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Biogeosciences
Discussions

Interactive comment on "Determination of plutonium isotopes in marine sediments off the Fukushima coast following the Fukushima Dai-ichi Nuclear Power Plant accident" *by* W. T. Bu et al.

W. T. Bu et al.

jzheng@nirs.go.jp

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We thank the referee for the constructive and detailed comments. It help us a lot to improve our manuscript. Our responses are detailed below.

General comment: This paper measured Pu isotope concentrations and isotope ratios in sediments collected near FDNPP at the western Pacific as well as in estuarine sediments of eastern Japan. And it concluded no detectable Pu contamination originating from the FDNPP accident injected to that region during the year since the accident. This paper presented the good quality of data enough for approaching the reliable conclusion. And further this document was well structured and well written. However, the

conclusion of no detectable Pu contamination from FDNPP accident could be misunderstood to be negligible Pu discharge from FDNPP accident even if there have been reports of Pu signals from the FDNPP accidents in soils and settling particles. The authors presented the evidences such as similar inventory of Pu to those before accident and negligible 241Pu in sediments. And, using two end-members of global fallout and PPG, the contribution of PPG was estimated based on 240Pu/239Pu ratio. Since Pu could be recycled within the water column, the amount of deposited Pu should be lower than that of discharged Pu. And since total discharged Pu quantity from FDNPP accident would be much lower than total amount deposited for past 48 yr, it would be difficult to identify the signals of Pu from FDNPP accident with inventory data. For 240Pu/239Pu ratio, PPG and FDNPP showed similar values, 0.36 and 0.303-0.330, respectively. And thus, it is also difficult to differentiate the origin of Pu from FDNPP accident with PPG. In conclusion, with Pu activity and isotope ratios in deep sea marine sediments, it seemed to be difficult to identify the contribution of Pu from FDNPP accident.

Response: Cs contamination from the FDNPP was detected in deep sea sediment traps one month after the accident (Honda et al., Biogeosciences Discuss., 10, 2455-2477, 2013). As Pu isotopes are more particle-reactive than Cs (Kd value of Pu is two orders of magnitude higher than that of Cs), the possible deposited or discharged Pu into the ocean would quickly incorporated with the marine sediments. The reviewer is right that the total discharged Pu from the FDNPP accident could be much lower than the total amount deposition for the past decades, which makes it difficult to identify the FDNPP Pu signal. However, the 241Pu results can give direct information. The FDNPP derived Pu was characterized with high 241Pu concentration (up to 34.8 mBq/g) and high 241Pu/239Pu atom ratio (> 0.1) (Zheng et al., Sci. Rep., 2:304, doi:10.1038/srep00304,2012). For all the sediment samples we measured, no 241Pu was detected except in ES4 at a depth of 3-4 cm, which had a 240Pu/239Pu atom ratio of 0.196 and a 241Pu/239Pu atom ratio of 0.0027, typically lower than that from the FDNPP. In addition, for the vertical distribution of 240Pu/239Pu atom ratio in the sediment

cores, the early deposition from the PPG was mainly distributed in the deeper layers (> 5cm). The 240Pu/239Pu atom ratio in the upper layer (< 5 cm) sediments was around 0.24 due to the mixing of the PPG fallout and the global fallout. From our results, we can see that all the 240Pu/239Pu values in the upper layer sediments were below 0.30. From these points of view, we concluded that no detectable Pu contamination from the accident in the marine sediments collected outside the 30 km zone from the plant site up to the sampling time. It has been modeled recently that if any discharged Pu contamination from the FDNPP occurred, it would remain in a very close area because of the low mobility of plutonium isotopes in the marine environment (Perianez et al., J. Radioanal. Nucl. Chem., doi 10.1007/s10967-013-2422-1, 2013). Thus for the marine environment inside the 30 km zone from the plant site, further researches are needed to give a more comprehensive conclusion. We revised our manuscript by adding the above discussed contents in page 14, line 25-28.

Specific comments: 1. p. 649 24 lines; organic matter content -> ignition loss. The weight loss by the ignition has often been used for organic matter content. However, it includes lattice water as well as organic matter, especially for fine-grained marine sediments. So, it is suggested that the ignition loss should be used instead of organic matter.

Response: We agree the correction that “organic matter content” should be replaced by “ignition loss” and it has been done in the revised manuscript.

2. p. 651 15-22, the concentration of 239+240Pu activity in. . . . .from 0.003 to 1.191 mBq/g -> the values are different from those in Fig.1 caption.

Response: In the revised manuscript, we changed the corresponding text to “the concentration of 239+240Pu in the Japanese estuary surface sediments ranged from 0.003 to 5.81 mBqg-1”; Accordingly, we changed the Fig.1 caption text to “The surface sediment Pu concentration range was between 0.003-5.81 mBqg-1”.

3. p. 652 4 lines, its marginal seas is given -> its marginal seas in the literatures

(references) is given.

Response: In the revised manuscript, we changed the corresponding text to “its marginal seas in the literatures (Buesseler, 1997; Dong et al., 2010; Hong et al., 1999; Ito et al., 2007; Kim et al., 2003; Lee et al., 1998; Lee et al., 2003; Lee et al., 2004; Lee et al., 2005; Liu et al., 2011a; Moon et al., 2003; Nagaya and Nakamura, 1992; Oikawa et al., 2011; Otosaka et al., 2006; Pettersson et al., 1999; Wang and Yamada, 2005; Yamamoto et al., 1990; Zheng and Yamada, 2005, 2006b,c) is given”

4. p. 655 24 lines-p. 656 9 lines, organic matter content -> ignition loss. The description for the relationship of Pu with organic matter seemed to be useless because there were little general relationships.

Response: We agree. Considering that our study was not focused on the Pu fractionation in the sediments. We removed Fig. 5 to supplement. The description was revised accordingly. See page 10, line 16-18.

Technical comments: 1. p. 659 1line, hypothesize -> Hypothesis.

Response: The revision has been done in the revised manuscript.

2. p. 660 eq. 3 RB -> RP

Response: The revision has been done in the revised manuscript.

Please also note the supplement to this comment:
http://www.biogeosciences-discuss.net/10/C422/2013/bgd-10-C422-2013-supplement.pdf

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Interactive comment on Biogeosciences Discuss., 10, 643, 2013.