

## Replies to Comment on bg-2020-491 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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Comments on "Impact of typhoons on particulate and dissolved  $^{137}\text{Cs}$  activities in seawater off the Fukushima Prefecture: results from the SOSO 5 Rivers cruise (October 2014) (bg-2020-491)"

Recommendation: Accept, with major revisions noted.

General comments: I reviewed the manuscript " Impact of typhoons on particulate and dissolved  $^{137}\text{Cs}$  activities in seawater off the Fukushima Prefecture: results from the SOSO5 Rivers cruise (October 2014) (bg-2020-491), submitted by Aoyama et al to Biogeosciences. The authors measured  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in the dissolved and particulate samples contaminated by the Fukushima Dai-ichi Nuclear power plant (FDNPP1) accident, which presented some new data. Their spatial distribution reflected the mixing of coastal water and open-ocean water. The  $^{137}\text{Cs}/^{134}\text{Cs}$  activity ratio derived from FDNPP accident is used to trace the source of riverine particle, which is very interesting. They also discussed the impact of typhoons on particulate and dissolved  $^{137}\text{Cs}$  activities in seawater off the Fukushima prefecture, but did not give a clear picture about the impact of typhoon on  $^{137}\text{Cs}$  activities in seawater. The novelty of this study needs to be improved. Additionally, decisions made with respect to data presentation combined with grammatical and other organizational errors result in a MS that lacks clarity and is difficult to follow. It is necessary to polish this manuscript by a native English speaker. Therefore, it is recommended to be published after major revisions.

**A: The manuscript will be revised in highlighting the novelty of the study that is to say two end-members mixing between river water and open sea water, the decoupling between dissolved and the organic particulate fraction extracted as shown by the trend of their respective  $^{137}\text{Cs}/^{134}\text{Cs}$  ratios with distance from the coast and finally the use of multi-days API that shows relationship between precipitations and  $^{137}\text{Cs}$  concentrations**

**In addition, as suggested the manuscript will be reorganized to give it more clarity and make it easier to read. Figures will be improved as well as English**

**The specific and technical comments indicated below will be taken into account.**

Specific comments

-Line 14: What is indicated by the dissolved activities.....?  $^{137}\text{Cs}$  or  $^{134}\text{Cs}$ ?

**A: Both  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  in dissolved form and extracted organic form decreased with distance from the coast due to mixing between river water and open sea water.**

-Line 18, "ranged from....to ....." means a range of variation, so the uncertainty in this sentence that "the ranged from  $0.01 \pm 0.00$  to  $0.12 \pm 0.01$ " is redundant? Please note this in the MS.

**A: The ratios shown here are based on two radioactivity measurements results, therefore ratios have also uncertainties.**

-Material and methods: What are the detection limits of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ?

**A: The detection limit for net  $^{137}\text{Cs}$  activity is about a few mBq per sample and that for  $^{134}\text{Cs}$  is about 10 mBq per sample. The measurements were carried out at the Ogoya underground laboratory which is located 270 meter water equivalent deep.**

-Lines 169-171, this sentence (the ratio of particulate  $^{137}\text{Cs}$  activity concentration.....) is confusing, please rephrase it.

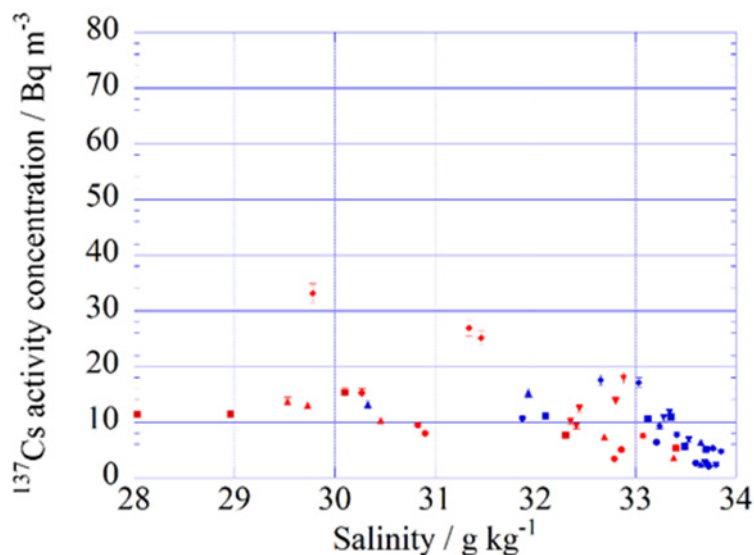
**A: Ok, we will do so.**

- Lines 179-181. This sentence is too long and needs revise to improve clarity and the flow....

**A: Ok, we will do so.**

-Lines 206-211, what's meaning that "data not shown or figure not shown"? Add in the Supporting information?

**A: all salinity data and radiocaesium activity concentration in dissolved form data are presented in Aoyama et al., 2020a. So, (figure not shown; data are in Aoyama et al., 2020a) at line 209-201 is correct and (data not shown) at line 207 should read as (figure not shown; data are in Aoyama et al., 2020a) in the original text. By the way, due to Rc1's comments, new figure4 will be in the coming revised article. So, both statements will be replaced as (see figure4, data are in Aoyama et al., 2020a).**



**Fig. 4. Relationship between salinity and  $^{137}\text{Cs}$  activity concentration in dissolved form.**

**Mano river transect: solid square**

**Niida river transect: solid circle**

**Odaka river transect: solid up-pointing triangle**

**Ota river transect: solid down-pointing triangle**

**Ukedo river transect: solid diamond**

**For 5 rivers transect, red is for surface data and blue is for bottom data.**

-Discussion section: The discussion was not enough and some conclusions are soft or from conjecturing, for example, "this pattern might reflect complex physical processes....."(lines 210-211); "Possible explanation of this finding are that the radiocaesium in the coastal seawater....."(Lines 236-239).

**A: We will revise the discussion and conclusion based on the statement regarding with novelty of this article. New findings and novelty in our article are shown below.**

**1, We highlighted simple two end-members mixing between river water and open sea water after flooding due to heavy rain showed by data in Fig. 8, Salinity vs.  $^{137}\text{Cs}$  activity concentration. Extrapolation to salinity equal zero gave us an appropriate number of  $^{137}\text{Cs}$  activity concentration in dissolved form at the mouth of the Ukedo river. This is the first data set of this kind.**

**2, Contrary to the dissolved phase,  $^{137}\text{Cs}/^{134}\text{Cs}$  ratio decay corrected to 11 march 2011 in the extracted organic fraction (extracted by conc.  $\text{H}_2\text{O}_2$ ) did not change with distance from the coast underlying the decoupling of these two phases Dissolved phase follows a classical two end-member mixing while the extracted organic phase is characterized by ratio of the FDNNP1 releases even at distance from the coast**

**3, The amount of precipitation for the day and  $^{137}\text{Cs}$  activity concentration were not correlated as shown in Fig. 12 (fig 17 see comment on figure 12-16 and 19 later in the text below), 1day API vs.  $^{137}\text{Cs}$  activity concentration which clearly indicated that a simple saying like "heavy rain is a cause of higher  $^{137}\text{Cs}$  activity concentration" is far from reality. Indeed, a timelag of 10 to 60 hours is generally observed between the time of the rains and the increase in the (total) flow of the rivers depending on the watershed scale including the magnitude of the slope gradient of the Tohoku region in Japan. In addition, the main inputs of radiocaesium from land to the ocean are via riverine particle fluxes (Sakuma et al., 2019). It is therefore necessary to consider also the delay due to the phase change of part of the caesium fixed on the particles towards the dissolved phase at river mouth due to change in physico-chemical conditions, especially ionic strength. According to Delaval et al (2020) two first-order reactions govern the kinetics of the process, with half-life reaction times of 1 h and a few days depending on the sites of cesium binding on the particles. Therefore, a delay of a few to several days can be observed between the time of heavy rains and the increase of dissolved cesium in the coastal area in connection with biogeochemical and hydrological processes.**

**These background knowledges underpin our finding that the dissolved radiocaesium activity concentration shows a correlation with several days API, and not with the amount of the precipitation of the day.**

-Conclusions section: The conclusion section seems long with too much information on some discussion that appears unnecessary. The conclusion should be rephrase.

**A: Yes, we will do it. RC1 also gave us similar comments. We will revise the conclusion section taking into account your comments as well as those of RC1, in order to make it clearer and better underline the results of this study. While these changes are provisional and will be revised again, some concluding paragraphs which will be included in the upcoming revised article are presented below to meet your request.**

**The detection of both  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in all dissolved samples demonstrated contamination from the FNPP1 accident with higher values in the surface layer compared to the bottom layer and a clear decrease with distance from the coast. The decrease of caesium activity concentration with increasing salinity reflected mixing of coastal water with open-ocean water.**

**At the stations very close to the coast relatively high  $^{137}\text{Cs}$  activities in all particles with values exceeding the dissolved  $^{137}\text{Cs}$  activities were observed. Beside,  $^{137}\text{Cs}$  activities were generally one or two orders of magnitudes lower in the extracted organic fraction than in the dissolved fraction reflecting the fact that the organic component of the particulate matter sampled is not the main carrier of cesium.**

The  $^{137}\text{Cs}/^{134}\text{Cs}$  activity concentration ratio in the dissolved phase changed drastically due to mixing with open-ocean water. In contrast, this ratio in the extracted organic fraction did not change with distance from shore or with  $^{137}\text{Cs}$  activity concentration and generally remained close to 1, even at locations far from the coast. This pattern indicates a decoupling between these two phases with the extracted organic fraction characterizing particle originating from land even at distance from the coast.

When considering  $^{137}\text{Cs}/^{134}\text{Cs}$  ratios, the source of radiocaesium in the coastal region north of FNPP1 should be related to releases from the core of Unit 1 of FNPP1, while in the coastal region south of FNPP1 the source should be a mixture of releases from the core of Unit 2 and the core of Unit 1 of FNPP1.

$^{137}\text{Cs}$  activities on the Fukushima coast and the modified 7-day API showed a good positive relationship with the exception of 56N canal of FNPP1. In fact, at this location, high  $^{137}\text{Cs}$  activities are observed apart from an event of heavy rain.

This study shows the need to better characterize the inputs of rivers in taking into account on one side their hydrologic characteristics and on the other hand the nature of their solid discharges and phase change from particle to dissolved form. Special attention should be paid to the organic fraction of the riverine inputs as well as their fate in the coastal environment in the area which has been impacted by the accident.

-Data availability: it should be moved in the Material and methods?

**A: As far as I understand, we need to have an independent section about Data availability based on Biogeoscience journal.**

-References: please unify the format of periodicals. For example, Scientific Reports (Line 349); J. Radioanal. Nucl. Chem. (Line 356).....

**A: Yes, we will do so.**

-Figures: these figures are not clear, please redraw.... For example, fig.2 and fig.9.

**A: Figures, 3 to 7 were revised and new figures are included in the reply to RC1. We will refine fig.2 and fig.9 later. We will delete figures 12-16 and 19 based on comments from RC1.**

**A: Technical issues stated below will be revised appropriately.**

-Line 36: 'Nishihara et al. (Nishihara et al., 2012)' should be 'Nishihara et al. (2012)'.

-Line 43: 'Miura et al. (Miura et al., 2020)' should be 'Miura et al. (2020)'.

-Line 57: '(Nagao et al., 2014)' should be 'Nagao et al. (2014)'.

-Line 115: '...the Low Level Radioactivity Laboratory At some stations, ....' should be '...the Low Level Radioactivity Laboratory. At some stations, .....'.

-Lines 128, 320, 324: 'Dataset of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  activity...' should be 'Dataset of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  activity...'.  
and  $^{137}\text{Cs}$  activity...'.  
-Line 188: '.....in the rivers, indeed Naulier et al. (Naulier et al., 2017)' should be '.....in the rivers. Indeed, Naulier et al. (2017)'.

-Line 224: 'Tsurutal et al., (Tsuruta et al., 2014)' should be 'Tsurutal et al. (2014)'.

-Line 227: '1.06 (+-10%)' should be '1.06 ( $\pm 10\%$ )'.

-Lines 235, 310: '0.92 (+-10%)' should be '0.92 ( $\pm 10\%$ )'.

-Lines 314-316: '7-day, 5-7day' should be '7-days, 5-7days'.

-Line 409, delete the Japanese language

-Lines 358-359, 410, 416, 420, 423, Superscript:  $^{137}\text{Cs}$

-Table1, "+-" changes " $\pm$ "

End of replies to RC2.

**We also need to revise a part of method section as below.**

**In the current text I made a copy paste mistake. We only used conc. H<sub>2</sub>O<sub>2</sub> with pure water, and did not use conc. HNO<sub>3</sub>. The Actual method for getting the organic fraction is given below.**

**On the filters there were yellow-brown matters for all surface layer samples. Then we poured H<sub>2</sub>O<sub>2</sub> step by step about 2 ml each time. We stopped adding H<sub>2</sub>O<sub>2</sub> when the color disappeared. The fraction dealt with in this paper corresponds to this soft extraction. We revised this part as appropriately as below.**

**In the current text, we stated that “Organic form of radiocaesium of the samples were obtained by disillusion of organic portion on the filter using concentrated nitric acid and concentrated hydrogen peroxide at all stations, then filled and dried up in a Teflon tube.”.**

**We will revise as below based on our lab note by my staff.**

**We performed filtration using membrane filter with a pore size of 0.45- $\mu$ m (Millipore HA). The filters were dried up and weighed to determine the mass of particles on the filter. Then, 4ml conc. H<sub>2</sub>O<sub>2</sub> were poured on the filter placed in a plastic try three times with an interval of a few days, 5 Dec.2014, 8 Dec. 2014, 10 Dec. 2014 for all treated filters. And 4ml pure water were added on 8 Dec. 2014. Then if orange or brown parts still exist, we added pure water 4ml and 4ml con.H<sub>2</sub>O<sub>2</sub> again on 15 Dec. 2014 to about half the number of filters. The end point was the visual inspection of the color disappearance. The solutions were put in a Teflon tube and measured at Ogoya laboratory after drying. Therefore, in the revised article we will mention extracted organic fraction instead of organic particles**

End of reply.