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Discussions

Interactive comment on "Export of ^{134}Cs and ^{137}Cs in the Fukushima river systems at heavy rains by Typhoon Roke in September 2011" *by* S. Nagao et al.

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Thank you very much for your comments. We have discussed their comments carefully and have made corrections which we hope meet with the approval.

Major comments One specific comment I would like to note is about the interpretation of the difference in values of the distribution coefficient (Kd) (Eqn. (1), p.2774, line 3) between those in the present study and those observed in other locations (other Japanese and Ukrainian rivers). They differ each other by more than two orders of magnitude (p.2773, line 12). According to the present authors' interpretation, the reason for the large difference is relevant to origins of suspended matter (p.2774, line

13-14; line 24-26). Namely, in the studied case, the suspended matter is a result of 'direct input of suspended solids eroded from the ground surface' (line 24-26), while the suspended matter in other referred studies is supplied 'from the watershed and resuspension of river bottom sediments by rain events'. I have two concerns about the interpretation.

1) Firstly no concrete evidences have been shown for identification of the sources in those cases (the present case and the literature ones). It seems that abovementioned assignment of the sources are rather speculative. 2) Secondly, addressing the variety of origins of suspended matter cannot solely explain differences in Kd values. One should further explain how the origin can influence on the nature of adsorption of cesium on suspended matter. Therefore, I feel it is necessary to reinforce the interpretation from logical and experimental aspects. Comparison of Kd values between those under a typhoon event and those under non-typhoon events may afford some experimental evidences, for example.

Answer The distribution coefficient, Kd, is an important parameter to estimate transport behavior of 137Cs in river systems using model simulation. The Kd is defined as Eqn. (1) under the equilibrium concentration of a radionuclide in the solid phase divided by the concentration of the radionuclide in the aqueous phase. However, after a heavy rain event, the radioactivity of 137Cs may vary with water discharge so that the Kd is 'apparent' value. At that time, river systems have two processes: 1) equilibrium of 137Cs sorption between river water and suspended solids, and 2) supplied 137Cs associated with suspended solids from river bottom, riverbank and surface soil. When the suspended solids associated with 137Cs irreversibly are supplied from the watershed, the Kd values apparently increase because of high 137Cs radioactivity content for the suspended solids. To understand the effects of input of suspended solids on Kd value, we compared the Kd values after heavy rain events due to typhoon in September 2011 and July 2012. The suspended solids concentration was 0.20–0.41 g L-1 for the September samples and 0.26–0.34 g L-1 for the July samples. The dissolved radiocesium concentration was almost constant, 0.0025–0.0046 Bq L-1 in both sampling date. The Kd value for the July samples is 0.20–0.43 x 106 ml g-1 and still high, though the values are 1/2–1/10 lower than that of 2011 September samples. These results suggest that the sources of particulate forms of radiocesium are important factors controlling Kd values at the second dispersion after the nuclear accident. The amount of 137Cs transported easily decreases with increasing time after the Fukushima Daiichi NPP accident.

We also discussed the 137Cs radioactivity in river bottom sediments and surface soil of riverside in the Same River referred from the Ministry of the Environment in Japan. The radioactivity of 134Cs and 137Cs in the bottom sediments from the lower Same River (Samegawa-Ohashi) was 700–770 Bq kg-1 on May 28, 2011 and 1800–2000 at high water flow on July 1, 2011 (Ministry of Environment in Japan, 2012). On the other hand, the radioactivity ranged from 10 to 250 Bq kg-1 during September 2011–November 2012 and 1–2 orders lower than the river suspended solids.

The radioactivity of 134Cs and 137Cs in the surface soil of riverside at Samegawa-Ohashi in the downstream of Same River ranged from 22 to 970 Bq kg-1 during September 2011–November 2012 (Ministry of Environment in Japan, 2012). The radioactivity at Idosawa-Ohashi in the upper downstream was from 70 to 900 Bq kg-1 during September 2011–July 2012, but 1500-7100 Bq kg-1 on September 2 and November 9 in 2012 at high water level condition. The results suggest that 134Cs and 137Cs were deposited on the ground surface in the riverside due to the effects of rain events.

Consequently, we consider that the reason for the large difference is relevant to origins of suspended matter, that is, the suspended matter is mainly a result of direct input of suspended solids eroded from the ground surface by rain events at the initial stage after the Fukushima Daiichi Nuclear Power Plant accident.

We corrected “3.3 Migration behavior of 134Cs and 137Cs in river systems” as follows:

Distribution coefficient (Kd) between suspended solids and river water is defined as Kd = Csolid/Cdissolved , (1) where Csolid and Cdissolved respectively denote the 137Cs concentrations in the suspended solids (Bq/g) and dissolved phase (Bq/ml). The fate and bioavailability depend strongly on the Kd and strength of the particle-contaminant association. Estimation of Kd values was conducted using measurement data presented in Tables 1 and 3. The Kd is 0.43–0.55 × 106 ml/g for the Natsui River and 4.1–5.0 × 106 ml/g for the Same River. These values are 1–2 orders higher than those of other Japanese rivers such as the Tone River and the Ishikari River (Hirose et al., 1990) and the Kuji River (Matsunaga et al., 1991) before the Fukushima Daiichi NPP accident. The Kd values of the Fukushima rivers are two orders higher than that of Ukraine after the Chernobyl accident (Matsunaga et al., 1998), which is regarded as supplying suspended solids from the watershed and resuspension of river bottom sediments by rain events.

The cumulative 134Cs and 137Cs inventory from the surface down to depth in undisturbed soils in Fukushima Prefecture confirms that >90% of the total 134Cs and 137Cs in the soil profile was found within the upper 5 cm layer at the cropland and grassland sites (Koarashi et al., 2012). The surface erosion processes in watershed have been studied using 137Cs derived from fallout as a tracer of suspended solids. Surface runoff generally does not occur in forested areas, but unmanaged Japanese cypress plantations often have little surface cover, and surface runoff is generated during large rainstorms (Miyata et al., 2007; Gomi et al., 2008). Fukuyama et al. (2010) have shown that, for different stand species, surface coverage and forest management practices affect the runoff of the surface-derived suspended solids at the catchment scale. To understand the effects of input of suspended solids on Kd value, we compared the Kd values after heavy rain events due to typhoon in September 2011 and July 2012. The suspended solids concentration was 0.20–0.41 g L-1 for the September samples and 0.26–0.34 g L-1 (Nagao unpublished data) for the July samples. The dissolved radiocesium concentration was almost constant, 0.0025–0.0046 Bq L-1 in both sampling date. The Kd value of 134Cs and 137Cs for the July samples is 0.20–0.43 x 106 ml g-1 and still high, though the values are 1/2–1/10 lower than that of 2011 September samples. These results suggest that the sources of particulate forms of radiocesium are important factors controlling Kd values at the second dispersion after the nuclear accident. Direct input of suspended solids eroded from the ground surface may be reflected in the higher values found for the Natsui River and the Same River after the heavy rain at the initial stage of the Fukushima Daiichi NPP accident.

The radioactivity of 134Cs and 137Cs in the surface soil of riverside at Samegawa-Ohashi in the downstream of Same River ranged from 22 to 970 Bq kg-1 during September 2011–November 2012 (Ministry of Environment in Japan, 2012). The radioactivity at Idosawa-Ohashi in the upper downstream was from 70 to 900 Bq kg-1 during September 2011–July 2012, but 1500-7100 Bq kg-1 on September 2 and November 9 in 2012 at high water level condition. The results suggest that 134Cs and 137Cs were deposited on the ground surface in the riverside due to the effects of rain events.

The radioactive content of 134Cs and 137Cs deposited in river bottom sediments was approx. 770 Bq/kg-sediment for the Natsui River and approx. 2000 Bq/kg for the Same River during May-July 2011, but 10-250 Bq kg-1 during September 2011-November 2012 (Ministry of the Environment, 2012). The radioactivity varied with sampling, although the samplings were conducted at fixed stations in each river. The river bottom sediments are sandy, so the apparent residence time of fine particles might be short in the Natsui River and the Same River. Therefore, the contaminated area around the river basin and river bottom sediments plays an important role as sources of particulate phase of 134Cs and 137Cs in river waters.

Fig. 6 presents a schematic illustration showing the export of radiocesium from the watershed to a river. Erosion of riverbank and the ground surfaces, and re-suspension of river bottom sediments occurs at rain events so that the amount of suspended solids increases and the radioactivity of 134Cs and 137Cs associated with riverine suspended solids also increases. Similar results have been reported for the Kuji River, running

through Ibaraki and Fukushima prefectures (Matsunaga et al. 1998). Increased erosion and radioactivity of 137Cs derived from global fallout have also been observed at the Kuzuryuu River in 2009 after rain events (Nagao, unpublished data).

Minor comments

1) Two similar terms, 'suspended matter' and 'suspended solids' are used in the manuscript. Is this wording really necessary? Answer: We revised one term, "suspended solids".

2) In Table 2, foot note c: 'Proceeding' should be 'Precedent' or 'Preceding'. Answer: We revised it to "Preceding".

3) In Table 4, adding a column of catchment area would help readers' understanding because the export flux is dependent on the area. Moreover, an indication of the major origin of exported 137Cs (global fallout, or a nuclear accident) for each case would also be helpful. Answer: We added two columns such as catchment area and the major origin.

Please also note the supplement to this comment:
http://www.biogeosciences-discuss.net/10/C1811/2013/bgd-10-C1811-2013-supplement.pdf

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