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The 129-Iodine content of subtropical Pacific waters: impact of Fukushima and other anthropogenic ^{129}I sources

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Abstract

Results obtained from a dedicated radiochemistry cruise approximately 100 days after the 11 March 2011 Tohoku earthquake and subsequent disaster at the Dia'ichi Fukushima Nuclear Power Plant show that Fukushima derived radionuclides in the nearby ocean environment had penetrated, on average, to ≤ 250 m depth (1026.5 kg m^{-3} potential density surface). The excess inventory of Fukushima-derived ^{129}I in the region ($\sim 150\,000$ km^2) sampled during the cruise is estimated to have been between 0.89 and 1.173 billion Bq (~ 136 to ~ 179 g) of ^{129}I. Based on a tight tracer–tracer relation with ^{134}Cs (or ^{137}Cs) and estimates that most of the excess cesium is due to direct discharge, we infer that much of the excess ^{129}I is from direct (non-atmospheric deposition) discharge. After taking into account oceanic transport, we estimate the direct discharge off Fukushima to have been ~ 1 kg ^{129}I. Although this small pulse is dwarfed by the ~ 90 kg of weapons-testing derived ^{129}I that was released into the environment in the late 1950s and early 1960s, it should be possible to use Fukushima derived ^{129}I and other radionuclides (e.g., ^{134,137}Cs) to study transport and entrainment processes along the Kuroshio Current.

1 Introduction

Events related to the Tohoku earthquake and subsequent tsunami on 11 March 2011 resulted in catastrophic damage to the Dai'ichi Fukushima Nuclear Power Plants (NPP: 37°25′ N, 141°20′ E), which released a broad suite of radionuclides into the environment via atmospheric plumes and direct discharge into the nearby ocean. Included in the release were radioisotopes of iodine (^{131}I and ^{129}I). Due to its short (~ 8 day) half-life ^{131}I has unique radiological hazards and is routinely measured for radiological assessments. On the other hand ^{129}I, which has a 15.7 million year half-life and is naturally produced via cosmic ray interactions with xenon and as a fission product

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of uranium, has less of a direct radiological hazard, but can be used to retrospectively infer ^{131}I release.

"Anthropogenic" ^{129}I has been produced and dispersed into the ocean via atmospheric testing of nuclear weapons and more recently, via reprocessing of spent nuclear fuel (Snyder et al., 2010 and references therein). Inorganic iodine is a micronutrient that is nearly conservative in seawater exhibiting only a weak depletion in surface waters relative to the deep ocean (e.g., Barkley and Thompson, 1960; Elderfield and Truesdale, 1980) and has a $\sim 3.4 \times 10^5$ yr residence time (Broecker and Peng, 1982). Due to its anthropogenic atmospheric weapons testing input function and long oceanic residence time, ^{129}I can be used as an oceanographic transient tracer, or from reprocessing facilities which act like point sources. The most well known examples takes advantage of the point source input into the North Atlantic, due to reprocessing at the La Hague (France) and Sellafield (England) facilities, to study the ventilation of the deep North Atlantic Ocean (e.g., Edmonds et al., 2001), and transport and mixing between the North Atlantic and the Arctic ocean (Smith et al., 2011 and references therein).

To investigate the input and dispersion of Fukushima derived radionuclides into the western Pacific a dedicated radiochemical and oceanography cruise was undertaken $\sim$ 100 days after the Tohoku earthquake. Here, we report the impact of the Fukushima release on ^{129}I in oceanic waters. Additionally, we present the results of a May 2011 trans-Pacific ($\sim 40^{\circ}$ N) survey of surface water samples.

2 Methods

Seawater samples were collected during two separate cruises (Fig. 1). The first was a trans-Pacific transit (Hong Kong to Long Beach, CA) utilizing a VOS container ship, the OOCL Tokyo (16–29 May 2011). Forty-five (45) surface samples and ancillary hydrographic data (temperature, salinity) were collected. The larger sample set was collected during a dedicated hydrographic and radiochemistry cruise onboard the R/V *Ka'imikai-o-Kanaloa* (KOK) 3–17 June 2011. The cruise plan for KOK1108b has been

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previously described (Buesseler et al., 2012). Briefly, the cruise consisted of thirty-two (32) sampling stations in a grid, 30 to 600 km offshore, to the east of Fukushima. This region is part of the Kuroshio–Oyashio confluence zone (e.g., Talley, 1993; Qu et al., 2001; Ito and Yasuda, 2010) composed of four different water masses (subpolar, subtropical, "Tsugaru", and transitional water) and bounded to the south by the eastward flowing Kuroshio Current.

Sampling onboard the KOK was conducted using paired Niskin bottles on a CTD/rosette. Our samples are from nearshore stations 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32 and offshore stations 5, 6, 7, 8, 9, 10, 11, 12 and focus on horizons less than 400 m water depth, but extending to depths as great as ~ 1000 m at some stations. On the OOCL Tokyo a continuous flow surface seawater line was used for sample collection, after the supply tap was opened and allowed to flow freely for several minutes. For sampling on both ships, bottles (0.5 L, HDPE, acid-cleaned with 2 % nitric acid) were rinsed several times with sample water prior to filling. Bottles were filled, sealed, and taped, and stored in the dark.

^{129}I analyses were made on total inorganic iodine. Iodine was extracted from seawater in a dedicated low-level ^{129}I preparation laboratory, using an adapted version (Tumey et al., 2013) of a commonly-used solvent extraction procedure (Fehn et al., 1992; Moran et al., 1998). Briefly, 0.5 mg of a very low ^{129}I iodine carrier (Woodward Iodine Corporation; $\sim 2\times10^{-14}$ ^{129}I/^{127}I) was added to a 250 mL aliquot of each seawater sample: i.e., a carrier to sample ratio of ~ 40 : 1. Through the addition of sodium sulfite and hydroxylamine hydrochloride dissolved inorganic iodine was reduce to iodide. The resulting iodide was oxidized to molecular iodine by the addition of nitric acid and sodium nitrite. Molecular iodine was extracted into chloroform and then back-extracted into an aqueous solution of sodium sulfite and potassium hydroxide. ^{129}I analyses were made on silver iodide precipitated by the addition of silver nitrate. The precipitated silver iodide was rinsed with MQ water (3X), dried, and mechanically mixed with niobium powder prior to being loaded into individual stainless steel target holders.

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Accelerator mass spectrometric analyses were made at the Center for Accelerator Mass Spectrometry (CAMS), Lawrence Livermore National Laboratory. Targets were analyzed in a sequence similar to that for ^{14}C at CAMS (e.g., Guilderson et al., 2003) and normalized against an in-house prepared dilution of NIST SRM 4949C with Woodward Iodide. Targets were analyzed such that samples with a $^{129}I/^{127}I$ ratio of $\geq 1 \times 10^{-11}$ were counted to ~ 3 % counting statistics. $^{129}I/^{127}I$ ratios for process blanks, prepared by running MQ water run through the full extraction procedure, averaged 3.8×10^{-14} and were not subtracted from unknowns. A small set ($n = 15$) of samples spanning $^{129}I/^{127}I$ ratios from $\sim 5 \times 10^{-12}$ to $\sim 1 \times 10^{-9}$ that were independently prepared and replicated over an eight month window encompassing the measurements of the KOK and OOCL Tokyo samples have a reduced chi-squared of 1.1 and are, on average, statistically indistinguishable from each other (Fig. 2). A subset of these results has been previously reported in Tumey et al. (2013).

In much of the open ocean, total dissolved iodine (speciated between iodate and iodide) is constant to a few percent of a relative concentration of $\sim 60\,\mu g\,L^{-1}$ (Barkley and Thompson, 1960; Elderfield and Truesdale, 1980; Nakayama et al., 1989) and is consistent with iodine concentrations in nearshore waters off Japan with salinity > 30 psu (Zheng et al., 2012). This value has been used in calculating the $^{129}I/^{127}I$ ratios derived from our measurements of seawater samples in this paper.

3 Results

We report results as both ^{129}I activity per m^3 and $^{129}I/^{127}I$ to afford simple comparison to results in other studies. ^{129}I results from the RV KOK1108b cruise and OOCL Tokyo are graphically shown as a function of potential density ($kg\,m^{-3}$) in Fig. 3a. Lowest $^{129}I/^{127}I$ ratios (^{129}I activities) of $\leq 4.5 \times 10^{-12}$ ($\sim 1.7 \times 10^{-6}\,Bq\,m^{-3}$) are observed in the deepest samples analyzed (~ 1000 m; densities ~ 1027.4 $kg\,m^{-3}$). ^{129}I increases with decreasing density (depth) to ~ 1026.9 $kg\,m^{-3}$ where there is a bifurcation. A suite of near constant values at $\sim 3.5 \times 10^{-11}$ ($\sim 1.5 \times 10^{-5}\,Bq\,m^{-3}$) track across (surface)

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densities to ~ 1024.0 kg m^{-3}, whereas the "spine" of the relation from higher densities continues to approximately 1026.4 kg m^{-3} and then the higher ^{129}I values scatter with no correlation to density from ~ 1026.4 to ~ 1024.5 kg m^{-3}. The maximum ^{129}I value is ~ 1.2×10^{-9} (~ 4.5×10^{-5} Bq m^{-3}). Higher ^{129}I values were obtained only for a subset of samples from depths shallower than ~ 250 m.

^{129}I results from the OOCL Tokyo surface samples are generally ~ 3.5×10^{-11} ^{129}I/^{127}I (~ 1.5×10^{-5} Bq m^{-3}) (Fig. 3a) and range from a low of ~ 2.1×10^{-11} (~ 8.3×10^{-6} Bq m^{-3}) to a high of ~ 1.3×10^{-10} (~ 5.2×10^{-5} Bq m^{-3}). There is no apparent relationship with density. The highest sample is from station 17 (37.32° N, 147.82° E) just to the northeast of Fukushima. The following station (18: 38.4° N, 150.45° E) is just under 4×10^{-11} (~ 1.5×10^{-5} Bq m^{-3}). As a function of longitude, ^{129}I values are similar to 145° W and then increase as samples are taken in the California Current System (Fig. 3b).

4 Discussion

4.1 Pre-Fukushima ^{129}I in the Western Pacific

In order to estimate the loading of Fukushima derived ^{129}I it is necessary to determine or estimate the "pre-event" ^{129}I concentration (activity) or ^{129}I/^{127}I ratio. This assessment can be done using internal tracer-tracer (e.g., ^{129}I-^{134}Cs or ^{137}Cs) relationships, "far-field" data from samples that are likely to be uninfluenced by Fukushima directly released effluent (but possibly influenced via atmospheric deposition), and local pre-Fukushima ^{129}I data.

Considerable effort has been devoted to the measurement of ^{134}Cs and ^{137}Cs in the samples obtained during the KOK cruise and an extensive data set has been published (Buesseler et al., 2012). These Cs isotope data, in conjunction with our ^{129}I KOK data, provide the basis for a tracer-tracer assessment of the "pre-event" ^{129}I levels. Because of its 2 yr half-life, any ^{134}Cs observed in the KOK sample suite and,

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more generally across the North Pacific, is Fukushima derived (Aoyama et al., 2012, 2013; Buesseler et al., 2011, 2012 among others). This is in contrast to ^{137}Cs which, due to its 30 yr half-life, has a small residual post-atmospheric weapons testing background of ∼ 1–2 Bq m^{-3} in surface waters (Aoyama et al., 2012, 2013; Buesseler et al., 2011 and references therein). Building on results presented in Tumey et al. (2013), but only using cesium-iodine pairs from niskin bottles collected at the same depth horizon on the same hydrographic cast ($n = 46$ for ^{134}Cs pairs and $n = 69$ for ^{137}Cs pairs), we can see that there is a strong first-order linear correlation between ^{134}Cs (^{137}Cs) and ^{129}I (Fig. 4). Graphically, there is a change in the slope of the relation when Cs activities are < 10 Bq m^{-3} and ^{129}I becomes approximately constant. There is a cluster of un-related data points at very low reported cesium activities. The latter feature may be related to the ∼ 1.5 Bq m^{-3} detection limits for the cesium radionuclides (Buesseler et al., 2012). The former feature: near constant ^{129}I for ^{134}Cs (and ^{137}Cs) < 10 Bq m^{-3}, although possibly a reflection of differential input is very likely the consequence of dilution of Fukushima effluent with a fixed ^{129}I/^{134}Cs (or ^{129}I/^{137}Cs) content diluted in background seawater. Using all of the reported WHOI data, the projected zero ^{134}Cs activity intercept has a ^{129}I/^{127}I ratio of 1.3×10^{-11} (95 % CI: $1.26–1.34 \times 10^{-11}$) or 4.5×10^{-6} Bq m^{-3} (95 % CI: $4.29–4.67 \times 10^{-6}$). If we restrict the analysis to results that are ≥ 1.5 Bq m^{-3} the intercept is 2.4×10^{-11} (95 % CI: $2.30–2.48 \times 10^{-11}$) or 8.6×10^{-6} Bq m^{-3} (95 % CI: $8.13–9.02 \times 10^{-6}$). Decay correcting the ^{134}Cs data from the reported 6 April 2011 reference of peak Fukushima input into the ocean to 11 March 2011 the date of the earthquake does not lead to a significant difference in these ^{129}I-^{134}Cs tracer-tracer intercept calculations of the "pre-event" ^{129}I concentration (activity) or ^{129}I/^{127}I ratio.

Although we expect that some portion of the OOCL Tokyo surface samples could be impacted by Fukushima derived ^{129}I, particularly those directly to the east of Japan, we can compare these "far-field" data to provide an alternative assessment of the "pre-event" ^{129}I levels. This provides insights on the spatial variability of the background ^{129}I, and the potential long-distance impact of Fukushima atmospheric fall-out. The OOCL

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Tokyo sample suite is, for the most part, remarkably consistent. If we consider the section from Hong Kong to the dateline (an arbitrary but useful reference), there is one station (#18: 37.32° N, 147.82° E) that is clearly elevated with a $^{129}I/^{127}I$ of 1.26×10^{-10} and the subsequent station (#19: 38.4° N, 150.45° E) has a ratio of 3.96×10^{-11}. Without those two samples, the average of the remaining Hong Kong – dateline samples is $(2.75 \pm 0.29) \times 10^{-11}$ (1 sigma sd, $n = 19$), which is not significantly different from the ^{134}Cs derived intercept of 2.4×10^{-11}. Heading into the eastern north Pacific surface values are similar: $(3.13 \pm 0.18) \times 10^{-11}$ (1 sigma sd, $n = 6$) to ~ 161° W. Going farther east and into the California Current System (CCS), ratios systematically increase to $(5.26 \pm 0.09) \times 10^{-11}$ for the last three stations (Fig. 3b).

Although not routinely measured on oceanographic samples, Suzuki et al. (2013) presented ^{129}I data from three hydrographic casts off the northeast coast of Japan. One cruise occurred in 2008 (OS08, KNOT station: 154.97° E, 43.97° N) and two cruises in 2009 (SY09, Joban C station: 142.22° E, 36.80° N; and Miyako from an undefined cruise: 145.0° N, 40.0° N). The casts contained numerous discrete samples in the upper 100 m and extended to depths approaching 3000 m. These stations allow us to assess the temporal variability in surface/mixed layer ^{129}I near Japan prior to March 2011. Using the nominally standard 0.125 ($kg\,m^{-3}$) density difference criteria to define the mixed layer, mixed layer values from the three profiles are (expressed as $^{129}I/^{127}I$): $(6.35 \pm 0.10) \times 10^{-11}$ ($n = 2$), $(4.47 \pm 0.12) \times 10^{-11}$ ($n = 3$), $(4.79 \pm 0.37) \times 10^{-11}$; (as $^{129}I\,Bq\,m^{-3}$): $(2.43 \pm 0.17) \times 10^{-5}$ $(1.77 \pm 0.57) \times 10^{-5}$, and $(1.89 \pm 0.14) \times 10^{-5}$, for the KNOT, Joban C, and Miyako stations, respectively. Similar to the KOK data, Suzuki et al. (2013) ^{129}I activities consistent with $\sim 2 \times 10^{-5}$ $Bq\,m^{-3}$ exist to densities approaching 1026.75 $kg\,m^{-3}$, indicating that for these three pre-event cruises ^{129}I was reasonably well-mixed locally to a depth of ~ 250 m. At higher densities (deeper depths), the KOK and pre-event Suzuki et al., data are indistinguishable (Fig. 5).

The three independent methods to determine the pre-event ^{129}I concentration, of which two are completely independent data-sets, have produced estimates that are consistent with each other and indicate that over the recent past surface ^{129}I activities

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have been $1–2 \times 10^{-5}$ Bq m^{-3}. As described by Suzuki et al. (2013) there may be a slight latitudinal dependence of ^{129}I in surface waters of the western subtropical north Pacific, although in the OOCL Tokyo data we do not observe any trend between 22° and 35° north latitude.

4.2 Estimate of ^{129}I/^{137}Cs and ^{129}I/^{134}Cs off Fukushima in June 2011

Using all of the WHOI cesium data we estimate the ^{129}I/^{134}Cs and ^{129}I/^{137}Cs activity ratios (unitless) of a hypothetical Fukushima end-member via Keeling plot analysis (i.e., 1/Cs activity vs ^{129}I/Cs), and obtain 3.9×10^{-7} (95 % CI: $3.74–3.97 \times 10^{-7}$) and 4.1×10^{-7} (95 % CI: 3.95–4.19), respectively. The ^{129}I/^{137}Cs end-member estimate is insensitive to the small (~ 1–2 Bq m^{-3}) post-bomb oceanic background activity. The ^{129}I/^{134}Cs end-member estimate from this data-set is slightly sensitive to the chosen reference date; decay correcting the ^{134}Cs data from the defined (Buesseler et al., 2012) 6 April 2011 reference point (inferred maximum direct discharge date) to the 11 March 2011 date of the earthquake yields an activity ratio of 3.7×10^{-7} (95 % CI: $3.62–3.84 \times 10^{-7}$).

4.3 Estimating the influence of ^{129}I atmospheric deposition

Because ^{129}I has a much lesser potential radiological health impact than ^{131}I or ^{134}Cs, observations of ^{129}I are not made as frequently. Many of the initial measurements by TEPCO, MEXT, and international radiological health effect assessments focused on ^{131}I and ^{134}Cs, and other short half-life radionuclides (e.g., Masson et al., 2012 and references therein). To estimate the potential impact of deposition of ^{129}I in the north eastern Pacific and to constrain its influence on surface waters sampled by the OOCL Tokyo locations where we observe a systematic increase in ^{129}I, we can take advantage of tracer–tracer relations and observations of surface ^{137}Cs and ^{134}Cs (Aoyoma et al., 2012, 2013).

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Our $^{129}I/^{134}Cs$ (or $^{129}I/^{137}Cs$) relation for the KOK sample suite, which we assume to be mainly due to direct release, provides one potential relationship to estimate far-field atmospheric ^{129}I deposition. However, it is likely to be incorrect because the chemistries of cesium and iodine are distinctly different, with iodine being much more volatile whereas cesium has a tendency to be more quickly scavenged onto aerosols and particles. Given that currently there are no direct measurements of ^{129}I in particulates and aerosols with corresponding cesium data, we estimate the atmospheric deposition from data derived from the US International Monitoring System and Comprehensive Test Ban Treaty networks (c.f. Biegaleski et al., 2012; Hoffman et al., 2012).

It should be noted that for the first month after the Tohoku earthquake and subsequent accident, the gaseous activity was ≥ 75 % of the total (gaseous and particulate) measured ^{131}I activity (Masson et al., 2012), consistent with the relative efficacy of scavenging. Transfer times for the conversion of volatile iodine to particulate is on the order of several weeks; hence, gaseous iodine provides a reservoir that can maintain and extend the influence of aerosol deposition of ^{131}I (c.f., Masson et al., 2012; Kristiansen et al., 2013). Thus, the exercise below should be considered one possible end-member based relation.

Post-event aerosol and particulate data document that the activity of ^{137}Cs to ^{131}I (decay corrected to release date) is 1–2 % (Masson et al., 2011), i.e., a $^{131}I/^{137}Cs$ (activity) ratio of ~ 66.7. From measurements around Fukushima NPP the atom/atom ratio of $^{129}I/^{131}I$ of combined dry and particulate (rain-out) deposition is 31.6 ± 9 (Miyake et al., 2012) for a reference (decay-corrected to) date of 15 March 2011 (the date of local maximum atmospheric radioactivity near the NPP), which when converted into an activity ratio is $\sim 4.5 \times 10^{-8}$. Taking the atom ratio back to 11 March 2011 yields an atom ratio of 22.4 and a $^{129}I/^{131}I$ activity ratio of $\sim 3.2 \times 10^{-8}$. Convolving the two relations yields a $^{129}I/^{137}Cs$ activity ratio of approximately 2.1×10^{-6} (referenced to 11 March). And, because Fukushima derived material has a $^{134}Cs/^{137}Cs$ activity ratio of 1 (Biegelski et al., 2013; Buesseler et al., 2011, 2012), also yield a $^{129}I/^{134}Cs$ activity ratio of 2.1×10^{-6} (referenced to 11 March), which is an order of magnitude larger

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than the direct release relation derived above from the KOK sample suite. It is this 2.1×10^{-6} $^{129}I/^{134}Cs$ activity ratio that we use in the following to estimate the influence of atmospheric deposition of ^{129}I.

To estimate the potential impact of atmospheric deposition we utilize the surface VOS NYK ^{134}Cs data of Aoyoma et al. (2012). These samples, although not from the exact location and date as the OOCL Tokyo samples (being collected from 31 March to 17 May), are reasonably contemporaneous. We decay correct the NYK ^{134}Cs data from the reported date of collection, which is the reference date for the decay-correction applied in reporting the ^{134}Cs data (M. Aoyama, personal communication, 2013), to 11 March 2011. Using the above derived $^{129}I/^{134}Cs$ activity ratio and for NYK samples that detected ^{134}Cs, the estimated excess ^{129}I (i.e., added upon the background surface ^{129}I) ranges from very low ^{129}I ($Bq\,m^{-3}$) values of 1.3×10^{-6} ($0.6\,Bq\,m^{-3}$ of ^{134}Cs) to 2.2×10^{-3} ($1028\,Bq\,m^{-3}$ of ^{134}Cs) (Fig. 6). The potential atmospheric deposition of ^{129}I into surface waters implied by the NYK data is, for the most part, similar to that observed by our OOCL Tokyo samples.

4.4 ^{129}I budget and mixing of Fukushima derived ^{129}I into the Western Pacific

As previously discussed, the events at the Dai'ichi Fukushima nuclear power plants released radionuclides into both the atmosphere and, as a consequence of fire fighting and containment efforts, directly released into the coastal ocean. Regardless of the potential chemical fractionation between cesium and iodine into volatiles, the gross correlation of ^{129}I and ^{134}Cs evident in Fig. 4 implies that the samples collected during the June 2011 KOK cruise were dominated by direct release, or the effects of different potential input pathways were effectively homogenized in the coastal and offshore environment during the time between release and sampling. This is consistent with seawater $^{131}I/^{137}Cs$ observations made by TEPCO and MEXT during March and April 2011 (c.f., Buesseler et al., 2011).

Not surprisingly, the penetration and mixing of Fukushima derived ^{129}I is similar to that of cesium (Buesseler et al., 2012) and limited to depths shallower than $\sim 250\,m$

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and densities less than 1026.6 kg m^{-3} (Fig. 7). Similar penetration depths are inferred from two ^{129}I hydrographic profiles from April 2011: KT11-06 station A: 140.83° E, 38.4 N; and B: 143.47° E, 38.28° N (Suzuki et al., 2013). Contrary to these independent methodologies and tracer data, the ^{129}I data of Hou et al. (2013), which were obtained from a sub-set of KOK samples, indicate penetration of ^{129}I to densities approaching 1026.9 kg m^{-3} or approximately 400 m (Fig. 8). Given the consistency between the Buesseler et al. (2012) cesium data and our ^{129}I results from the same cruise, and the consistency between our ^{129}I data and that of Suzuki et al. (2013) we infer that there are unresolved issues with the Hou et al. (2013) ^{129}I data at all depths. Thus, we restrict the discussion of the ^{129}I results and estimated excess ^{129}I budget to our data.

Following the region analysis of Buesseler et al. (2012) we construct average ^{129}I profiles for the nearshore and offshore regions of the KOK cruise (Fig. 9a). For internal consistency we define the pre-Fukushima ^{129}I content using the ^{134}Cs zero-intercept for ^{134}Cs data > 1.5 Bq m^{-3} (^{129}I/^{127}I 2.4×10^{-11}, 0.9×10^{-5} Bq m^{-3}) and, via simple subtraction, calculate the excess inventory as a function of depth (Fig. 9b). The excess inventory would be ~ 10 % lower using the OOCL Tokyo observations (2.75×10^{-11}) and ~ 25 % lower if we chose 3.5×10^{-11} from the near constant ^{129}I values as a function of density (c.f., Fig. 3a) to define the pre-event ^{129}I background. The nearshore region (50 000 km^2) has an estimated excess ^{129}I inventory of 1.31×10^{-2} Bq m^{-2} and that of the offshore region (100 000 km^2) is 5.17×10^{-3} Bq m^{-2}. This leads to an upper estimate excess inventory of ~ 1173 million Bq of ^{129}I (8.38×10^{23} atoms or ~ 179 g) and a lower bound estimate of 890 million Bq (~ 136 g) of ^{129}I. Direct quantification of the uncertainty of this estimate is, due to the potential clustering of profiles and sampling a heterogeneous domain, difficult. Errors quoted in similar studies have been ±30 % or more (Buesseler et al., 2012).

To determine the actual ^{129}I direct discharge release requires a model-based assessment of the mixing (dilution) within, and transport out of the KOK sampled region between the initial release and the cruise, ~ 3 months later. Such estimates are very likely to be model dependent and require independent knowledge of the relative influ-

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ence of atmospheric and direct discharge of ^{129}I. An ocean model forced with NCEP-NCAR reanalysis and satellite altimetry as described in Rypina et al. (2013) and using the KOK sampled region 2 PBq excess inventory of cesium (Buesseler et al., 2012) implies that ∼ 82.5 % of the direct oceanic discharge and ∼ 95 % of the atmospheric deposited Cs had been advected out of the region the KOK sampled by the time of the cruise. The convolution of the two source terms and export indicate that more than 95 % of the excess cesium in the region sampled by the KOK came from direct discharge (Rypina et al., 2013). If the model results are accurate, we can use a similar dilution scaling argument to estimate an admittedly uncertain direct discharge of ∼ 1 kg of ^{129}I. Moreover, if precise and accurate, the cesium modeling assessment allows us to estimate the actual direct discharge ^{129}I/^{134}Cs (and ^{129}I/^{137}Cs) relation from:

$$0.95[^{129}\mathrm{I}/^{134}\mathrm{Cs}]_{\mathrm{direct}} + 0.05[2.1 \times 10^{-6}]_{\mathrm{atm}} = 3.7 \times 10^{-7}_{\mathrm{observed}}$$

Solving yields 2.8×10^{-7} (unitless activity ratio) for ^{129}I/^{134}Cs and 3.2×10^{-7} for ^{129}I/^{137}Cs.

4.5 California coastal current ^{129}I

There were only a few NYK samples in the eastern Pacific that detected ^{134}Cs and thus implicate atmospheric deposition of ^{129}I, whereas the OOCL Tokyo data show that ^{129}I in eastern surface waters is consistently elevated compared to the far western and central Pacific (Fig. 5). Washout of atmospheric ^{129}I by precipitation into western North American watersheds and subsequent run-off into the CCS is a potential avenue for the observed trend in ^{129}I as the OOCL Tokyo came into the CCS and approached Long Beach, CA. Indeed, analysis of seaweed samples collected in April 2011 from Southern California document the presence of ^{131}I inferred to be from Fukushima (Manley et al., 2012). However, these observations and inferences do not preclude a possible North American ^{129}I point source that could influence the ^{129}I content of the CCS, namely the Columbia River (Kilius et al., 1994; Moran et al., 2002). More recent (1998–

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2005) monitoring of ^{129}I in the Columbia river and watershed indicate elevated values near Richland, WA compared to upstream at Priest Rapids Dam (Patton, 2004, 2009), where ^{129}I average concentrations range from $\sim 4.6\times10^{-6}$ Bq L^{-1} to $\sim 1.8\times10^{-6}$ Bq L^{-1}. Flow rates at Priest Rapids are approximately 3000 m^3 s^{-1} (Patton, 2004, 2009) which implies a potential ^{129}I source function of ~ 1.2 to $\sim 3.1\times10^{23}$ atoms yr^{-1}. A simple salinity based two-end-member model using the Richland ^{129}I concentration (0 psu, 1.8–4.6 $\times 10^{-3}$ Bq m^{-3}) and an "unperturbed" average eastern Pacific surface value of 1.2×10^{-5} Bq m^{-3} (33.27 psu) from the OOCL Tokyo data allow us to infer that the CCS maximum values that we observe (2.0×10^{-5} Bq m^{-3}) could be accommodated with only a fraction of a percent of direct dilution of surface ocean waters with Columbia River water. Thus, it is possible that recirculation within the CCS has allowed the ^{129}I signature from the Columbia River to accumulate. Further isolation of the ^{129}I source and overall impact of atmospheric deposition of ^{129}I in eastern Pacific surface waters will require additional samples and radiochemical tracer-tracer analysis that can uniquely distinguish Fukushima derived fall-out from the Columbia River or other potential point sources.

4.6 Penetration of ^{129}I into subsurface water masses

Although the region off Fukushima has, due to the confluence of the Kuroshio and Oyahio currents, more complicated dynamics than the "open" Pacific, we can explore the ^{129}I data in the context of a conventional transient oceanographic tracer. In this region, isopycnals are shallower and slightly compressed relative to that of the open Pacific; i.e., isopycnals deepen to the south and east entering the open North Pacific. Major reference density horizons include 1026.2 kg m^{-3}, which outcrops at 38–40° N during winter and slopes downward to 500 m in the center of the subtropical gyre, and 1026.8 kg m^{-3}, the main density of North Pacific Intermediate Water (NPIW; a well-defined low-salinity, well-oxygenated water mass at 300–700 m depth which spreads across the North Pacific (Reid, 1965; Talley, 1993)). NPIW is unique in that waters of

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this density do not outcrop in the open Pacific; NPIW is ventilated in the Sea of Ohkotsk and near the Kurils with (some) subsequent modification in the Gulf of Alaska (Aydin et al., 1998; Guilderson et al., 2006). At the time of the R/V KOK cruise (~ 100 days post event) we did not observe entrainment or mixing of Fukushima derived radionuclides to densities greater than $\sim 1026.5\,\mathrm{kg\,m^{-3}}$ (Fig. 3a). The major impact of Fukushima nuclides was confined to lower densities due to the rapid expunging (relatively short residence time) of surface waters in the confluence zone and the region sampled by the KOK (e.g., Buesseler et al., 2012; Rypina et al., 2013). Moving forward in time, we anticipate that small residual (relative to the large initial direct discharge that was swept offshore) radionuclides could be mixed into higher densities. This is confirmed by the results of Suzuki et al. (2013), who observe elevated ^{129}I to $\sim 1026.9\,\mathrm{kg\,m^{-3}}$ in samples off Fukushima in September, five months after the accident. This is not to imply that the Fukushima radionuclides have significantly impacted ("labeled") NPIW and waters of deeper densities, just that the enhanced mixing in the Oyahio–Kuroshio confluence region can lead to the local input of some of the Fukushima related products at these densities.

The pre-anthropogenic $^{129}\mathrm{I}/^{127}\mathrm{I}$ of the ocean has, via the analysis of sediments and archived macrophytes, been estimated to be $\leq 1.5 \times 10^{-12}$ (Moran et al., 1998). This estimate is consistent with the deep (sigma-t > 1027.6) results of Suzuki et al. (2013) who report ratios equivalent to 1.2×10^{-12} (±100 %) to as low as 3.5×10^{-13} (±1000 %). The present day open Pacific distribution of anthropogenic or "bomb" ^{129}I, uninfluenced by reprocessing or related activities, as a function of depth (density) should reflect that of a conventional "transient tracer" such as tritium except that (compared to tritium with a ~ 12 yr half-life) there is no appreciable radioactive decay. To demonstrate this point we compare our results and the pre-event data of Suzuki et al. (2013) to tritium observations obtained in 1993 (WOCE P10) and in 2004 from a CLIVAR reoccupation of WOCE P02. Tritium data were obtained from the CLIVAR & Carbon Hydrographic Office (http://cchdo.ucsd.edu/pacific.html). Although from the same general location near Japan, transport and dynamics means that the comparison is obviously not a direct

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comparison of the same water masses at the exact same time. The tritium data are decay corrected to a common reference year of 2010. We chose this year to be close to the 2008 and 2009 pre-event data of Suzuki et al. (2013) and our observations in 2011. For visual clarity we restrict our (KOK) data to non-Fukushima impacted samples and densities greater than 1026.5 kg m^{-3} (Fig. 10). Tritium and ^{129}I data exhibit the same general curvature and describe a typical (vertical) diffusive transient tracer profile with input at the surface (cf. Fig. 10: Kelley and Van Scoy, 1999). The curves indicate approximately the same vertical diffusivities based on tritium and ^{129}I; the shape of the tritium and ^{129}I profiles, as a function of density and if converted into a common depth domain, are similar.

In contrast to these consistent atmospheric testing sourced transient tracer data, ^{129}I data from the central equatorial and western Pacific during a 1997 IAEA cruise (Povinec et al., 2003, 2010, 2013) present a large and regional elevation in ^{129}I above not only pre-anthropogenic ^{129}I/^{127}I ($\leq 1.5 \times 10^{-12}$) but also above the data of Suzuki et al. (2010, 2013) and our own "non-Fukushima influenced" data (Fig. 11). If real, the data from the IAEA 97 cruise indicate a significant perturbation to the North Pacific pre-anthropogenic ^{129}I budget at all depths; i.e., extending to 5000 m. ^{129}I data from stations 2 and 3 as a function of depth (P. Povinec, personal communication, 2013) and stations 6 and 7 (digitized from Fig. 3 presented in Povinec et al., 2012) were placed on density horizons using the corresponding CTD data collected during the cruise on the R/V *Bosei Maru*. We assume that the IAEA 97 cruise stable iodine concentrations were similar to open ocean values. The IAEA 97 cruise ^{129}I values are nearly invariant below 1000 m for the western subtropical north Pacific at ^{129}I/^{127}I equal to $\sim 2.8 \times 10^{-11}$ and those from the low latitude north Pacific are $\sim 5.3 \times 10^{-11}$. Upper ocean samples are also distinguished by their high values of $(5.9 \pm 1.1) \times 10^{-11}$ (1sd, range: 3.9×10^{-11} to 7.8×10^{-11}) for depths less than 800 m (< 1027.0 kg m^{-3}) at stations 2 and 3, and similar densities near Bikini and Einewetak (depths shallower than 500 m) average $(6.6 \pm 2.4) \times 10^{-11}$ (1sd, range: 3.2×10^{-11} to 1.1×10^{-10}).

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To estimate this possible perturbation to the North Pacific pre-anthropogenic ^{129}I budget we simplify the region contained by stations 2, 3, 6, and 7 (Povinec et al., 2003, 2012) to a triangle with an area in excess of 2.4×10^{6} km^{2}. We fit a smoothed polynomial to the merged ^{129}I data presented in Fig. 9 for densities greater than 1026.9 and equal interval interpolate the data with a step of 0.1 at 0.05 density horizons (e.g., 1026.95, 1027.05, 1027.15). For shallower depths, and to account for the small ^{129}I latitudinal dependence in surface waters observed between 36 and 44° N, we use a constant ^{129}I/^{127}I of 2.8×10^{-11}, which is consistent with our OOCL Tokyo data for stations south of 35° N. Differences were then calculated for the corresponding density horizons after averaging/compositing stations 2/3 and 6/7. Excess ^{129}I (as atoms m^{-3}) were calculated and, after translating the density horizons back into the depth domain, integrated. This admittedly coarse assessment yields an excess burden of 17–29 kg ^{129}I relative to our composite post-bomb ^{129}I profile in Fig. 10, or 21–32 kg of excess ^{129}I relative to pre-anthropogenic ^{129}I. This is 20–32X larger than what we estimate the direct release from Fukushima NPP to have been (~ 1 kg). To put this "anomaly" in perspective, the total global release of ^{129}I during atmospheric weapons testing is estimated to have been 90 ± 50 % kg (Snyder et al., 2010). From the available ^{129}I release data it is difficult to ascertain the validity of the proportionally large ^{129}I input implied by the IAEA 97 cruise data, which is equivalent to ~ 30 % of the estimated global fallout from all atmospheric weapons testing. Intriguing as the question of where this regional pulse of ^{129}I may have gone in the intervening decades may be, it is our hypothesis that samples passing through Monaco since at least 1997 and continuing to present have inadvertently picked up ^{129}I from the more common "environmental" samples with high ^{129}I that are handled by the IAEA. Such inadvertent ^{129}I contamination is not unprecedented (Szidat et al., 2000).

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5 Conclusions

We determined the excess burden of Fukushima NPP derived ^{129}I in a ∼ 150 000 km^2 region offshore Fukushima as observed in June 2011, ∼ 100 days after the catastrophe, to be ∼ 136 to ∼ 179 g of ^{129}I. Modeling (Rypina et al., 2013) of the excess ^{134}Cs and ^{137}Cs allows us to infer the total discharge into and through this region to be ∼ 1 kg of ^{129}I with the majority inferred to be via direct discharge. Similar to other Fukushima derived radionuclides, the penetration of ^{129}I was generally relegated to depths shallower than 1026.6 kg m^{-3}, or ∼ 250 m. We estimate the pre-event surface water ^{129}I/^{127}I from ^{129}I-^{134}Cs pairs to have been 2.4×10^{-11} which is similar to surface water samples unlikely affected by Fukushima collected on a trans-Pacific transect: 2.75×10^{-11}.

The seawater samples describe a ^{129}I/^{137}Cs and ^{129}I/^{134}Cs unitless activity ratio of 4.1×10^{-7} and 3.7×10^{-7} respectively, with the ^{134}Cs being decay corrected to 11 March 2011. The ^{129}I/^{134}Cs activity ratio is slightly sensitive to the decay correct reference date. If we use the 6 April 2011 date of maximum discharge (eg., Buesseler et al., 2011), the activity ratio is 3.9×10^{-7} and indistinguishable from ^{129}I/^{137}Cs (95 % CI overlap). Utilizing the ocean model estimate of the relative influence of atmospheric and direct injection allows us to estimate that the direct discharge component had a ^{129}I/^{134}Cs activity ratio of approximately 2.8×10^{-7} and, not surprisingly nearly the same value, 3.2×10^{-7} for ^{129}I/^{137}Cs. The activity ratio of direct discharge is an order of magnitude less than that implied by aerosol and related data: 2.1×10^{-6}.

Surface water samples from in and near the California Coastal Current System have slightly elevated ^{129}I values compared with waters to the west. Samples taken during a May 2011 VOS container ship transit document approximately constant ^{129}I/^{127}I values from Hong-Kong to ∼ 161 W of $\sim 3 \times 10^{-11}$ or less whereas values in the CCS are $\sim 5.3 \times 10^{-11}$. Although it is possible that these slightly elevated values are due to atmospheric deposition of Fukushima releases, a more probable explanation is that the CCS is impacted by ^{129}I from the Columbia River.

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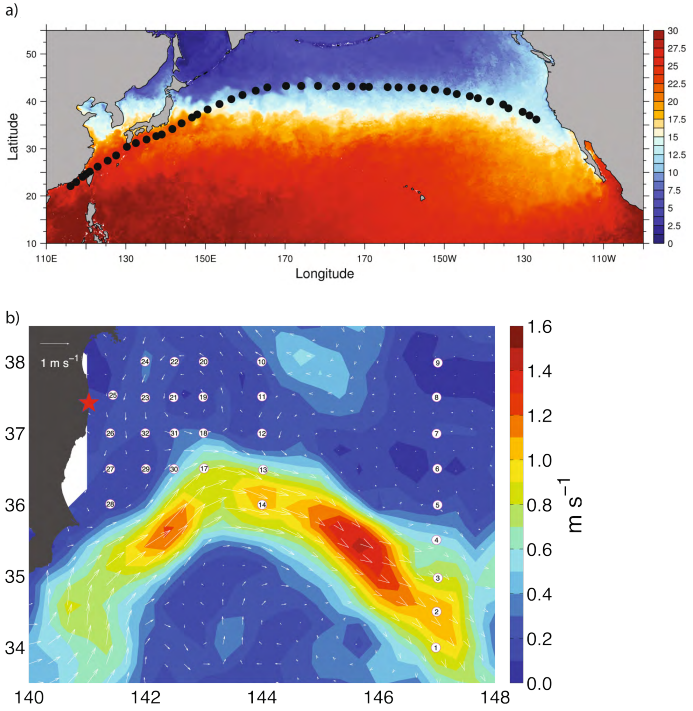

Fig. 1. (a) Surface sample locations collected via the OOCL Tokyo (16–29 May 2011) overlain on level 4 Global high resolution sea surface temperature for 23 May 2011 (http://podaac.jpl.nasa.gov/dataset/JPL-L4UHfnd-GLOB-MUR). The Kuroshio, which in general tracks the zero curl of the wind stress, separates cooler sub-Polar water (blue colors) from warmer subtropical water (orange to red colors). **(b)** Hydrographic profile stations for the R/V *Ka'imikai-o-Kanaloa* cruise 1108b (3–17 June 2011) overlain on the first derivative of sea surface height which clearly delineates the Kuroshio Current as it sweeps eastward off Japan. Arrows are current velocity vectors. Figure modified from http://www.whoi.edu/page.do?pid=67796 and Buesseler et al. (2012).

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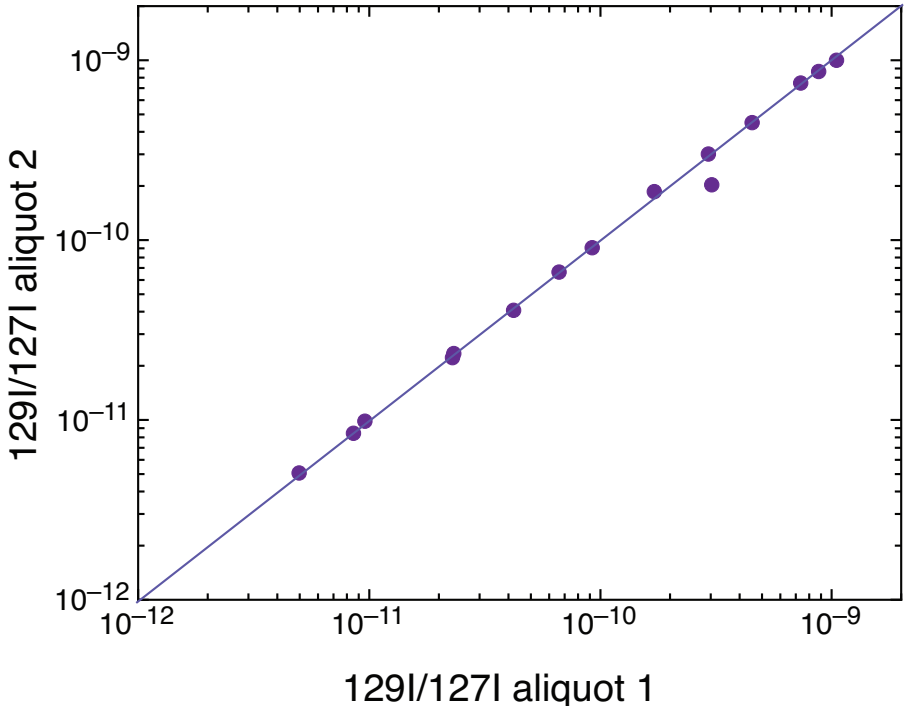

Fig. 2. Independently prepared ^{129}I replicate analyses prepared over an 8 month window are statistically indistinguishable from each other (reduced chi-squared of 1.1).

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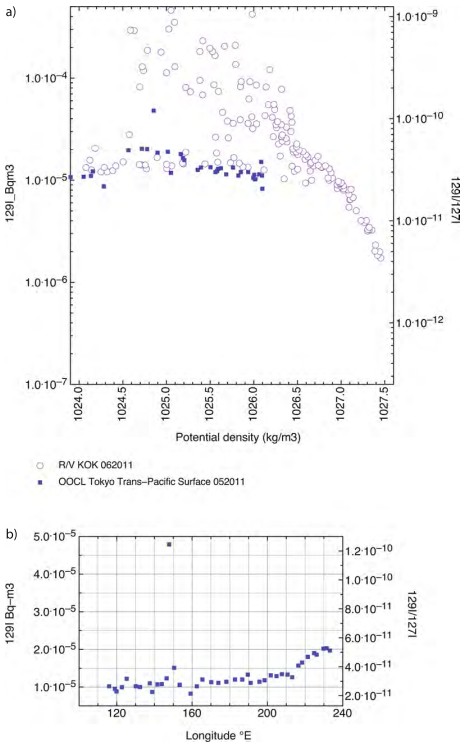

Fig. 3. (a) ^{129}I as a function of potential density from the R/V KOK 1108b (open circles) and OOCL Tokyo samples (filled squares). **(b)** Surface ^{129}I from the OOCL Tokyo as a function of longitude. For clarity error bars are not plotted. For most of the data the uncertainties are the same size as the symbols. Results are shown as ^{129}I Bq m^{-3} and the equivalent approximate ^{129}I/^{127}I ratio.

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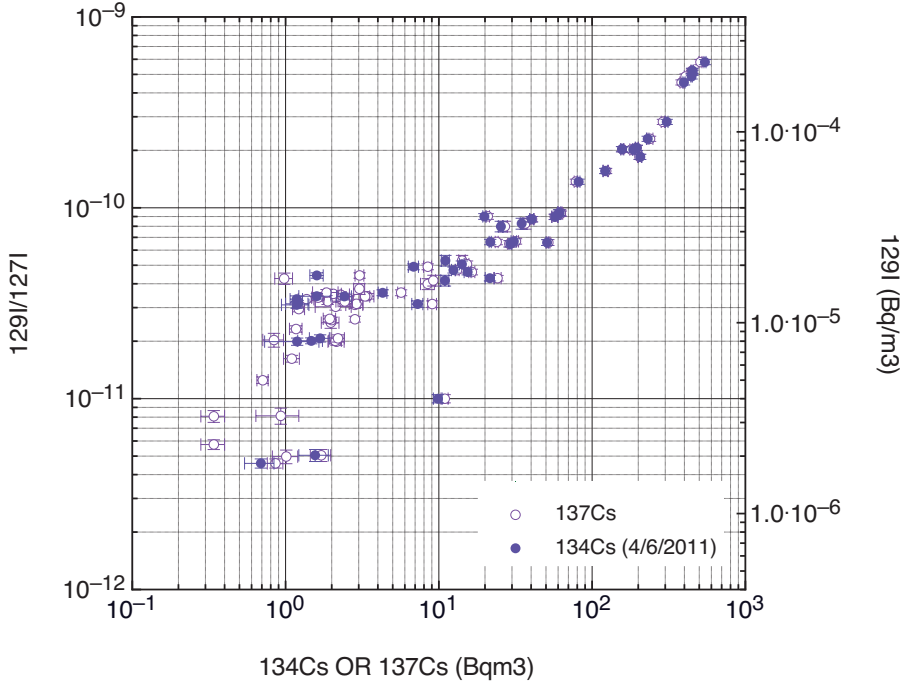

Fig. 4. ^{129}I as a function of ^{134}Cs (and ^{137}Cs) for exact same samples. Cesium data are from Buesseler et al. (2012). ^{134}Cs has a reference (decay-corrected) date of 6 April 2011: the date of the highest direct discharge as indicated by TEPCO and MEXT monitoring.

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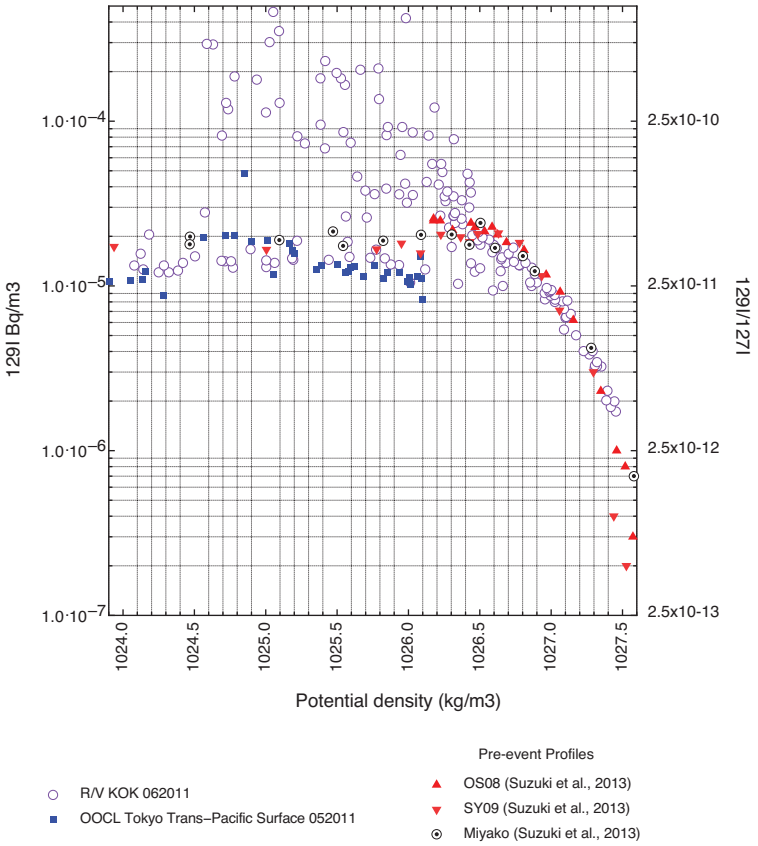

Fig. 5. ^{129}I data from the KOK and OOCL Tokyo and data from three hydrographic stations (Suzuki et al., 2013) that were collected in 2008 and 2009: OS08 (filled triangles), SY09 (upside down triangles), Miyake (bulls eye). For clarity error bars are not plotted. For most of the data the uncertainties are the same size of the symbols. The exception to this includes the lowest values of Suzuki where uncertainties are 100–1000 %.

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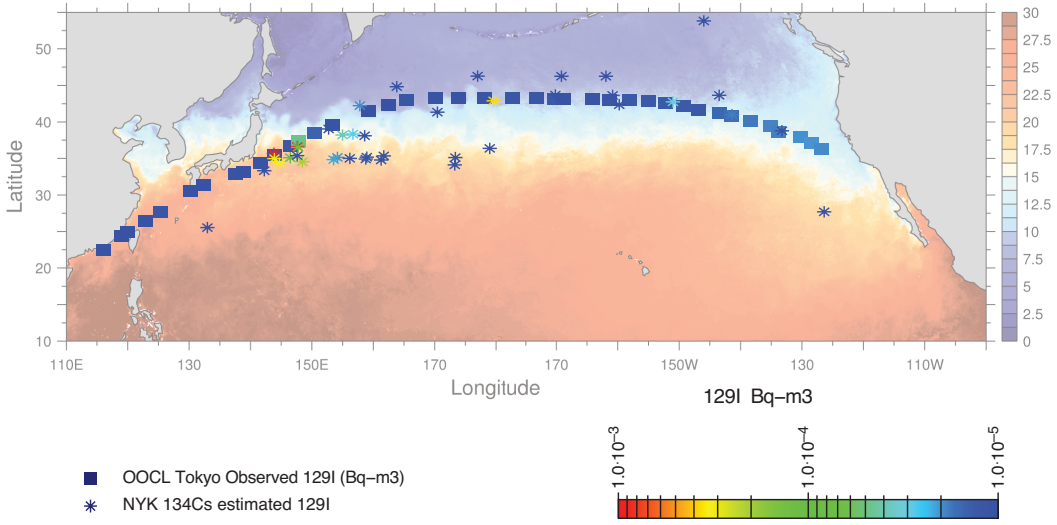

Fig. 6. Estimated ^{129}I via atmospheric deposition (by way of ^{137}Cs and ^{134}Cs) for NYK surface samples (asterisks) collected at about the same time as the OOCL Tokyo (filled squares). NYK cesium data from Aoyama et al. (2012, 2013). The excess ^{129}I estimated via ^{134}Cs has been added to an average surface ocean background as described in the text. Translucent raster 23 May 2011 sea surface temperature whereas discrete points scale with ^{129}I Bq m^{-3}.

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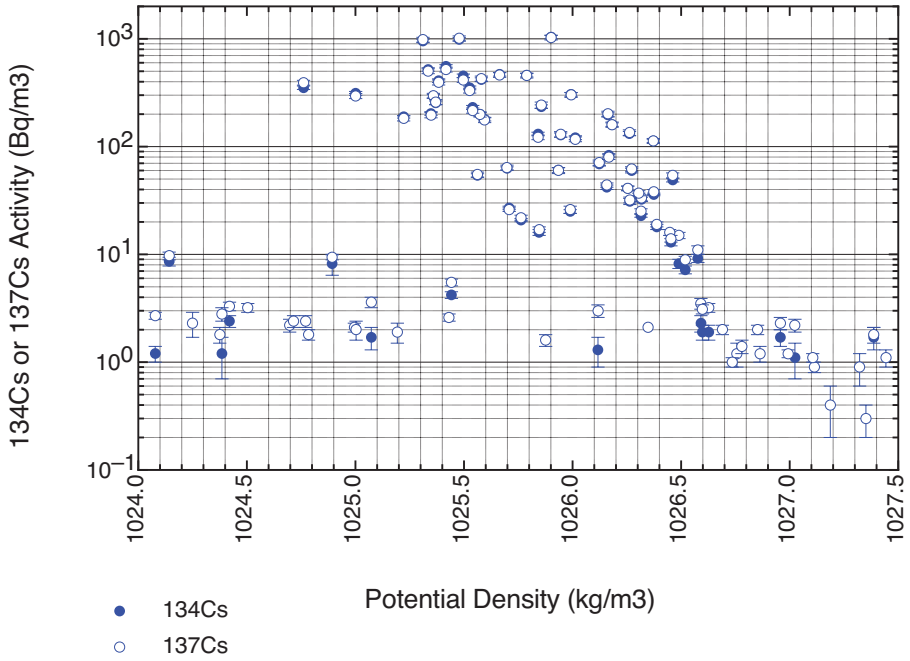

Fig. 7. ^{134}Cs (filled circles) and ^{137}Cs (open circles) as a function of potential density. ^{134}Cs data are referenced (decay-corrected) to 6 April 2011. Not shown are the samples that were non-detects. Data of Buesseler et al. (2012).

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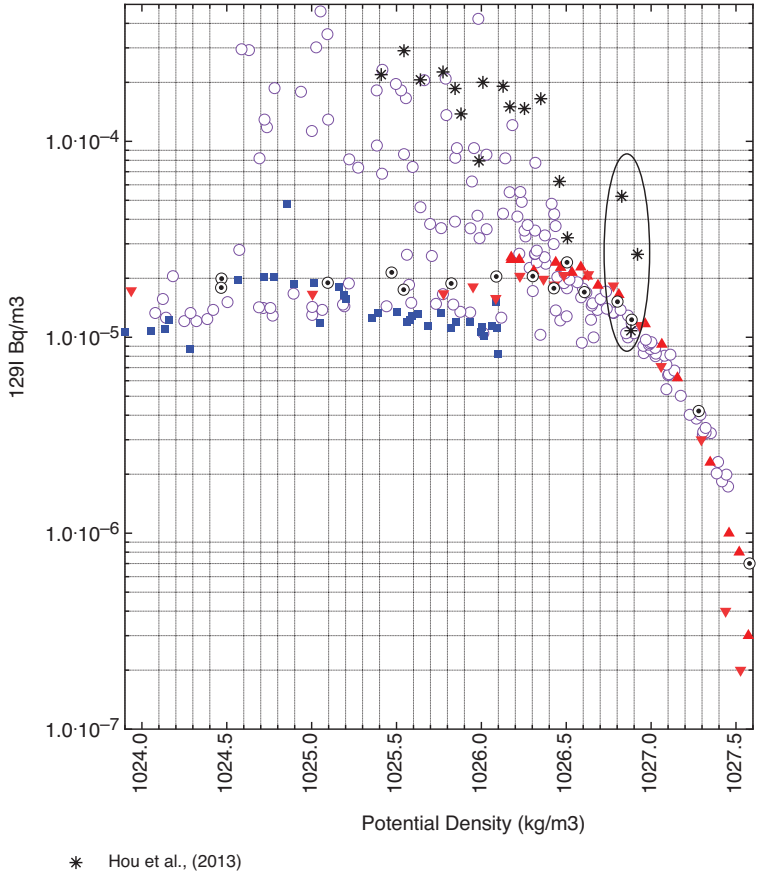

Fig. 8. ^{129}I data from Fig. 5 with those of Hou et al. (2013: astericks) as a function of density. Note the anomalous elevated 129I at 1026.8–1026.9 (kg m^{-3}).

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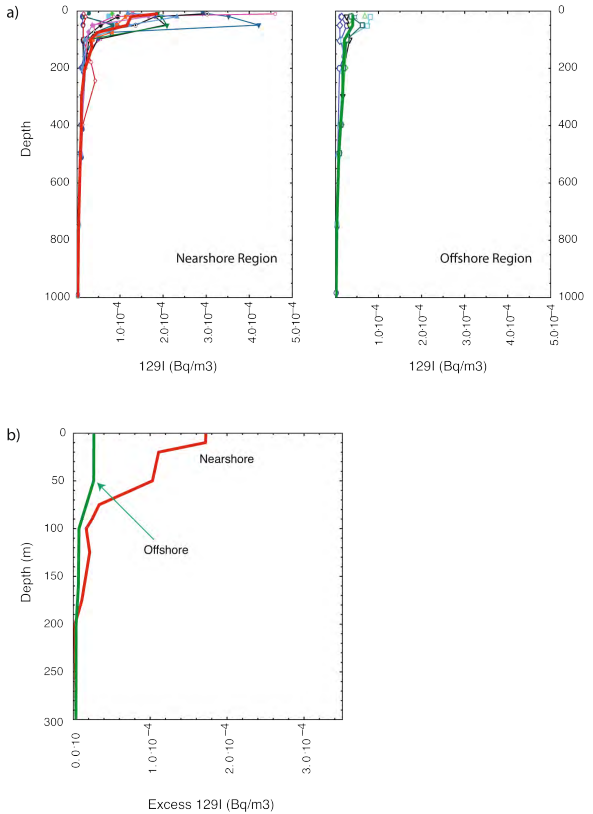

Fig. 9. (a) ^{129}I as a function of depth for all profiles presented for the nearshore (Stations 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32) and offshore regions (Stations 5, 6, 7, 8, 9, 10, 11, 12) as defined by Buesseler et al. (2012). **(b)** Excess ^{129}I for the nearshore and offshore regions for depths above 300 m.

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The 129-Iodine content of subtropical Pacific waters

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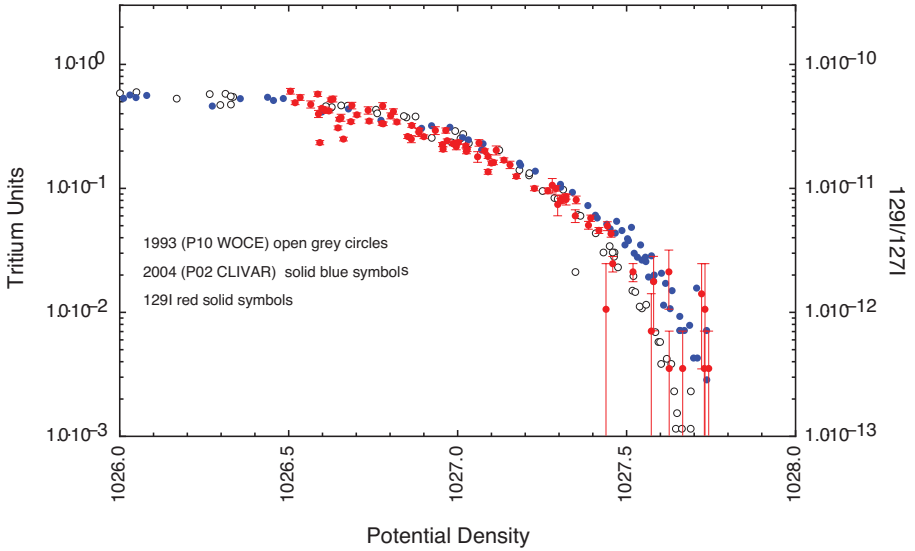

Fig. 10. Tritium and ^{129}I produced during atmospheric weapons testing will have a similar input function and penetration history in the ocean. Pre-Fukushima ^{129}I data, as ^{129}I/^{127}I from three hydrographic profiles to the east of Japan (Suzuki et al., 2013), and data from the R/V KOK for densities greater than 1026.5 kg m^{-3}(filled red circles). Tritium data from selected stations near Japan from WOCE P10 in 1993 (stations 79, 81, 83, 85, 88, 90: open circles) and the CLIVAR reoccupation of P02 in 2004 (stations: 11, 15, 19, 23, 28: filled blue circles). Tritium data have been decay-corrected to a common reference of 2010.

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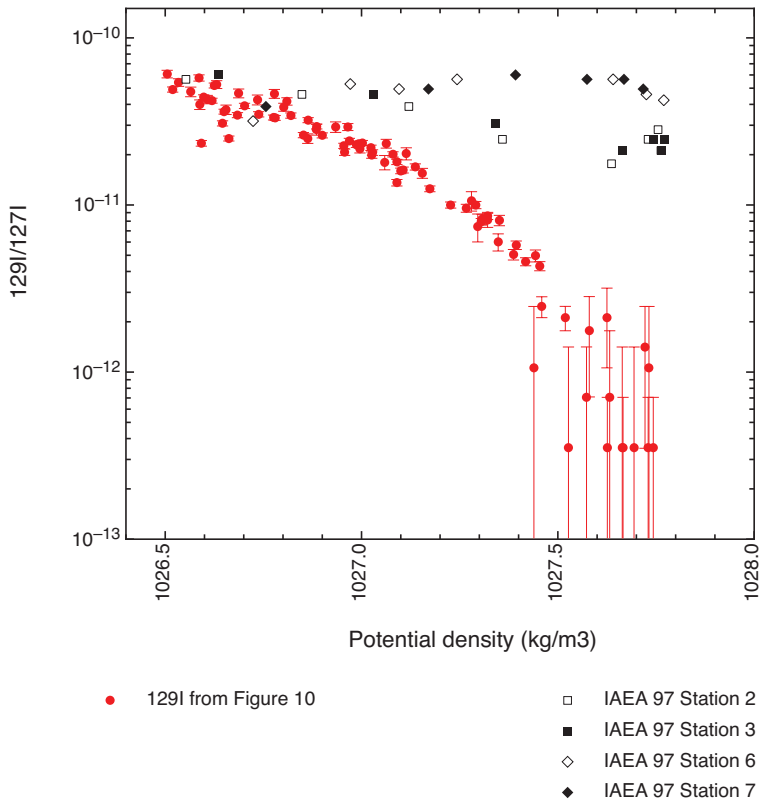

Fig. 11. ^{129}I data from Fig. 10 (filled red circles) with that from four stations during the IAEA 97 cruise presented in Povinec et al., 2010 and a sub-set replotted in Povinec et al., 2013. Note that regardless of location: western subtropical North Pacific (stations 2, 3: squares) or low latitudes near Bikini/Enewetak (stations 6, 7: diamonds) that the IAEA 97 data imply a significant ^{129}I excess at all depths.

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