

Biogeosciences Discuss., referee comment RC1  
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## Comment on bg-2021-10

Anonymous Referee #1

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Referee comment on "Tritium activity concentration and behaviour in coastal regions of Fukushima in 2014" by Michio Aoyama et al., Biogeosciences Discuss.,  
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This study estimated the tritium flux from rivers, Japan, and Fukushima Nuclear Power Plant after its incident caused by the tsunami in 2014 and during typhoon Hagibis and following typhoons in 2019.

However, the manuscript is partially difficult to read because a lot of information is missing and there are lots of typo. The contents of the text should be reconsidered. There are several topics which should be sufficiently discussed such as tritium in marine biota, difference in tritium between 2013-2014 and 2019, and the validation of the flux calculation and so on. All of the figures are very difficult to understand, particularly Figure 1 indicating sampling stations. I recommend that this figure is required to be revised.

L17 "Abstract."

I could not find out the information of the location of stations and areas the authors presented here. Because the authors described the concentration ranges at the specific stations (i.e. 56 north and soso 5 rivers) without any explanation, readers cannot distinctly understand what the authors intend to say. Define any abbreviations used in the abstract. This abstract seems to be kind of results and discussion section. As suggested in submission section of this journal (<https://www.biogeosciences.net/submission.html>), the abstract should be short, clear, and concise even though there is no limitation. This

abstract must be short.

L17-18 "SoSo 5 rivers cruise in 2014"

What rivers did the authors indicate. Readers never know where those rivers are located.

L18 "Tomioka port"

Where is this port?

L19 "Fukushima coast"

What coast is indicated? The Fukushima coast is composed by meridionally oriented coastline that is over 100 km long.

L21 "FNPP1"

Please specify this abbreviation.

L21 "around 200-500 Bq m<sup>-3</sup>"

This phrase seems to be redundant. " are in the range of 200-500"?

L21-22 "slightly lower than"

Is that statistically appropriate?

L24 "56N of the FNPP1"

This station is same as 56 north canal? Please specify.

I think that indicating concentration range is important because tritium flux from this area could affect tritium level in the surrounding "seawater", and is prevailing the riverine discharge.

L25 "both north and south of the FNPP1 site"

What areas did the authors indicate?

L25-26 "in river waters."

What rivers did the authors indicate? SoSo 5rivers?

L26-27 "at the stations located both north and south of the FNPP1 site"

Are those stations are different from above surrounding waters both north and south of the FNPP1? Please define the difference.

L34-35 "The open-water  $^3\text{H}$  activity concentration 35 contribution to coastal waters"

Does the phrase "open water activity concentration" refer to the activity concentration of tritium at cesium-137 = 0 on the x-axis? It seems difficult to understand from the abstract alone, while the authors indicate intercepts by standardized major axis regressions.

L37-39 "The  $^3\text{H}$  and  $^{137}\text{Cs}$  fluxes to the coastal region of Fukushima based on the open-water movement, freshwater flux from the rivers based on their respective catchment, and mean monthly precipitation were estimated."

From this concept, it is difficult to follow how to calculate the fluxes. In addition, there are no data on Cs-137 fluxes in this abstract.

L44-45 "which is one order of magnitude larger than those estimated using  $^3\text{H}$  activity concentration in the FNPP1 port."

Is this tritium flux at the FNPP1 port same as, "1.9-4.5 GBq day<sup>-1</sup>  $^3\text{H}$  flux at  $^3\text{H}$  using the

H activity concentration at the port"? Or is it Tomioka Port? Readers can be confused because one station has several different station names. The abstract also seems to have two different ports, so when the authors indicate a port, as " the port", the reader may not understand which port is referred to.

L47-51 "The  $^3\text{H}$  activity concentration of TFWT in the fish filets collected close to the FNPP1 site ranged from  $97 \pm 11 \text{ Bq m}^{-3}$  to  $144 \pm 11 \text{ Bq m}^{-3}$ , which were similar to the  $^3\text{H}$  activity concentrations in the surrounding seawater, in agreement with the knowledge that the bioconcentration factor of  $^3\text{H}$  is approximately 1. In contrast, higher values were found in TOBT, which can be linked to life- history traits."

Discussion on the activity in fish has not conducted. I recommend that the authors delete it.

L52 "1 Introduction"

The explanation concerning tritium in the environment is too long and is going out of focus on the local area of this study.

L103-107 "Momoshima et al. (Momoshima et al., 1987) measured the  $^3\text{H}$  activity concentration in various environmental materials around a typical nuclear power station, the Genkai Nuclear power plant, in Japan in 1983 and 1984. They found that the elevated  $^3\text{H}$  activity concentration was observed on one occasion in pine needles and surface soil after noting a high  $^3\text{H}$  activity concentration of  $220\text{--}10000 \text{ Bq m}^{-3}$  in the seawater."

Again, this study focuses on the fluxes from rivers and port of the FNPP1, but the relationship between tritium level in seawater and that in marine organisms is not sufficiently discussed.

L107-111 "However, no incidental increase in  $^3\text{H}$  levels was observed in atmospheric water vapour, hydrogen, and methane. Masson et al. (Masson et al., 2005) presented free-water  $^3\text{H}$  and organically bound  $^3\text{H}$  levels in the French coastal marine environment, from Concarneau to Gravelines, along with the  $^3\text{H}$  levels in the seawater. The matrices selected for their specific survey included seawater, seaweed, molluscs, crustaceans, and"

The same comment as above mentioned.

L120-121 "In this paper, we present results of the  $^3\text{H}$  activity concentration observed during the SoSo 5 rivers cruise and at the Tomioka port and Hasaki and discuss"

From the explanation given above by the authors, I do not think that the readers can relate the objectives of this study.

L120-121 "SoSo 5 rivers cruise and at the Tomioka port and Hasaki and discuss the behaviour of  $^3\text{H}$  in the coastal region of Fukushima."

Please specify. From figure 1, it is hard to find out where stations are. The authors should modify the station map of figure 1.

L122-124 "We also present results on the  $^3\text{H}$  contents found in the fish filet collected close to the FNPP1 site. These results are also discussed using the already published  $^3\text{H}$  activity concentrations of river and open-ocean waters."

As commented in the abstract, there is no discussion of tritium levels in marine biota. I think that the authors have only discussed tritium flux from rivers and FNPP1, and made comparison of the flux from between river and the area in the proximity to the FNPP1. The authors should revise the objectives.

L177-179 "In 2014–2015, the  $^3\text{H}$  activity concentrations at coastal stations of Mano-1, Niida-1, Odaka-1, Uedo-1, and Tomioka port ranged from  $90 \text{ Bq m}^{-3}$  to  $175 \text{ Bq m}^{-3}$ , while the  $^3\text{H}$  activity concentrations at Niida-5, Odaka-5, and Uedo-5 stations,"

It is difficult to understand where stations are located from Fig.1

L179 "11–15 "

This range was 12-16 km in the abstract. Which is correct?

L186-187 "Therefore, the  $^3\text{H}$  activity concentration at 56N of the FNPP1 site was significantly high compared to that in the surrounding waters both north and south of the FNPP1 site, as shown in Figure 2."

The comparison between data of 56N site with DL of  $<1600 \text{ Bq/m}^3$  and that in other areas with DL of  $< 100 \text{ Bq/m}^3$ ? is not appropriate because the mean value and concentration range can be different among 56N and other stations if DL is different between them.

L194-203 " $^3\text{H}$  activity..."

The explanation on the activity concentration of tritium in fishes is only here. The authors should delete all the sentences related to this topic since it is difficult to relate the level of tritium in fishes to the main body of this study from only this information and less discussion.

L206-207 "Fukushima Dai-ichi Nuclear Power, hereafter FNPP1, "

This specification should be done in the Introduction section.

L208 "FNPP2"

FNPP2? Please specify. This is not plotted on Fig 1.

L210-211 "in general"

What is "general" for? The difference can't be observed from Figure 4 since the both two shapes of plots are almost same from this resolution of Fig. 4. And sampling dates are different between river and port stations. Change of water/weather conditions due to different sampling date should have affected the tritium and Cs concentration in two sampling stations. Furthermore, the authors should check the difference in activity concentrations using statistical analysis.

L239-241 "This estimate ( $0.05 \pm 0.03$  PBq) is slightly lower than that obtained by Povinec et al. (2013) using the same method ( $0.3 \pm 0.2$  PBq) for the samples collected in June 2011 from the south of the FNPP1 site during the KOK cruises (Buessler et al., 2012)."

I do not think that 0.05 PBq is slightly lower than 0.3 PBq. The explanation of the difference is necessary.



L250-254 "During.... 10 during the period from 2013 to 2016 (Figure are not shown)."

A figure showing the decline of the ratio with time is necessary. A detailed explanation of the reason is highly required because there could be process causing fraction between Cs and tritium during the water movement, other than the decontamination effort of TEPCO. Salinity change could also affect the ratio, however there were no description concerning the change in the ratio along the salinity gradient.

L254-256 "This fact may consistent with lower the  $^3\text{H}/^{137}\text{Cs}$  activity ratio observed in stagnant water (Nishihara et al., 2012) and in coastal waters close to FNPP1 (Povinec et al., 2013, Takahata et al., 2019)."

I think this sentence is grammatically wrong.

L258 "Matsumoto et al. (Matsumoto et al., 2013)"

Is that correct reference style? Please check.

L260-261 "Tsukuba, Kashiwa, and Hongo"

Indicating location and values on figure would be easy to understand for readers.

L266 "Tamura-shi"

Same as above comment

L268 "Niigata city"

Same as above comment

L269 "no significant"

Did the authors check statistically?

L273-274 "by  $^3\text{H}$  levels around  $70 \text{ Bq m}^{-3}$  as indicated previously."

Was the level consist with the observed values during the typhoon period?

L277 "< 650"

550 is minimum value from Aoyama et al., 2021c. Why did the authors describe like <650? All readers wrongly recognize that DL is about 650.

L277 "(Aoyama et al., 2020c), "

This is not listed in Ref.

L277 "which were slightly higher than"

The authors should carry out statistical analysis.

L277-278 "that in precipitations"

Were those values obtained from the same area? The minimum concentration by Fukushima Pref was over 300 Bq/m<sup>3</sup>. Since the detection limit of the tritium analysis conducted by Fukushima Prefecture could be higher than that by Tagomori et al., it may be impossible to make an appropriate comparison.

L281-282 "approximately 300– 500"

<330-510 is correct range.

L287 "Hokkaido, Gifu, and Okinawa

Same as other areas: Please indicate location.

I do not know where those are located. For example, from the map, Hokkaido is the name of a province whose area exceeds 80,000 km<sup>2</sup>. Tritium levels are dependent on sampling points, so information on sampling location may be necessary.

L290-293 "that the 3H .... decreased afterward. "

The authors should conclude this trend from the statistical analysis. For example, was there significant difference in tritium between 2014 and 2018-2019?

L297 "24 Bq"

It is too low. 240 Bq/m<sup>3</sup>?

L298 "Watanabe et al., 1991"

This study is not cited in Ref

L306 "in Table 4"

Table 4 is not listed in this manuscript. Or estimated values from the intercept in Table 1?

L311-313 "before the ...or 5–8 GBq day<sup>-1</sup> from both FNPP1 and FNPP2. "

Please explain how the authors estimated the amount of the discharge.

L313-316 "After the FNPP1 accident," no difference was observed before the accident among the monitoring stations."

After the FNPP1 accident, relatively lower tritium values suddenly appeared on Fig. 8. I guess that analytical procedure during the year periods of 2011-2019 seemed to be different from before 2010. The authors should explain that. I could not find out where those values were observed. The authors should indicate sampling points.

L323 "0.05 m s<sup>-1</sup>"

How did the authors obtain this rate?

L323-325 "freshwater flux from the rivers ... along the Fukushima coast."

References concerning river discharge are necessary. Sakuma et al. (2019) in Table A2? If so, please indicate this on the manuscript. Please explain how to calculate the fluxes. From parameters on Table A2, it is hard to understand. Also, I could not find out how the authors obtained those parameters.

L327-329 "We obtained the estimates for June 2014 and October 2014, and also with very heavy rains encountered in October.... are listed in Table 3"

Please describe more detailed explanation for the estimation of flux.

L332 "(Kanda 2012)"

This is not listed in references.

L333 "Monoageba"

?

L336-339 "In contrast, considering ....  $^3\text{H}$  flux from the FNPP1 site was found to be 28 to 86 GBq day<sup>-1</sup> as shown in Table 3,"

Please describe more detailed explanation for the estimation of flux for readers.

L340-341 "One of the reasons ... and the port of FNPP1."

I could not understand what the authors explain. Please explain more detail. I think this topic is the most important in this study.

L342 "Conclusion"

I do not think this section is conclusion.

L416 "References"

Please check the reference style. The number of <sup>137</sup>Cs or <sup>134</sup>Cs should be superscript. Also please check references whether these are cited in the main body. There are papers written by another language.

L518-520

?