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Interactive comment on “The 129-Iodine content of subtropical Pacific waters: impact of Fukushima and other anthropogenic ^{129}I sources” by T. P. Guilderson et al.

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Overview statement: We compared our ^{129}I results with the ^{129}I data from three Suzuki et al. (2013) depth-profile stations, OS08, SY09 and Miyako. These stations were occupied prior to the events leading up to the release of radionuclides from Dai’ichi Fukushima and provide a picture of the background ^{129}I in the subtropical North Pacific. Suzuki et al. (2013) also presented three relatively-sparse ^{129}I depth-profiles from hydrographic stations that were occupied about one month before the R/V KOK cruise in early June 2011. These three profiles; KT1106A (4/26/11), KT1106B (4/30/11) and KT1106C (5/1/11), are indistinguishable from our own data and at den-

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sities greater than 1026.5 kg m⁻³ (depths greater than ~250 m) are indistinguishable from the pre-event depth-profile stations (see figure below).

The commenter (Dr. Hou) suggests that the 129I depth-profile data from three of the stations (FS1, ES2 and FS2) of the KH-11-07 cruise reported by Suzuki et al. (2013) should also be included in our Figure 8 plot. However, given that 1) the KOK cruise occurred in early June (two months after maximum direct discharge) and 2) the KH-11-07 cruise occurred in early August (four months after maximum direct discharge), then the doubling of the length of time since maximum direct discharge between the two cruises would have substantially increased the impact of ocean dynamics on the mixing and entrainment of the Fukushima release to depth. Hence, we feel it is clear that the KH-11-07 data do not provide information on the penetration depth of Fukushima radionuclides at the time of the KOK cruise. A similar argument exists with regards to the KT 11-27 ST08 and K8 data which were collected in late October. Indeed, we specifically point this out in the manuscript (page 19949, lines 8-16). In this context, we reiterate our statement that "...we infer that there are unresolved issues with the Hou et al. (2013) 129I data at all depths."

We also note that the coordinates we give for KT11-06 A (142.83° E 38.40° N) are decimal equivalents to the coordinates given in Suzuki et al. 2013 (142°50' E 38°24' N).

Comment 1: The commenter is incorrect in his assertion that "The exact same sets of data presented in this article have been reported by the same authors in JRNC (Tumey et al. 2013)..." An examination of the JRNC article and this paper should have informed the writer that the data presented in Tumey et al. (2013) consisted of ~50 analyses, with 16 of them being from the OOCL Tokyo. In this paper we present in excess of 200 data points (i.e., ~150 new results) of which 40 are from the OOCL Tokyo sample suite and the remainder from the R/V KOK cruise. Further, we explicitly noted in this paper (page 19939 lines 12-13) that a subset of the data presented in this paper have been previously presented in Tumey et al., 2013 (J Radioanal Nucl Chem

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296:957–962). As is evident from the discussion in this paper, the larger and more extensive data-set has allowed us to independently estimate the excess ^{129}I derived from Fukushima in the region sampled by the KOK and to explore the tracer-tracer relationships.

Comment 2: The commenter points out a useful clarification with regards to iodine sample preparation. Our procedure does call for addition of nitric acid following the sulfite and hydroxylamine additions to reduce the pH prior to addition of sodium nitrite. This addition enables the reduction of iodate to iodide before the nitrite addition which oxidizes iodide to molecular iodine. We have tested the effectiveness of this method using an in-house seawater QA/QC material which was prepared with iodide carrier, iodate carrier, and with no carrier. All three measurements yielded the same intrinsic $^{129}\text{I}/^{127}\text{I}$ ratio for the seawater thus demonstrating complete recovery of all inorganic iodide fractions. In the revised version we will clarify the sequence of nitric addition so that there is no future misunderstandings by other readers.

Comment 3: The commenter is correct: $\sim 1.2 \times 10^{-9}$ is $\sim 4.5 \times 10^{-5}$ Bq-m⁻³. We missed this typographical error during proof-reading. This will be corrected in the final version.

Comment 4: The commenter is correct that we have used an average value for the concentration of iodine in open ocean seawater (page 19939 lines 14-19); this average value was based on the cited work of Barkley and Thompson (1960), Elderfield and Truesdale (1980), Nakayama et al. (1989) and Zheng et al. (2012). The writer is incorrect in his assertion of a $\pm 10\%$ “typical” analytical uncertainty of the measurements presented in this paper; for most results presented in this paper the uncertainty is $\sim 3\%$ (see page 19939 lines 5-6). While it is clear that, for example, a sample that had an iodine concentration of $55 \mu\text{g l}^{-1}$ rather than $60 \mu\text{g l}^{-1}$ would have an $\sim 10\%$ higher $^{129}\text{I}/^{127}\text{I}$, this level of uncertainty in the iodine concentration does not change the major interpretations of the manuscript.

Comment 5: The commenter is correct in stating that the distribution of atmospheric

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weapons testing produced ^{129}I would have an initial heterogeneous input function in space and time. However, we do note that the residence time of the surface mixed layer (globally) is ~ 25 years. Given that the bulk of the ^{129}I input from atmospheric weapons testing likely occurred between 1961–1962 ($\sim 75\%$ of the global yield was in this window) the ocean has had a significant amount of time to mix and smooth out surface values in much of the temperate and even tropical ocean. Analogies exist with the present day surface distribution of other similarly produced transient tracers (e.g., ^3H , ^{14}C , ^{137}Cs). Empirically, the comparatively small range of surface ^{129}I values observed between $20\text{--}45^\circ\text{N}$ in the North Pacific implies that this is the case. We perhaps could have better articulated the various potential sources of ^{129}I into the Pacific. Obviously there are other (usually small) sources of anthropogenic ^{129}I in the Pacific that could have a local influence, e.g., Tokai Mura (c.f., Snyder et al., 2010). Although not the focus of the paper, we perhaps could have better articulated the various potential sources. A case in point is the potential influence of ^{129}I from the Columbia River (e.g., Kilius et al., 1994; Moran et al., 2002) for which we have data that is relevant and as discussed in this paper (section 4.5).

Comment 6: Please refer to page 19941 lines 5–8: In order to avoid potential representation error, the comparisons of ^{129}I and ^{137}Cs , ^{134}Cs were done on the exact same samples: i.e., same cast and niskin bottles. The comparison presented in the paper consists of 46 (^{134}Cs – ^{129}I) and 69 (^{137}Cs – ^{129}I) pairs. There are fewer ^{134}Cs pairs due to ^{134}Cs results being below detection limit.

Comment 7: We agree with the implication of the comment that the chemistry and reaction rates for iodine in gaseous and particulate form are not known very well. In detail the rate will be dependent on availability of condensation nuclei, droplet size, water vapor saturation state, air parcel history, etc. However, there does seem to be a consensus that the conversion of gaseous to particulate (including rain washout) is on the order of several weeks (e.g., Achim et al., 2012; Caput 1993; Kristianesen et al., 2013; Uemetsu et al., 1988). We neglected to include the appropriate references and

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will rectify this.

Comment 8: We thank the commenter for directing us to Thakur et al., paper which we had not seen. We note that much of the atmospheric data presented in Thakur et al., is common (ie., same primary source) as the references that we utilized: Biegeleski et al., (2012); Hoffman et al., (2012); Masson et al., (2012). We agree that observations of Fukushima fallout in Europe may not be the most appropriate in deriving a Pacific-centric ^{129}I deposition. In section 4.3 we laid out the assumptions that we used to estimate atmospheric $^{129}\text{I}/^{134}\text{Cs}$ (or $^{129}\text{I}/^{137}\text{Cs}$). To the best of our knowledge and at the time of submitting our paper, the Miyake et al., (2012) data on samples collected in Japan were the first direct ^{129}I data that would be relevant to estimating the potential of atmospheric deposition in the Pacific. Unfortunately, we were unable to locate ^{134}Cs or ^{137}Cs data from the exact same samples as presented in Miyake et al 2012.

Comment 9: We would disagree with the commenter that constructing ‘average’ or composite profiles for determining the excess of ^{129}I (or other Fukushima radionuclides such as ^{134}Cs , ^{137}Cs , e.g., Buesseler et al., 2012; or “bomb tracers such as ^{14}C ” e.g., Broecker et al., J. Geophys. Res., 90, 6953-6970, 1985 and a whole series of related papers) is meaningless. The writer is correct that there is the possibility of representation error and undersampling a non-homogenous tracer field. This is why such estimates have (comparatively) large errors. Be that as it may, a first order perturbation budget needs to start with the initial set of observations (e.g., those presented) and then move forward with advection-diffusion (model) based estimates in isopycnal space (e.g., Beherens et al., Env. Res. Letts, 7, 034004, 2012; Dietz et al., Ocean Sci., 8, 319–332, 2012; Rypina et al., 2013, among many).

Comment 10: We are confused by this comment. As documented in Tumej et al., (2013) and further demonstrated in the larger data-set presented in this manuscript, there are indeed numerous western subtropical Pacific surface water samples with an $^{129}\text{I}/^{127}\text{I}$ ratio less than $\sim 3\text{E}-11$. The derivation using ^{134}Cs vs ^{129}I yields a pre-event $^{129}\text{I}/^{127}\text{I}$ of $\sim 2.4\text{E}-11$. This is consistent with the empirical observations from

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the OOCL Tokyo surface data set which average $2.75(\pm 0.29, 1\text{-sigma sd})$ E-11 from Hong Kong to the dateline. If there is a significant semi-permanent latitudinal dependence in surface water ^{129}I with slightly lower $^{129}\text{I}/^{127}\text{I}$ water at lower latitudes than one might want, or need, to use the value from the south: as much of the water to the east of Fukushima is brought north and east via the Kuroshio. The larger question is whether or not there was, in surface waters, a small amount of Fukushima derived ^{129}I from atmospheric transport and deposition. For these reasons we used the internal consistency of the ^{134}Cs vs. ^{129}I relationship. In the manuscript we noted that the excess ^{129}I burden in the region sampled by the KOK will change by 10-25% depending on which pre-event ^{129}I is chosen (cf., page 19946 lines 15-18).

Comment 11. We thank the commenter. The figure call on page 19947, line 20 should indeed have been to Figure 3b.

Comment 12: The Columbia River is known to be a potential source of ‘anomalous’ or elevated ^{129}I due to past activities at the Hanford Site. In an ideal world we would have an additional and specific conservative tracer to elucidate the dilution of Columbia River water into the California Current System (CCS). Unfortunately over the timescale of interest, salinity in surface water is not conservative and thus we cannot use a direct dilution estimate from it. This is why we resorted to a simple estimate using ^{129}I . If one were to use salinity with the following end-members: 33.27 psu for the eastern subtropical North Pacific and average value of the CCS samples of 33.09 (i.e., the hydrographic data of the OOCL Tokyo sample set) then one would estimate something on the order of a half a percent dilution by freshwater. Over the years for which relevant data are available; 1998-2005, the activity of ^{129}I just downstream from Hanford had an annual average of $\sim 1.8 \times 10^{-6}$ (Bq-L⁻¹) in 2005 to $\sim 4.8 \times 10^{-6}$ (Bq-L⁻¹) in 1998 and an average across all of the years of $\sim 3.1 \times 10^{-6}$ (Bq-L⁻¹). From a two-endmember mixing model using the range of ^{129}I endmembers observed from downstream of Richland and the measured value one ends up with a range of dilution of, again, a few fractions of a percent: $\sim 0.2\text{-}0.5\%$. The total flow of the Columbia River from Richland (~ 3000

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m³-sec⁻¹) increases to ~ 7500 m³-sec⁻¹ at Astoria Oregon. Assuming no additional significant ¹²⁹I sources between Richland and the river's mouth requires an additional dilution factor of ~ 2.5 . The end result is as we state: that only tenths of a percent (nominally by volume) of Columbia River water are required to increase the 'background surface ¹²⁹I of the subtropical eastern north Pacific from $\sim 1.2 \times 10^{-5}$ Bq-m⁻³ to that in the CCS $\sim 2 \times 10^{-5}$ Bq-m⁻³.

Comment 13: We agree that it would have been ideal if the deeper samples presented by Suzuki et al., (2013) had smaller error bars. Be that as it may, the data reported are, as we and others have noted, consistent, given their uncertainties, with each other and the pre-anthropogenic ¹²⁹I ratio of Moran et al. 1998.

Moran $\sim 1.5 \times 10^{-12}$ Suzuki $1.2 \times 10^{-12} \pm 100\%$: gives a range of (0.0-2.4)E-12 Suzuki $3.5 \times 10^{-13} \pm 1000\%$: gives a range of (0.0-3.5)E-12

Indeed, one may infer that the data presented by Suzuki give an upper limit of the pre-anthropogenic ¹²⁹I of $2.4-3.5 \times 10^{-12}$, or less.

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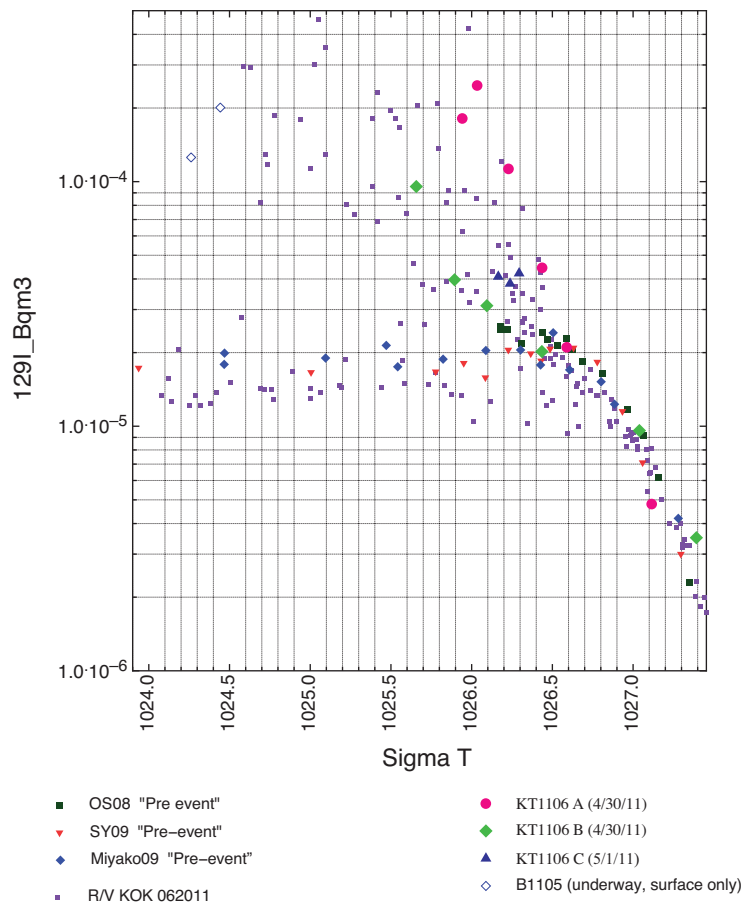


Fig. 1.

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