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Discussions
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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.

T. P. Guilderson et al.

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Reply to reviewer one comment's. Numbers are referenced as per reviewer.

Comment 1 As mentioned by the authors in Figure 1, the transect used for the sampling lies in a critical oceanographic position that separates the cold Arctic-related front from the warm subtropical-related front. Along this part of the Pacific, the Oyashio Current brings cold Arctic water that flows south and circulates counterclockwise in the western north part. The effect of this water parcel and possible inclusion of 129I from the Arctic Ocean has not been considered. Although this current may bring small amount of 129I, but these can be significant when compared to the also small amounts discharged

from the Fukushima. Furthermore, it is not clear why the depth penetration of the 129I signal is deeper in near shore compared to off shore and what mechanisms control this feature.

Reply:

The reviewer is correct that we did not explicitly attempt to tease out any potential influence of mixing of subpolar water masses on the 129I content off Fukushima. In part this is because we believe, and as other published data imply, the influence of Fukushima dominates the 129I story for samples collected on KOK 1108b. The trans-Pacific VOS sample set only has temperature and salinity data and thus it would be difficult to quantitatively assess via any tracer-derived mixing model the relative influence of subpolar versus subtropical water. We do note that the VOS Tokyo samples that came from surface waters between 8-10°C (stations 21-34) likely sampled predominantly subpolar surface water. The 129I/127I for these samples averaged ∼3x10-11. The coldest (water) sample of 6.7°C had a 129I/127I of 2.9x10-11 which is similar to the average. The non-Fukushima influenced 129I/127I as presented in our paper have ∼10% variability. This may be the level of variability inherent in surface waters, or (as noted by this reviewer, see comment 2) could be due to assumptions on the concentration of stable iodine.

From these data we could infer that there is little "visible" influence of purported Arctic 129I in our sample set. We also note that the mean flow across the Bering Strait is from the Pacific into the Arctic. From 11 years of current meter data that span the Bering Strait the mean flow of 0.7-1.1 Sv is into the Arctic with only sporadic (weeklong duration) flow to the south (Woodgate et al., 2012). Far western Canada Basin surface water 129I in does not appear to have changed between 1995 and 2008 (discrete data from 1995, 2001, 2006, and 2008), and is consistent with mostly only atmospheric weapons deposition of 129I (Smith et al., 2011). This is in contrast to other portions of the Arctic, which are influenced by the reprocessing signature of Sellafield and La Hague. To the best of our knowledge, these are the most recent relevant data, but

admittedly do not include surface waters of the Chuckchi Sea or Bering Strait proper.

With regards to the comment asking why the penetration of Fukushima derived radionuclides is found at deeper depths near shore as compared to off-shore, this is due to the increase turbulence and shear associated with shallow topographic features (e.g., shelf/slope) to the east of Japan. We will add a specific statement to this effect in the revised version of the paper.

Comment 2 The authors uses a common value of 127I to calculate the 129I/127I ratio which adds a weak component to the interpretation as it is quite known that 127I concentration can vary a lot in ocean water with respect to depth and distance from shores.

Reply:

The commenter is correct in a general sense that dissolved inorganic iodine can vary significantly across the total possible types of water masses encountered in oceanic environments including estuaries. This is because river waters tend to have low iodine concentrations. In general, the potential range of iodine concentrations is actually much reduced for open ocean and coastal (non-estuarine) environments and is ∼10%. To the level that iodine is mostly conservative and would behave as a passive tracer (e.g., more like salinity), the salinities encountered in the sample suite from the KOK average 34.16 with a total range of 33.51 to 34.78, which would imply a limited range of iodine concentrations. Indeed, this is reflected in the iodine concentration data of Hou et al. (2013) for samples from the R/V KOK where the average concentration is 58 ± 2.6 (1-sigma sd) μg-L-1 (range of 54-62μg-L-1).

Comment 3 It is difficult to understand the representation of the about 1 kg of 129I direct discharge. Is that a direct discharge just into the sea or even to the atmosphere? How much is the accuracy of the estimate?

Reply:

The direct discharge is that discharged directly into the ocean. This terminology follows previous papers discussing Fukushima and which distinguish 'direct' oceanic discharge from 'atmospheric' release. In the revised version of the manuscript we will clarify the useage of 'direct' discharge.

Our estimate is based on the excess inventory of 129I determined in the region sampled by the R/V KOK. Based on the available data, including that presented in Buesseler et al. (2012), and inferences from modeling, the majority of the 'excess' 129I is derived from direct discharge with only a minor contribution from atmospheric deposition.

There are several sources of uncertainty in the estimate of direct discharge. The first is the uncertainty in the 'excess' 129I in the oceanic region sampled by the R/V KOK. In effect this is the representation error of sampling a non-homogeneously mixed tracer and is often ∼30% or more (e.g., page 19946, lines 24-25). This 'excess' inventory then needs to be assessed within the context that the observations were ∼3 months after the initial release. This requires the use of an ocean advection-diffusion transport model to determine how much of the release had been advected out of the region that was sampled. In our manuscript we build off the work of Buesseler et al. (2012) and Rypina et al. (2013) to scale the excess 129I. Therefore direct discharge inferences are going to be model dependent (page 19946/47 lines 28-29, 1): i.e., a different ocean model, even forced with the same winds, could result in different estimates of retention and dilution. Without additional independent, or even an ensemble of model estimates from the same transport model using different parameters (e.g., horizontal and vertical diffusivity), it would be difficult to put a fixed number to the error. It is very likely at least ±50% but it could be larger.

It may be illustrative to put the estimated 129I releases from Fukushima in context with the more studied, Chernobyl event which released ∼6 kg of 129I (Snyder et al., 2010). The estimated Fukushima atmospheric 137Cs release is 10-16 PBq (e.g., Biegeleski et al., 2012) which when convolved with the estimated 129I/137Cs activity ratio of at-

mospheric release (page 19944 lines 19-29, page 19945 lines 1-3) implies ~3-5 kg of 129I. Combined with our estimate of 1 kg of 129I via direct (oceanic) discharge yields a total release of ~4-6 kg of 129I. We also note that precipitation during onshore flow, i.e., an east wind, over March 14-16 and March 21-22 resulted in significant (2.1-2.9 PBq) 137Cs fallout to the west and north of the NPP. Thus up to 1 (of the ~3-5) kg estimated 129I released into the atmosphere may have ended up falling out back on Japan. The above estimate is dependent on the assumptions as discussed in the manuscript attempting to relate the washout of 134Cs and 137Cs to 129I without any (to our knowledge) 129I/134Cs or 129I/137Cs data. In our estimate we went through a correlation exercise using the 129I/131I data of Miyake et al. (2012). Such a back of the envelope calculation should be verified with direct observations.

Comment 4. The authors further use a term called excess inventory of 129I calculated from 134Cs data. As the authors also mention in the text, the chemistry of these two isotopes is rather different in aquatic environment in terms of ionization forms and uptake (iodine is taken by the biomass while Cs is rejected). Therefore the estimate of the excess inventory should be considered with large errors.

Reply:

The range that we give for the excess 129I inventory in the region sampled by the R/V KOK is 136-179 grams (page 19946, lines 16-22). The reported range is due to the possible choices of the pre-event (pre-Fukushima) 129I/127I background value: the 129I zero 134Cs intercept versus the western Pacific values observed in the OOCL Tokyo sample set (~2.8x10-11), or the 'by-eye' line of ~3.5x10-11 of surface values observed in the R/V KOK suite. The uncertainty for the excess 129I inventory for any of these calculated values then needs to take into account the potential representation error, which from similar studies is ±30%.

An admittedly crude estimate of the iodine flux lost via sinking phytoplankton can be derived taking advantage of the long-term sediment trap programs and shorter term

floating traps at the KNOT K2 and S1 sites (e.g., Honda et al., 2006; Honda et al, 2013; Kobari et al., 2013). If we assume a carbon export flux of 20-100 mg-m-2-day-1 and an I/C ratio of ~ 1x10-4 this yields an iodine flux up to ~0.01 mg iodine m-2-day-1. We can then scale this to the ~150,000 km2 region sampled by the RV KOK and yield an upper bound of an instantaneous export of ~1500 kg iodine-day-1. For the instantaneous comparison with the excess inventory of dissolved 129I we can use ~5x10-10 which is in the mid-range of the elevated values that we observe which yields ~0.0008 grams of 129I (versus 136-179g in the dissolved excess inventory). Therefore, relative to the instantaneous dissolved 129I excess inventory for the region sampled by the R/V KOK that exported out of the surface waters is estimated to be very small.

There are obvious assumptions with taking this estimated daily flux and expanding to the ~100 days post event and what the 129I/127I ratio of the integrated (time/space) exported C might be. Be that as it may, if we hypothesize that the integrated average 129I/127I was ~5x10-10, this yields a very uncertain 129I pelagic total flux of ~ 0.08 grams 129I. If the time-area integrated 129I/127I ratio was 5x10-9 this ends up being ~0.8 grams. This number is still less than a percent of the instantaneous excess dissolved inorganic 129I inventory and does not substantially alter the budget estimates presented in the manuscript. It is more difficult to make an assessment of what the local coastal (seaweed) budget of 129I might be even taking into account the higher iodine concentration in seaweeds (range of 10s to ~3000 μg I/gram seaweed).

We will include statements in the revised manuscript specifically stating that our estimates neglect any potential biological uptake and subsequent trophic level transport or export to the deep ocean via sinking POC.

Comment 5 Iodine as element is taken by and adsorbed to planktons, algae, seaweeds and even larger organism. This feature adds further uncertainty to the estimates of 129I given in the paper. Unfortunately, consideration of this aspect has been neglected in the paper which is vital for the understanding of iodine budget in the ocean.

Reply:

The reviewer is correct that we have focused our effort on the dissolved inorganic 129I budget. We have no direct-129I data on biological materials to assess that which would be in higher trophic levels, POC, or in coastal macrophytes. Without these basic data any extrapolation to what might be the biological 129I reservoir would be highly uncertain

Comment 6 The relatively high values observed along the Californian current seem strange as also mentioned by the authors. The source as from the Columbia River needs further input.

Reply:

If we had 134Cs/137Cs data for the same samples (which we do not) then we could quantitatively assess if the increased 129I values were due to atmospheric deposition of atmospheric releases from Fukushima. Because the CCS data are so consistent, in lieu of the variability we might expect from atmospheric deposition, we infer/hypothesize that the increased levels of 129I that we observe in the California Current System are due to 129I from the Columbia River.

There are related data to support such a hypothesis.

Data include a study of Kilius et al. (1994) who, using seaweed collected between 1988-1993, document increased levels of 129I in the Columbia River estuary (see figure below taken from their paper). These secondary data are consistent with elevated 129I measured in the Columbia River by Moran et al. (2002) who for 1996 estimated from direct observations an annual discharge, at the time, of ∼8 g of 129I. The 25-65 129I grams-year-1 near Hanford (pg 19948 lines 1-16) is an upper limit of the 129I that would be available to exit the Columbia River. The observations in Richland do not include any biological uptake/fixation during transit or other loss terms.

From an oceanographic standpoint it is very possible to accumulate 129I from the

Columbia River in the CCS. A back of the envelope estimate using the area of the CCS (∼2x107 km2), the depth of the CCS (250 to 500 m) and estimates (observed and via regional ocean modeling) of the flow (∼2 Sv) implies that waters in the CCS have a residence time up to 20 years.

Additionally, 129I in CCS waters sampled at Scripps Inst. of Oceanography (La Jolla, CA) are, and have remained, elevated relative to more open Pacific surface values (Chang et al., 2012, poster presentation at Fall AGU 2012 meeting). We believe that it would be difficult for continuously elevated 129I values at La Jolla to be the result of Fukushima atmospheric deposition in the CCS.

As we specifically remark (pg 19948 lines 12-16), further isolation of the source of 129I will require tracer-tracer (i.e., a variety of radiochemical isotope) analyses.

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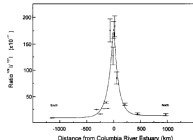Interactive comment on Biogeosciences Discuss., 10, 19935, 2013.

Fig. 2. The $^{129}I/I$ ratios ($\times 10^{-11}$) plotted as a function of the distance from the mouth of the Columbia River. The solid curves are derived from a least square fit to the southern and northern portions of the measured iodine isotope distribution.

Fig. 1.

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