Replies to Comment on bg-2021-10 by anonymous referee #2

Dear anonymous referee #2,

We submit our replies to your comments as below.
Thank you very much for your comments which greatly contribute to the improvement of our manuscript.

Best regards,

Michio
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Reply to Referee#2:

This was a difficult paper to read/review due to the lack of flow and logical sequencing in the paper. Moreover, the grammar and writing style made it difficult to interpret the precise intent of the authors. The paper does not appear to have been proof-read carefully – there are duplicate redundant sentences (e.g., lines 130-132, 138-139) and figure labels do not always match what is in the figure caption (e.g., Figure 4).

Reply: In some parts of the main text, there are obvious redundant sentences as RC2 pointed out. The Y-axis label in figure 4 should be 3H, not 137Cs. We will recheck all the text, tables and figures again before submitting a revised version.

The data, 3H and 137Cs data collected over 2014-2018 from the coastal waters of eastern Japan near the Fukushima Nuclear Power Plant and two sites further south, seem to be looking for a home. Although Biogeosciences hosted a special issue dedicated to the Fukushima event, it is unclear to me if Biogeosciences is the 'best' journal for these newer monitoring style data compared to a journal more specific to radionuclides and radiochemistry (e.g., Journal of Environmental Radioactivity, Journal of Radioanalytical and Nuclear Chemistry).
Reply: Since this article is not only dedicated to radionuclide activity, but also to their dynamics including the issues of freshwater fluxes, Biogeosciences appears very appropriate.

The authors expend a good deal of writing for background on atmospheric weapons testing derived tritium (^{3}H). I do not believe this was a good use of space in the manuscript. Of the ^{3}H produced by weapons testing, and if we use 1963 as the initial time zero, less than ~6% of weapons testing ^{3}H is still in the environment (atmosphere, terrestrial, ocean reservoirs). The authors could significantly shorten and tighten the introduction to simply state the background ^{3}H (and ^{137}Cs) in the western subtropical Pacific, that controlled releases from FNPP elevated coastal water ^{3}H prior to the earthquake/tsunami induced cataclysm, and then go straight into line 120: "In this paper, we present…"
Reply: As you pointed out as well as referee#1, we will shorten the introduction.

The authors could also state the purpose or what they were looking to explore/understand. Were they looking to better understand/constrain the relative influences of FNPP impacted

submarine groundwater discharge versus surface (river) input of ^{3}H and ^{137}Cs on coastal water concentrations?

Reply: We revised the introduction and stated the objectives of this article more clearly at the end of the introduction as below.

In this paper, we present results of the ^{3}H activity concentration observed during the SoSo 5 rivers cruise and at the Tomioka port and Hasaki, a pier of the Hasaki Oceanographical Research Station of the Port and Airport Research Institute, and discuss the behaviour of ^{3}H in the coastal region of Fukushima. An assessment of various contributions to both ^{3}H and 137Cs fluxes into the coastal area close to the FNPP1 site was carried out taking into account i) riverine flux of these radionuclides based on precipitation amount on the catchments of several rivers, ii) fluxes estimation using from the FNPP1 site by previously reported method (Kanda 2012, Tsumune et al., 2012, 2013, 2020) and iii) fluxes from open water towards based on the speed of the coastal current and the activity concentrations characterizing these open water. All of these results are discussed together with 3H activity concentrations in river and open sea waters already published.

Does the different physical chemistry of cesium and tritium lead to different input functions in the coastal waters (eg., cesium will desorb off particles when it gets to higher salinity)? This is particularly relevant with regards to submarine groundwater discharge which is a significant source of 137Cs (e.g., Sanial et al., 2017 www.pnas.org/cgi/doi/10.1073/pnas.1708659114) post direct discharge (eg., Buesseler et al, 2012).

Reply: RC2's comment that "submarine groundwater discharge which is a significant source of 137Cs (e.g., Sanial et al., 2017 www.pnas.org/cgi/doi/10.1073/pnas.1708659114) post direct discharge (eg., Buesseler et al, 2012)". We do not find such impact when we analyzed the data around the coastal region of Fukushima. Although Sanial et al., 2017 presented a very interesting phenomenon, it was put into evidence at a specific area south of FNPP1 site where waters were highly contaminated by initial direct release. Their estimates might be overestimated because most of the area is rocky. We observed that 137Cs and 3H activity concentration along the Fukushima coast showed a maximum at FNPP1 site with decreasing activities both north and south of FNPP1 site. If the flux from the contaminated beach was significant at the time of our sampling, the distribution of 137Cs activity concentration along the coast should show a different shape.

Key takeaways:

From the TEPCO 56N canal data, it is pretty clear that FNPP is (still) a source of 3H, regardless of the sensitivity of their methods being limited to > 1650Bq-m-3.

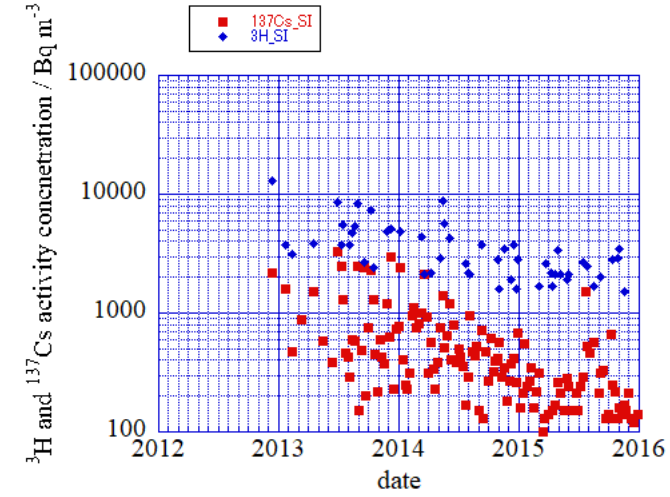
Reply: Yes, it is.

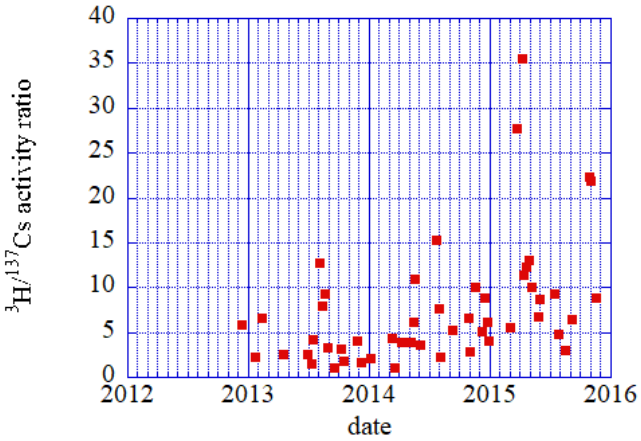
The Aoyoma et al., additional data capture the input of 3H and 137Cs into coastal waters.

Reply: Yes, this point is one of key issues of this article. Thanks,.

One of the most intriguing aspects of the data is the 3H/137Cs ratio that has varied post direct discharge in 2011 to the newer data. The authors do not provide a credible discussion/interpretation of this observation.

Reply: Ok, both 3H/137Cs activity ratio and their respective activities during the period 2012 -2016 are showed in figures below. 3H/137Cs activity ratio increase over time as mentioned in our text and not decline, while activity concentrations of 3H and 137Cs at 56N, shows a different decreasing trend.





In the current text, we stated “During the period from 2013 to 2016 at 56N of FNPP1, the 137Cs activity concentration decreased around two orders of magnitude lower due to decontamination effort of TEPCO while the 3H activity concentration decreased gradually, then the 3H/137Cs activity ratio tended to increase from 1 to 10 during the period from 2013 to 2016 (Figure are not shown). “ Only we need to do here is to revise from “two orders” to “one order”. We keep the other part of the sentence because it is the correct description of the trend of the two radionuclides. Since the observation at 56N was carried out very close to the source, we can assume that the observed trend was little affected by the water movement but mostly represents the change in the characteristics of the source. If the observation was done after some movement, salinity may affect the ratio as RC1 pointed, but this is not the case here.

So, in the revised main txet, this part will be as below;
Between 2013 and 2016 at 56N of FNPP1, the 137Cs activity concentration decreased by about one order of magnitude due to decontamination effort of TEPCO while the 3H activity concentration decreased only gradually, leading to an increase of the 3H/137Cs activity ratio from 1 to 10 (Figure are not shown).

What are the uncertainties on the flux (input) estimates? Are there any ‘real’ differences in the estimates provided in eg., table 3?
Reply: The uncertainties on the flux budget depend on the uncertainties of each parameter entering into r the calculation.
For the activity of 3H and 137Cs in open water, uncertainties are around 10-20%. For the river waters at Ukedo and Tomioka, uncertainties are around 10 %. For the activity of 3H and 137Cs at 56N of FNPP1 and in the port of FNPP1, variability is large and uncertainties are around 50 -100 %. Therefore, the order/rank of estimated values is real. Therefore, in the main

text, we thought it safe to say that “Regarding the 3H fluxes, the largest source comes from the open-water inflow from the north of FNPP1, with 52 GBq day-1 while the rivers north of FNPP1 contribute 3–6 GBq day-1. From the port of FNPP1, we used Kanda's method (Kanda 2012), ,,,, that led to 3H fluxes in the range of 1.9–4.5 GBq day-1 in three cases in 2014 and 2019, which is comparable with the 3H fluxes from the rivers located north of FNPP1.” In other words, our estimate is at least correct as an estimate of the relative importance of the different contributions.

End of reply to RC2.