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

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The Guilderson et al. paper illustrates the importance of assembling a detailed inventory of iodine-129 in the world’s oceans, particularly in light of the Fukushima nuclear disaster. As mentioned in the introduction, iodine-129 can be used to provide retrospective dosimetry estimates of iodine-131 releases. The authors use two sampling transects to determine the variations in iodine-129 relative to stable iodine and radio-cesium, present the spatial distribution and speculate as to the amount of iodine-129 discharged into the oceans directly due to the accident (137 - 179 g). The authors note (section 4.3) that iodine-129 measurements are not made as frequently as I-131, but it

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is precisely the longer half-life of iodine-129 that has allowed many more samples to be made of iodine-129 than of iodine-131 following the nuclear accident (see, for example, Miyake et al, 2012, *Geochem. J* or retrospective dosimetry papers on Chernobyl). As a side note, it is worth mentioning that while the North Sea and Northeastern Atlantic Oceans have been adequately studied, only sporadic sampling for iodine-129 has been carried out for the rest of the world's oceans.

(1) While the author's presume a large pulse from the Columbia River, for example, due to the Hanford Project, the data we have is limited to: 1) seaweed (Kilius et al, 1994) who found that radioiodine was effectively scavenged out by macroalgae within 100 km of the Columbia River estuary; 2) River water and water from local streams (Moran et al., 2002), which does not seem to be much different from regional background values; and 3) shoreline springs, streams and monitoring wells surrounding the Hanford project (Patton et al., 2003, 2009). The Patton studies show that iodine-129 is localized and has not migrated as extensively as Tritium, for example, and the authors of the Hanford studies surmise that even in river water it is rapidly sorbed and scavenged out by sediments. I mention this because in order to make a qualitative assessment as to the contribution of iodine into the world's oceans, we need quantitative data in all of the world's oceans, preferably with repeated sampling over time. The complete absence of data along the Pacific Shores of the United States is not unique. While there is ample evidence for a large iodine-129 plume associated with the Savannah River project (Kaplan et al, *Environ. Sci. Technol.*, 2011), there has been no sampling for I-129 in seawater along the mid-Atlantic states either. In the Pacific Ocean, the are studies of Bikini and Einewetak do not include background transects, so it is hard to say whether the "bomb pulse" currently extends to any appreciable distance away from these two point sources. Guilderson's work is a start in the right direction, but I think there should be some reservation in providing a firm interpretation of the data until we have know more. (2) The authors tendency, for example, to be somewhat dismissive of the penetration depth of Hou et al. (2013) as a series of "unresolved issues", the regional elevation of I-129 in pre-Fukushima waters indicated by Povinec et al (2013) as

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"if real", and values from Monaco as "inadvertent contamination", simply highlights the level of uncertainty even with the interpretations of this paper until we have more data. At present, there is more data regarding $^{129}\text{I}/\text{I}$ ratios in rivers, lakes, and streams near coastal areas. While ^{129}I concentrations tend to be lower in streams than in shallow seawater, the ratios tend to be remarkably similar (Snyder et al., 2010). Because of this, it is essential that when iodine carrier is added to samples, that the stable iodine concentration of the sample is known, in order to actually calculate $^{129}\text{I}/\text{I}$ ratios in seawater, since the iodine concentration can change. (3) (Incidentally, Guilderson's paper is unclear about what the $^{129}\text{I}/\text{I}$ ratio of the carrier that was used is, if the value varied between sample runs and what errors were propagated as a result of this.). Although the iodine ratios from coastal surface waters are no substitute for actual seawater samples, perhaps they can be used until we actually have a more complete data set of marine values. (4) It might be mentioned that there is more data from Japan that could be incorporated into the interpretive section of this paper. Ohno et al. (Geochem J. 2012) provide $^{134}/^{137}$ ratios of 0.98 in shallow sediments around Fukushima, Toyama et al. (J. Env. Rad., 2012) provides a look at the variations in atmospheric deposition of ^{129}I prior to the accident. Incidentally, the Ohno paper shows $^{131}\text{I}/^{137}\text{Cs}$ values that are much lower than those assumed in section this paper, which highlights the uncertainty of these assumed ratios. (5) Finally, although the author's present estimated $^{129}\text{I}/\text{I}$ values to "afford simple comparison", they choose to plot this against potential density, rather than depth, which makes it difficult, if not impossible, to compare side-by-side with other studies that present values relative to seawater depth.

Response

We thank the reviewer, Dr. Snyder, for making the time to review the submitted manuscript. We agree with many of the thoughts incorporated in his commentary. We reference our replies to the numbers that we have inserted into his above commentary

(1) While the author's presume a large pulse from the Columbia River, for example, due to the Hanford Project, the data we have is limited. . .

Reply:

We concur that the available data is limited. In an ideal world there would be routine analyses on river waters to better understand the temporal variability of ^{129}I export to the ocean.

In regard to the elevated ^{129}I values in the California Current System: we approached the VOS surface samples from the standpoint that the CCS values were clearly above the central North Pacific surface samples and that one possible explanation was atmospheric deposition of Fukushima-derived ^{129}I . Unfortunately, the collected VOS samples were small volume, which precluded the characterization of the samples for a suite of radiochemical tracers. But, given the NYK-samples-based indication that atmospheric deposition was not the source of the elevated levels, we explored the possibility (i.e., hypothesized) that the source of the elevated ^{129}I signature was re-circulation within the CCS of freshwater derived from a river with a known history of elevated ^{129}I .

We concur that, based on the data presented in Moran et al., (2002), the Columbia River is not the only river in North America that exhibits elevated ^{129}I , Moran et al. (1998; IAEA SM-354/101; and tabulated in their 2002 paper) document that the Hood River (which flows into the Columbia River from the south ~ 170 west of Hanford) has higher ^{129}I than other rivers with similar iodine concentrations. They hypothesized that, although not influenced by direct liquid-discharge from Hanford, atmospheric releases from the Hanford complex have perturbed the drainage basin/watershed providing a source of ^{129}I . We do note that although Patton et al. (2003, 2009) surmise that current and recent releases of ^{129}I have been ameliorated by adsorption and sequestration in sediment, this does not necessarily negate our hypothesis that the signal that we observe is due to elevated Columbia River ^{129}I . One should remember that the measurements presented in the PNNL reports have a detection limit of $\sim 10^{-5}$ which is multiple orders of magnitude above the data presented in our paper and the sensitivity of the analysis. At best, the PNNL data place (an AMS relevant) upper limit on the ^{129}I

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available for downstream export.

More importantly the Columbia River dominates the CCS freshwater budget. The Moran et al. data indicate ~7-8 grams of 129I export at the mouth of the Columbia River in 1996. Given the history at Hanford, we can assume that the elevated 129I directly measured by Moran et al., occurred for multiple years on either side of their observation. Although the overall flow of the CCS approaches 2 Sv with an up-to-~20 year mean transit time, the age probability distribution function of this advection and diffusion likely has long tails. The recirculation within the CCS, and the long history of release from Hanford/Columbia River convolved with the residence time of waters in the CCS implies that 129I could accumulate in the CCS.

Notwithstanding the above and the current wording of our discussion of the elevated CCS levels in the paper, we will edit section 4.5 endeavoring to make it very clear that we are hypothesizing that the source of the 129I is the Columbia River, and do not mean to imply that our results are sufficient to conclusively identify the cause of the elevated 129I levels in the CCS. We will include a similar modification in the conclusion section.

(2) The authors tendency, for example, to be somewhat dismissive of the penetration depth of Hou et al. (2013) as a series of "unresolved issues", the regional elevation of I-129 in pre-Fukushima waters indicated by Povinec et al (2013) as "if real", and values from Monaco as "inadvertent contamination", simply highlights the level of uncertainty even with the interpretations of this paper until we have more data.

Reply

With regards to the data of Hou et al. (2013) and Povinec et al. (2010; 2013), we were not being pejoratively dismissive of their work. As the reviewer points out, there is a paucity of data at all scales (temporal and spatial) in the Pacific against which one can compare new data sets. This is one of the reasons why we put our data in the context of those presented by Suzuki et al. (2013). In particular, we were concerned about low

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129I abundance data given recent statements in the literature that “no (recent) seawater samples have been observed with close to pre-anthropogenic levels” (Hou et al., 2013). This is in contrast to both the data sets presented by Suzuki et al. (2013, and also 2010) and our data sets, which indicate low-level ($\leq 1 \times 10^{-11}$) 129I at potential densities greater than 27.3 kg/m³ in the far western subtropical Pacific. We do, however, stand by our original statement that the 129I data presented by Hou et al. (2013), in the context of the currently available cesium data and our own 129I data, stand out as having elevated 129I values at density surfaces which do not appear to have been influenced by Fukushima-derived radionuclides at the time of the R/V KOK sampling. Visually comparing our figure 7 (cesium data of Buesseler et al., 2012) and figure 8 supports such a statement. The Povinec et al. (2013) data imply near surface 129I values throughout the whole of the water column that they sampled; for the Bikini and Enewetak stations (6 and 7, respectively) this is to 4400 and 2500 meters water depth, respectively, and for the ‘background’ stations to the northwest (stations 2 and 3) to 5000 meters. The implied excess 129I (several 10s of kg), although not impossible in the scope of uncertainty of weapons testing produced 129I, is difficult to reconcile.

The results presented by Hou et al., were from samples initially collected for the group at Monaco. Given the information in their respective papers the extraction and preparation of targets were done in different locations, and analyzed at several different AMS laboratories. We hypothesized that the contamination of the two independent sample sets occurred at Monaco or due to issues at Monaco (eg., contaminated sample containers) because that is the common feature to the sample sets. We note that although process blanks were prepared and reported by Hou et al., that the inference is that these were not field blanks using the same collection vessels (cubitainers or HDPE bottles).

(3) Incidentally, Guilderson’s paper is unclear about what the 129I/I ratio of the carrier that was used is, if the value varied between sample runs and what errors were propagated as a result of this.

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Working carrier solutions were generated by dissolving measured masses of Woodward iodine. Over the course of preparing and measuring the OOCL Tokyo and R/V KOK sample suite we used several separately prepared carrier solutions. As indicated in our paper (page 19939, line 6), process blanks, prepared using these carrier solutions, were measured along with the various batches of samples. The measured $^{129}\text{I}/^{127}\text{I}$ ratios obtained for the process blanks averaged 3.8×10^{-14} . The measured $^{129}\text{I}/^{127}\text{I}$ ratios for unknown samples were orders of magnitude higher than this very low ^{129}I level in the process blanks and results obtained for unknown samples were not corrected for the ^{129}I introduced through the addition of the carrier. For each batch of unknown samples, the measurement results were reduced using the appropriate batch or date-specific carrier solution, and uncertainties (including the measurement uncertainty in the carrier ^{127}I mass and the uncertainty in the assumed seawater iodine concentration) were propagated appropriately in a fashion consistent with the algorithms presented in Bevington & Robinson. Additionally, we note that for the majority of the replicate analyses presented in our figure 2, individual samples in each replicate pair were actually prepared using separately prepared carrier solutions.

(4) Ohno et al. (Geochem J. 2012) provide $^{134}\text{I}/^{137}\text{I}$ ratios of 0.98 in shallow sediments around Fukushima, Toyama et al. (J. Env. Rad., 2012) provides a look at the variations in atmospheric deposition of ^{129}I prior to the accident. Incidentally, the Ohno paper shows $^{131}\text{I}/^{137}\text{Cs}$ values that are much lower than those assumed in section this paper, which highlights the uncertainty of these assumed ratios

Reply

We thank you for the Ohno reference. We did not know of it. The $^{131}\text{I}/^{134}\text{Cs}$ (or $^{131}\text{I}/^{137}\text{Cs}$) ratio is less than that chosen/estimated by us in estimating the potential impact of atmospheric transport and deposition of ^{129}I in the north Pacific. Thus, our estimated impact may be an upper limit. We will revise section 4.3 to reinforce the uncertainty in the assessment and remark that other data (e.g., Ohno et al.,) would imply slightly lower impact in e.g., the NYK VOS sample set.

The Toyama et al., data set is a wonderful addition to the broader radiochemistry community. The data give credence to our interpretation of the composite western subtropical ^{129}I profile in figure 10 being mainly driven by a singular forcing function (atmospheric weapons testing derived ^{129}I).

(5) Finally, although the author's present estimated $^{129}\text{I}/\text{I}$ values to "afford simple comparison", they choose to plot this against potential density, rather than depth, which makes it difficult, if not impossible, to compare side-by-side with other studies that present values relative to seawater depth.

Reply

We note that in the chemical oceanographic community that it is common to present data as a function of density. Doing so, as opposed to as a function of depth, more accurately presents oceanographic properties and the influence of isopycnal mixing and diffusion. Water of the same density is not always at the same depth and thus one can be influenced by apparent fits/misfits that are a consequence of the reference frame. The subtropical gyre at some level has the appearance of a deep bowl. Thus lines of constant density slope down from the far western Pacific into the central Pacific and the distance between isopycnals at one geographic location will change, i.e., the thickness of a particular water mass can vary (see figure below for the 26.8 potential density surface). Off Japan, this results in a compression of water masses (density surfaces) as a function of depth relative to regions to the south and east.

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Depth [m] @ Potential Density Anomaly $\text{kg}\cdot\text{m}^{-3} = 26.8$

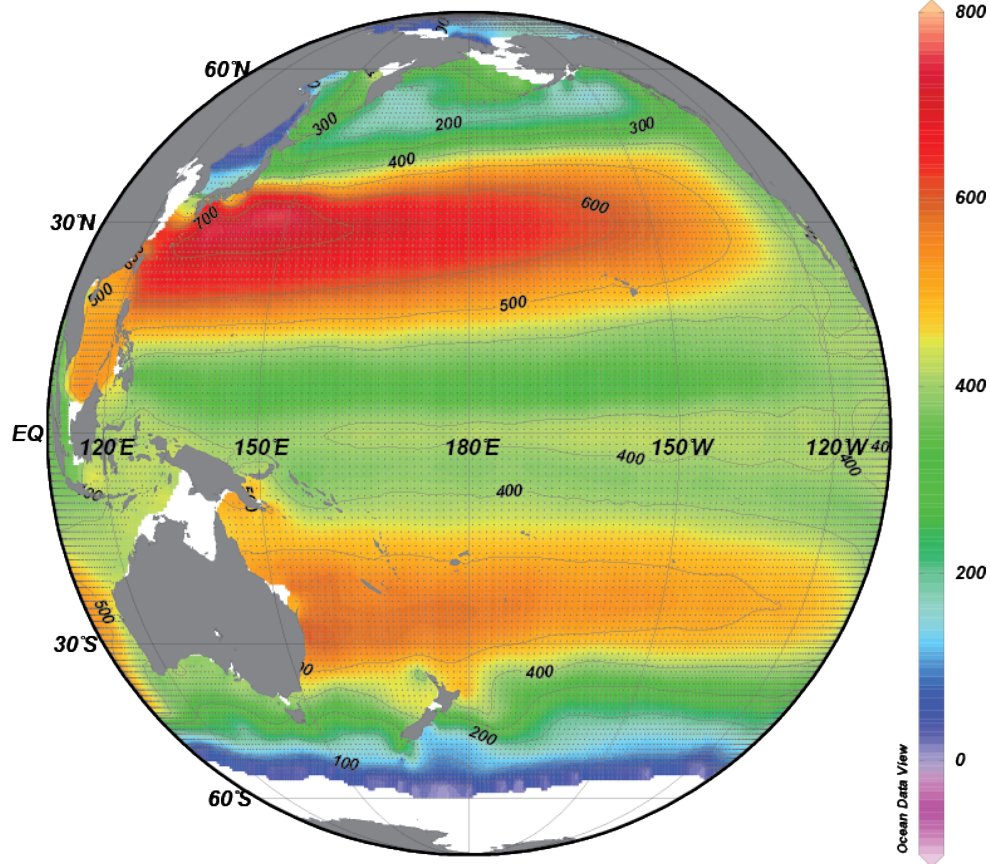


Fig. 1.