b] *Department of Geosciences, University of Arizona, Tucson, AZ, USA*
[c] *Department of Geosciences, National Taiwan University Research Center for Future Earth, Taipei, Taiwan*
[d] *Department of Chemistry and Biochemistry, University of California San Diego, La Jolla, CA, USA*
[e] *Isotope Climatology and Environmental Research Centre, Institute for Nuclear Research, Debrecen, Hungary*

HIGHLIGHTS

- Iodine-129 monitored continuously at Scripps Pier since 2011
- Iodine-129 variations observed but not attributable to Fukushima Nuclear accident.
- We expect that I-129 is diluted to a level where it cannot be observed.
- Iodine-129 can be observed in the Columbia River due to Hanford facility.

GRAPHICAL ABSTRACT

In June 2011, a few months after the disaster, we began a surface ocean ^{129}I monitoring program, with samples from Scripps Pier, La Jolla, California, USA. After 7 years of ocean transport, a distinct signal has not yet arrived at Scripps Pier. We have however, recorded an interesting systematic seasonal ^{129}I time series record that stems from surface circulation variations along the California coast.



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ABSTRACT

Radionuclides from the Fukushima Daiichi Nuclear Power Plant were released directly into the ocean as a result of the Great East Japan Earthquake on March 11, 2011. This material became entrained in surface ocean currents and subsequently transported for great distances. In June 2011, a few months after the disaster, we began a surface ocean ^{129}I monitoring program, with samples from Scripps Pier, La Jolla, California, USA, with the expectation that surface currents originating off the east coast of Japan would eventually carry radionuclides to the La Jolla site. After 7 years of ocean transport, a distinct signal has not yet arrived at Scripps Pier. We have however, recorded an interesting systematic seasonal ^{129}I time series record that stems from surface circulation variations along the California coast. To provide a more comprehensive picture of the ^{129}I budget in coastal surface waters

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[*] Corresponding author at: Department of Geosciences, University of Arizona, Tucson, AZ, USA.
E-mail address: jull@email.arizona.edu (A.J.T. Jull).

[1] Present address: State Key Laboratory of Isotope Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou, China

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off the west coast of the U.S., we also include ^{129}I data from the Columbia River, and offshore sites along the coast of Washington State. Anthropogenic nuclides are carried by the Columbia River into the Pacific Ocean from the vicinity of the decommissioned Hanford nuclear facility. We find highly elevated ^{129}I/^{127}I values in the Columbia River, downstream from the Hanford site, but this anthropogenic ^{129}I becomes significantly diluted once it reaches the Pacific Ocean. Nonetheless, its imprint persists in surface seawater off the west coast of the U.S. that has significantly higher ^{129}I/^{127}I levels than other surface sites in the Pacific Ocean.

1. Introduction

On March 11, 2011, the Great East Japan Earthquake and subsequent tsunami triggered nuclear shutdowns, failures, and partial meltdowns at the Fukushima Daiichi reactor complex. In the weeks following this catastrophe a series of radioactive releases occurred, including large quantities of iodine radionuclides. Shortly after the accident, elevated amounts of ^{131}I were detected in Japan and a portion of this anthropogenic pulse was rapidly transported around the world in the atmosphere (Hsu et al., 2012; Adachi et al., 2013). In addition to atmospheric fallout, contaminated water was also directly injected into the ocean. Seawater samples of anthropogenic radionuclides incollected south of the nuclear plant discharge channel, 30 km from the coast, were observed to contain high levels of ^{137}Cs and ^{129}I. It is estimated that during the accident, a total 2.4–7 GBq of Iodine-129 was discharged into the ocean (Povinec et al., 2013; Guilderson et al., 2014). Many studies of the radioactive plume in the ocean from this accident have been conducted (Buesseler et al., 2011; Aoyama et al., 2013; Cascuberta et al., 2017; Charette et al., 2013; Guilderson et al., 2014; Povinec et al., 2013; Smith et al., 2015, 2017). The elevated level of radionuclides is a concern for human health, and highlights the need for monitoring efforts that can help us understand how radionuclides spread through the atmosphere and ocean, on a global scale.

Iodine-129 is a long-lived radionuclide with a half-life of 15.7 million years. It has been shown to be a useful tracer for environmental research due in part to its high mobility in nature, and significant anthropogenic production (Edwards, 1962; Muramatsu and Ohmomo, 1986; Santschi et al., 1996; Raisbeck et al., 1995; Schmidt et al., 1998; Hou et al., 2009; Povinec et al., 2010). There are two natural production pathways for ^{129}I: 1) by spontaneous fission of ^{238}U, and 2) by the interaction of cosmic-rays with xenon in the atmosphere; however over the past 70 years, anthropogenic ^{129}I sources have overwhelmed natural production. These are dominated by nuclear fuels reprocessing activities, but also include above-ground nuclear testing and accidents. At present, global ^{129}I in surface ocean waters are about 1–2 orders of magnitude higher than in the estimated pre-nuclear era ^{129}I/^{127}I values of between 5×10^{-13} and 1.5×10^{-12} (Fabryka-Martin et al., 1984; Moran et al., 1998; Edmonds and Morita, 1998; Biddulph et al., 2006). It has also long been known that the industrial nuclear fuels reprocessing plants in Sellafield (England) and LaHague (France) are the primary sources of anthropogenic ^{129}I in Atlantic Ocean surface waters (Yiou et al., 1994, 2002), and that this ^{129}I can now be found everywhere around the globe (Xing et al., 2015; Fehn and Snyder, 2000).

There have been a number of historical accidental anthropogenic ^{129}I releases to the environment, including the Chernobyl reactor accident (Paul et al., 1987; Kutschera et al., 1988) and a total of seven nuclear submarine accidents (Alfimov et al., 2004; Povinec et al., 2010; Hu et al., 2010). The Chernobyl disaster released between 40-48 GBq ^{129}I (Aldahan et al., 2007; Kashparov et al., 2003) directly to the atmosphere. The estimated total range of ^{129}I values released as a result of the Fukushima accident overlap with those for Chernobyl, from 6.6G Bq ^{129}I (Steinhauser et al., 2014) to 55G Bq ^{129}I (Hou et al., 2013). Although the amounts are comparable, it should be noted that in the case of Fukushima, 80% of the total radioactivity was released directly to the ocean. Nuclear submarine accidents have also released radioactivity directly into the ocean, but these are negligible in comparison to the amount of radioactivity released in the Fukushima accident. The estimated total radioactivity released from Fukushima is in the range of 340–800 PBq (Steinhauser et al., 2014), whereas the estimated total radioactivity released from nuclear submarine accidents is <370 GBq (IAEA, 2001; Hu et al., 2010).

A relatively large ^{129}I/^{127}I signal was observed immediately following the Fukushima accident in the ocean along the east coast of Japan. Suzuki et al. (2013) reported that before the accident, seawater ^{129}I near Fukushima was $0.94–1.83 \times 10^7$ atoms/l, while after the accident, ^{129}I increased to $1.08–89.8 \times 10^7$ atoms/l, with an average of 9.3×10^7 atoms/L, approximately 8 times higher than before the accident. Hou et al. (2013) measured an ^{129}I/^{127}I ratio of 2.2×10^{-9}, in surface seawater collected 40 km offshore the Fukushima site. Guilderson et al. (2014) estimated that the amount of ^{129}I directly released into the ocean was ~1 kg. As regards atmospheric transport, Herod et al. (2013) measured ^{129}I/^{127}I ratios in precipitation in Vancouver and Saturna Island, Canada immediately following the Fukushima accident and showed that ratios of up to 11×10^{-9} were observed about 6–10 days after the accident. They further noted variable ^{129}I increases in precipitation during the following 9 months.

How this seawater with elevated ^{129}I levels is transported, and how long it takes for the anthropogenic ^{129}I to become diluted with naturally low ^{129}I seawater is still unknown. Since the North Pacific Gyre provides a potential pathway for surface seawater from the east coast of Japan to cross the Pacific, we collected seawater samples weekly- biweekly from the Scripps Pier, La Jolla, California. We expected that by establishing an ^{129}I time series, we might be able to identify a pulse of anthropogenic ^{129}I that corresponded to the Fukushima accidental release. At the same time, this study provided us with an opportunity to quantify the amount of anthropogenic ^{129}I that is currently being added to the Pacific Ocean from the Hanford site, at the mouth of the Columbia River.

2. Materials and methods

Seawater samples were collected at weekly to biweekly intervals at Scripps Pier (32°52′N, 117°15.4′W), University of California San Diego, from an intake pipe 10 m below the surface. Two iodine extraction methods were used in this study.

1. The first method followed Biddulph et al.,(2006). The seawater sample (500 ml) was filtered with a 0.22 μm membrane filter. Before starting the iodine extraction process, H_3PO_4 and $NaHSO_3$ were added to convert all iodate into iodide (I^-). To this solution, 3 to 5 ml of $CHCl_3$ was added in a separatory funnel, and 5 drops of 1 M $NaNO_3$. The sample was then shaken vigorously and time was allowed for the immiscible settling of $CHCl_3$, at the bottom of the funnel. The $CHCl_3$ was decanted into a 20 ml glass scintillation vial and 10–15 mg Ag powder was added to the solution. This process was repeated 5 times and the resulting solution was allowed to react overnight. The $CHCl_3$ was then evaporated under a fume hood. The dried Ag + AgI residue was rinsed with distilled water 3 times and dried.
2. The second method is modified from Hou et al., 2010. After filtering with a 0.22 μm membrane, $NaHSO_3$ and HCl were added to acidify the water to pH of 1–2. The water sample was then passed through an AGI-X4 anion exchange column, which was washed with H_2O and 0.5 M $NaNO_2$, and eluted with 2 M $NaNO_2$. Centrifuge and

coprecipitation steps were then performed to get AgI and AgCl to coprecipitate.

Once dry, the samples were pressed into copper cathodes for measurement by accelerator mass spectrometry (AMS). Measurements were performed at the University of Arizona with a 3 MV NEC Pelletron accelerator. The +4 charge state was selected at a terminal voltage of 3 MV with a transmission of 10%. Errors include statistical uncertainties and a random machine error of 4.3%. We used a chemical process blank for the blank correction. The blank produced about 0.1 counts/s while the seawater signal yielded about 2 counts/s. Samples were normalized and calibrated with 2 standards with $^{129}I/^{127}I$ of 63.57 $\times 10^{-12}$ and 8.38×10^{-12}. All the samples were measured for 200 s, repeated 6 times for a total counting time of 1200 s. The $^{129}I/^{127}I$ data show that for the same period of seawater collection, the two methods give similar $^{129}I/^{127}I$ results.

3. Results and discussion

Our $^{129}I/^{127}I$ data show that from June 2011 to May 2018, $^{129}I/^{127}I$ in seawater off La Jolla, California coast varied from 40 to 80×10^{-12} with a variable seasonal cycle with relatively high values in the Fall and lower values in the summer (Fig. 1 and Supplementary Table 1). This effect is clear in 2011, 2012, 2013 and 2017, but less obvious in the data for other years.

We note these values are within the range for the California current system measured by Guilderson et al. (2014). We can also use the known value of iodine in surface seawater (Tusnogai and Henmi, 1971) to show that these ratios are equivalent to ^{129}I concentrations of 0.75 to 1.5×10^6 atoms/l. We can compare these values to the expected input of ^{129}I from the Fukushima NPP accident. To do this, we need to look at the transport of other radionuclides across the Pacific Ocean. Smith et al. (2015, 2017) measured ^{137}Cs along a sampling line (Line P) from the British Columbia coast (approximately 1300 km). They observed an increase of approximately 6-8 mBq/l ^{137}Cs during the period up to 2014. We can assume that this seawater would eventually travel down the California coast. However, unpublished data available online (http://www.ourradioactiveocean.org/results.html) show minimal ^{137}Cs along the southern California coast (<2 Bq/m^3) in recent measurements. Using the activity ratio from the radioactive waters measured by Povinec et al. (2013) and others off the Japanese coast for 129/137 of 0.5–5 $\times 10^{-6}$, we can estimate that the ^{129}I signal equivalent to the measured ^{137}Cs on the west coast would be equivalent to 0.3–1.2 $\times 10^6$ ^{129}I atoms. Therefore, a signal of this magnitude should be observable at San Diego. However, from the data shown in Fig. 1, it is not clear that any excursion attributable to the Fukushima event can be convincingly determined. In order to assess this question further, we also need to evaluate other possible sources of ^{129}I.

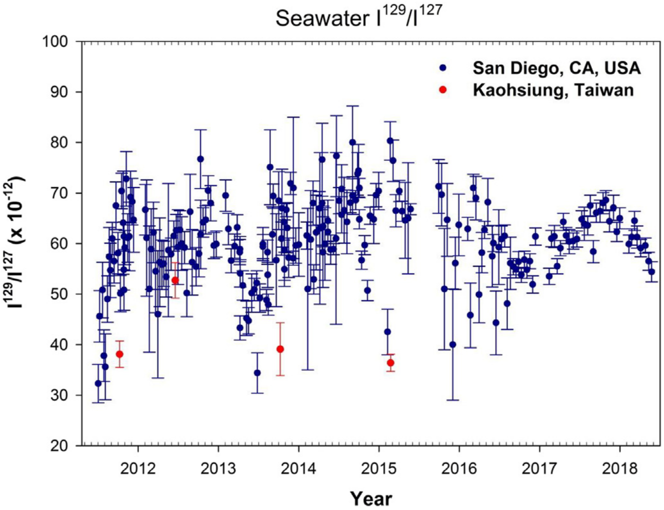

Fig. 1. Seawater $^{129}I/^{127}I$ data from June 2011 to May 2018 in San Diego, CA, USA and Kaohsiung, Taiwan, on a monthly scale.

3.1. Pacific Ocean ^{129}I background value

Before the Fukushima accident, anthropogenic ^{129}I sources in Pacific Ocean waters were derived from nuclear weapons testing that took place in the 1950s–1960s, a Japanese nuclear fuels reprocessing plant, and the Hanford (Washington, USA) nuclear facility (Suzuki et al., 2008). Overall, the Pacific $^{129}I/^{127}I$ background value is ~30×10^{-12} while within the California Current System it is ~50×10^{-12} (Guilderson et al., 2014). This higher amount of ^{129}I could be coming from the Hanford site in Washington, USA. The Columbia River has an average flow rate of 7252 m^3/s (Moran et al., 2002) and is the largest river in North America by volume that discharges into the Pacific Ocean. Suzuki et al. (2008) reported that the Hanford site had released considerable amounts of ^{129}I, about 266 kg, into the atmosphere from 1944 through 1972. Within the Hanford separation area water wells, concentrations of 10^{10}–10^{17} atoms/l ^{129}I have been reported (Brauer and Rieck, 1973). The Washington State Department of Health reported that during 1944–1972, 1778 Tbq of ^{131}I was released into the Columbia River. This implies a contribution of around 0.78 Mbq of ^{129}I released to the Columbia River, if we adopt a ratio of the fast fission yields ($^{129}I/^{131}I$) = 1.03/3.36 (https://www-nds.iaea.org/sgnucdat/c3.htm).

Kilius et al. (1994) collected 13 different species of seaweed sample at 15 sites near the Columbia River mouth in the Pacific Ocean and in the Columbia River Estuary; $^{129}I/^{127}I$ levels reached 176.3×10^{-11}. It is likely that this elevated ^{129}I is still influencing the adjacent river and groundwater system, as well as the nearby ocean. To better understand the possible ^{129}I source from the Hanford site, 15 river water and seawater samples in Oregon and Washington, USA were also collected and analyzed for our study (Fig. 2). High levels of $^{129}I/^{127}I$ were found in several river water samples (Tables 1 and 2). The highest $^{129}I/^{127}I$ water sample that we measured came from the Columbia River, near the Hanford site. The $^{129}I/^{127}I$ reached 5.31×10^{-8}, which is >1000 times higher than the California Current System background value. Also, $^{129}I/^{127}I$ values decrease rapidly with distance, from the Hanford site. At the mouth of the Columbia River, $^{129}I/^{127}I$ is 5.03×10^{-8}. As river water mixes with ocean water, ^{129}I is diluted significantly by seawater. In a seawater sample collected 100 miles south of the Columbia River mouth, at Cannon Beach, $^{129}I/^{127}I$ is only ~75×10^{-12}. This effect can also be seen in the difference in $^{129}I/^{127}I$ between the coastal Long Beach, WA sample and the river sample from Astoria, OR.

A similar situation north of the Hanford site along the Columbia River is observed. In Lake Washington, where the water source is groundwater that may be contaminated by the Hanford site, $^{129}I/^{127}I$ is ~2500×10^{-12}, and just a small distance to the north, at Puget Sound, the ocean water $^{129}I/^{127}I$ decreases to ~100×10^{-12}.

We can estimate the 129I concentrations in atoms/l from the river water samples, given the iodine concentration in the Columbia River measured by Moran et al. (2002). Using these values, the ^{129}I concentrations vary from to 4.0 ± 0.2 to 42.1 ± 1.6 10^7 atoms/l, or from 26 to 275 times the average ocean value. Using these estimates and the flow rate of 7252 m^3/s given by Moran et al. (1982), we calculate an inflow rate of ^{129}I from the Columbia River into the Pacific Ocean of 9 to 95×10^{21} atoms/yr, or 0.01 to 0.13 GBq ^{129}I/yr.

We conclude that the contribution of ^{129}I to the California Current System from the Hanford site is measurable, and may explain the relatively elevated regional Pacific background value of 50×10^{-12}.

3.2. Intra-annual ^{129}I variations

From June 2011 to May 2018, $^{129}I/^{127}I$ in seawater off La Jolla, California coast varied from 40 to 80×10^{-12}, which is similar to the Pacific

Fig. 2. Location of sampling sites in the Hanford vicinity.

background value, with a minor overprint from the Hanford site. This result disagrees with a report by Stan-Sion et al. (2015), who suggested a significant $^{129}I/^{127}I$ increase occurred in the late spring of 2013. We found no evidence for enhanced ^{129}I and their reported value of ~5 × 10^{-9} is inconsistent with all other published values from the Fukushima accident (Hou et al., 2013; Guilderson et al., 2014). To better investigate the variation of ^{129}I through time at Scripps Pier, we compared our ^{129}I data with chlorophyll, salinity, and temperature (Fig. 3). Chlorophyll is often used as an indicator of upwelling, and ^{129}I should be very sensitive to upwelling as well. However, the data in Fig. 3 do not show any straightforward correlation between the chlorophyll time series and our $^{129}I/^{127}I$ time series. The $^{129}I/^{127}I$ time series do not follow the salinity and temperature time series trends either. One possibility to explain these differences is that ^{129}I and ^{127}I represent different reservoirs. We expect that the differences involve biological processes, since inorganic iodide-iodate isotopic exchange is very slow (Myers and Kennedy, 1950). Volatile iodine species, especially methyl iodide (CH_3I) rapidly exchange iodine between the ocean and atmosphere

Table 1
$^{129}I/^{127}I$ results of samples from Pacific Northwest locations ($\times 10^{-12}$).

Sampling sites	June 2014	October 2014	January 2018	March 2018
Victoria, BC, Canada (48° 25′21″N, 123° 22′9″W)		90.6 ± 4.6		
Puget Sound, WA (47°34′47″N, 122°24′32″W)	101 ± 3	69.2 ± 6.8		67 ± 2.2
Lake Washington, WA (47°45′27″N, 122°15′53″W)	2108 ± 93			
Lake Washington, Medina, WA (47°36′54″N, 122°14′21″W)		2920 ± 170		
Long Beach, WA (46°20′24″N, 124°04′17″W)		475 ± 26		
Columbia River (Maritime Museum), Astoria, OR (46°11′24″N, 123°49′17″W)		5030 ± 220		
Columbia river, Rooster Rock State Park, OR (45°32′50″N, 122°14′10″W)	53,100 ± 2000			27,400 ± 430
Columbia river, OR (45°38′55″N, 121°55′15″W)			11,520 ± 190	
Multnomah Creek, OR (45°34′34″N, 122°06′56″W)	4570 ± 320		5200 ± 140	
Cannon Beach, OR (45°53′40″N, 123°57′51″W)	72.7 ± 5.2	77.4 ± 6.1		145 ± 6
Fogarty creek, OR (44°50′19″N, 124°03′00″W)	1000 ± 90			
Depoe Bay, OR (44°48′52″N, 124°03′47″W)	87.5 ± 3.1	91.6 ± 4.6		91.6 ± 4.6
Heceta River, OR (44°08′08″N, 124°07′34″W)	247 ± 25			
East Hood River, OR (45°29′47″N, 121°33′46″W)			5090 ± 130	
Log Boom Lake Wash, OR ()				2065 ± 46

Table 2
Estimated ^{129}I concentrations (atoms/l) from Pacific Northwest locations.

Sampling site	June 2014	October 2014	January 2018	March 2018
		(10^6 atoms/gl)		
Victoria, BC, Canada (48° 25′21″N, 123° 22′9″W)		1.69 ± 0.09		
Puget Sound, WA (47°34′47″N, 122°24′32″W)	1.89 ± 0.05	1.29 ± 0.06		1.25 ± 0.04
Lake Washington, WA (47°45′27″N, 122°15′53″W)	7–23			
Lake Washington, Medina, WA (47°36′54″N, 122°14′21″W)		7–23		
Long Beach, WA (46°20′24″N, 124°04′17″W)		8.87 ± 0.05		
Columbia River (Maritime Museum), Astoria, OR (46°11′24″N, 123°49′17″W)		39.9 ± 1.8		
Columbia river, Rooster Rock State Park, OR (45°32′50″N, 122°14′10″W)	422 ± 16			217 ± 3
Columbia river, OR (45°38′55″N, 121°55′15″W)			91.4 ± 1.5	
Multnomah Creek, OR (45°34′34″N, 122°06′56″W)	36.3 ± 2.5		41 ± 11	
Cannon Beach, OR (45°53′40″N, 123°57′51″W)	1.36 ± 0.10	1.45 ± 0.11		2.7 ± 0.1
Fogarty creek, OR (44°50′19″N, 124°03′00″W)	18.7 ± 1.7			
Depoe Bay, OR (44°48′52″N, 124°03′47″W)	1.63 ± 0.06	1.71 ± 0.09		1.71 ± 0.09
Heceta River, OR (44°08′08″N, 124°07′34″W)	2.0 ± 0.2			
East Hood River, OR (45°29′47″N, 121°33′46″W)			11.9 ± 3.0	
Log Boom Lake Wash, OR ()				5–16

Estimates are based on the iodine concentrations in the Columbia and Hood River (1.7ug/l; Moran et al. 2002), average surface sea water (and 0.5ug/l; Tusnogai and Henmi, 1971), and an estimated range of 0.5-1.7ug/l for Lake Washington.

(Saiz-Lopez et al., 2012). Once the marine iodine is moved into the atmosphere by sea spray, it can travel as aerosols until it is rained out. Iodine has ample opportunity to be removed from the surface waters of the California Current as volatile species and are carried by prevailing winds over the continent. There is no doubt that ^{129}I will be very sensitive to upwelling, since there is an order of magnitude more ^{129}I in the surface as compared with deep waters, but the signal will also be affected by removal by volatile species through air-sea interaction. This process is affected primarily by wind speed (Fuge and Johnson, 1986). We have also done an initial evaluation of the periodicity of the ^{129}I data, which suggest a quasi-annual signal. The prime component appears to be ~0.9 year but there may also be modulation of the signal on a time scale of several years, as evident from Fig. 1. Detailed analysis of this time series will require improved evaluation of the data, which we expect to publish in future.

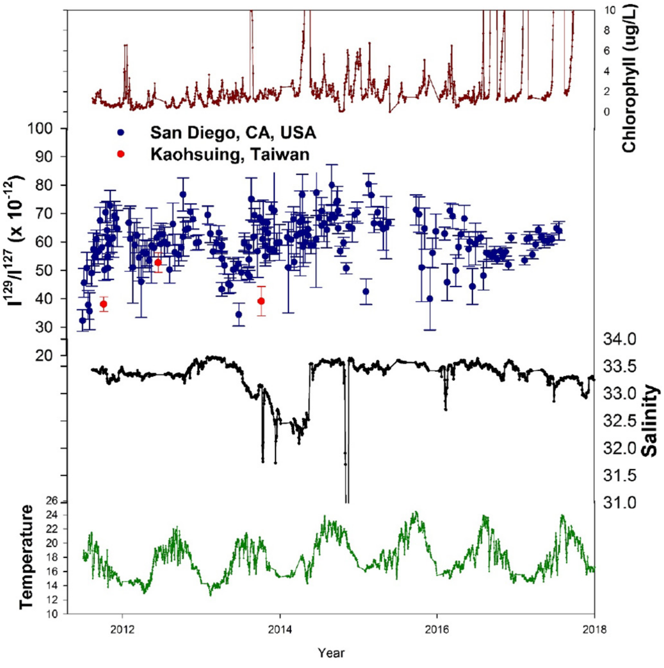

Fig. 3. Scripps Pier chlorophyll, salinity and temperature data.

3.3. Comparing ^{129}I with model estimates

There have been several model results for the transport of radionuclides from Fukushima. Han et al. (2013) estimated 3.2 to 3.9 years for Fukushima radiogenic material to reach the west coast of the U. S. Povinec et al. (2013) suggested that Fukushima signal will arrive at the U. S. west coast in 4–5 years which implies 2015–2016.

We used the OSCURS model to estimate this timing (Bonjean and Lagerloef, 2002; see http://www.pfeg.noaa.gov/products/las/OSCURS.html). Among several runs using different initial locations and dates, from 22nd March to 20th April 2011, only a few results predicted that the water would reach the Eastern Pacific (Fig. 4). This reflects in part the sensitivity to initial model parameters, such as the starting location, which can only be selected as the number of miles offshore. One of the OSCURS model results indicated that water from Fukushima, Japan could reach the Northeast Pacific coast by May 2014. This model result is supported by some reports of debris arriving along the northwest coast of the United States that year.

Although the OSCURS model shows that water from Fukushima, Japan could reach the Northeast Pacific coast in May 2014, we have not observed any significantly elevated ^{129}I/^{127}I values from Scripps Pier up until July 2017. Rossi et al. (2013) also predicted that contaminated (^{137}Cs) water should reach the northwestern American coast at 45°N in 2014, based on a regional eddy-resolving simulation. They also suggested that these waters would reach the Californian coast sometime later (2016–2025), due to the offshore Ekman drift at the coast, which is associated with coastal upwelling. In addition, it was recognized that mixing with relatively depleted upwelled water could obscure the signal through dilution. This model may offer an explanation for our experimental observations. The other possible reason could be that the elevated ^{129}I seawater had already become thoroughly diluted through mixing as it traveled across the ocean and is therefore not detectable above the background level.

4. Conclusion

Overall, the Northeastern Pacific has a higher ^{129}I/^{127}I background value of 50 × 10^{-12} than other parts of the Pacific which are 30 × 10^{-12}. This higher background level is due to radiogenic iodine input from the decommissioned Hanford site. Since the accidental release at the Fukushima Daiichi Nuclear Power Plant, radionuclides have been transported and deposited all over the world. However, after seven years of travel, no contaminated seawater from Fukushima disaster has yet reached La Jolla, San Diego, USA. Although OSCURS

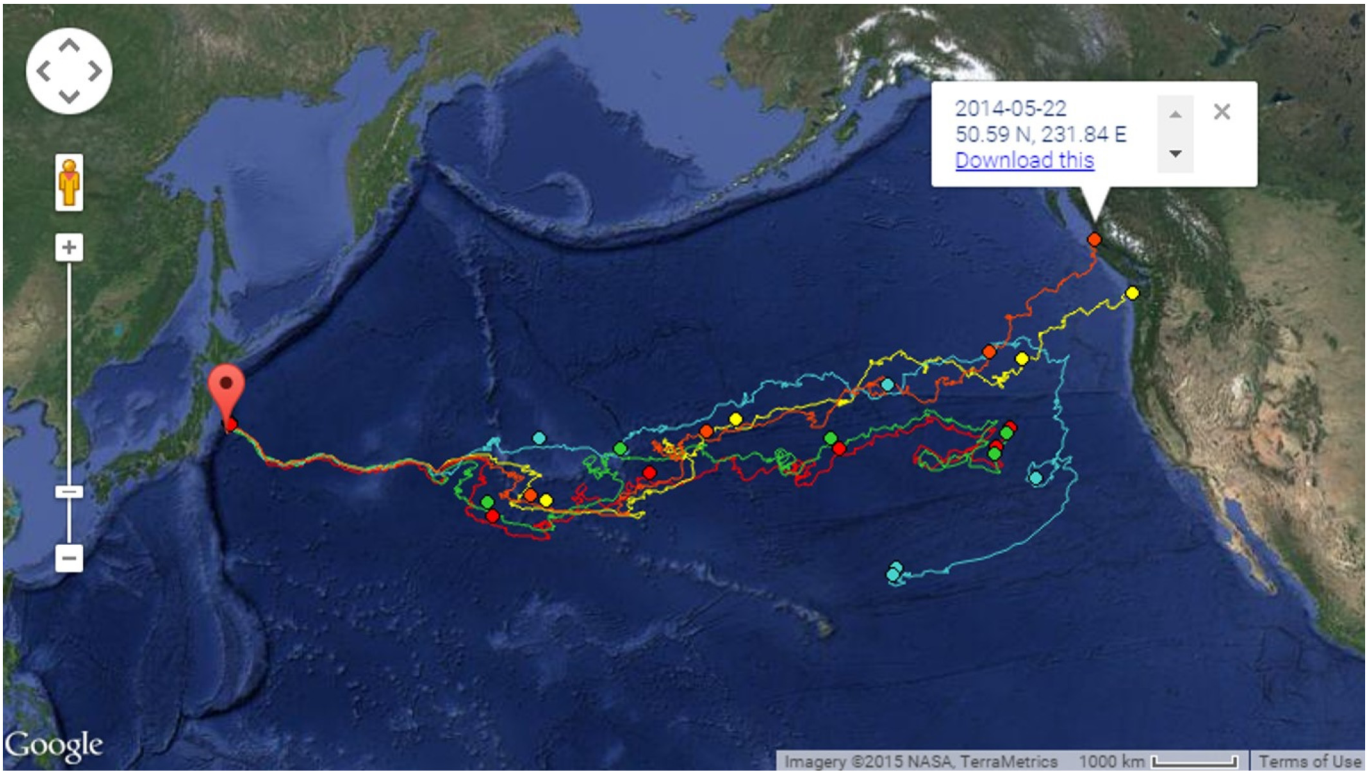

Fig. 4. OSCURS model simulation.

model results are limited by a range of possible initial parameters, such as starting time and precise location, one model result indicates that water from Fukushima, Japan could reach the Northeast Pacific coast in May 2014. This agrees with a previous model study of Han et al. (2013), that suggested it would take 3.2 or 3.9 years for the Fukushima radiogenic material to reach the U. S. west coast. However, as we did not observe a clear ^{129}I/^{127}I pulse in our monitoring, it appears that the currents carrying the excess ^{129}I either did not travel to La Jolla, or the excess ^{129}I was diluted in transit, possibly by upwelled water. This may allow us to set an upper limit on the amount of ^{129}I detectable at the Scripps Pier. Improved analysis of the existing dataset along with new measurements may allow us to separate that signal from the other contributions to the variability evident in Fig. 1.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.06.372.

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