Grain size distributions of radiocesium and Plutonium activity concentration in sediments collected off the Niida River estuary

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After the accident at the Fukushima Dai-ichi Nuclear Power Station (FDNPS) happened in March 2011, large amounts of radionuclides released from the FDNPS into the terrestrial and marine environments. The total amounts of ^{134}Cs and ^{137}Cs released from the accident were estimated as 18 PBq and 15 PBq, respectively. In contrast, those of ^{238}Pu, ^{239}Pu and ^{240}Pu amounts were estimated as 0.0019 PBq, 0.0000032 PBq and 0.0000032 PBq and these amounts were not many compared to abundance before the accident (Report of Japan government to the IAEA Ministerial Conference on Nuclear safety, 2011). Based on the Pu atom ratio, it was estimated that the release of Pu from the accident was negligible in the marine environment (Bu et al., 2015). However, previous reports focused on the river (Evard et al., 2014) and offshore area (e.g. Zheng et al., 2012, Bu et al., 2013, 2015) and the lack of information on the distribution and behavior of plutonium in the estuarine area hampered the understanding of the process of radionuclide transport from river to ocean. In this study, the Niida River estuary was focused on, because the upstream portion of this river is located in Iidate Village, which was an area of high radiocaeasium deposition from the accident. We discussed temporal and vertical distributions of radiocaeasium and plutonium based on the results of the radiocesium (^{134}Cs, ^{137}Cs) and plutonium (^{239}Pu, ^{240}Pu, ^{241}Pu) activity concentrations and plutonium atom ratios (^{240}Pu/^{239}Pu, ^{241}Pu/^{239}Pu) according to grain size in sediments.Sediment core samples at three monitoring stations (NR1: 37˚39' N, 141˚04' E, water depth: 25 m, NR2: 37˚41' N, 141˚09' E, water depth: 30 m, NR4: 37˚38' N, 141˚08' E, water depth: 35 m) were collected in mid-October 2013. Collected sediment cores were cut into 1 cm thick slices and dried. Then, the dried sediments were separated into four classes, based on grain sizes, using several mesh sizes: granules (grain size larger than 2 mm); very coarse to coarse sand particles (1-2 mm); coarse to very fine sand particles (0.063-1 mm); and silt to clay particles (smaller than 0.063 mm). Radiocesium (^{134}Cs and ^{137}Cs) activities were measured for each grain size class using high-purity gamma ray spectrometry and then corrected to the sampling date. Plutonium (^{239}Pu, ^{240}Pu and ^{241}Pu) were extracted and concentrated based on Wang et al. (2017) and measured using SF-ICP-MS (Zheng et al., 2006; Zheng, 2015).

Fractions for the classes of granules, very coarse sand, coarse to very fine sand, silt to clay particles were: 0.0-35%, 0.013-35%, 38-99%, and 0.0-29%, respectively. The fractions for coarse to very fine sand particles represented more than 70% of the total particle amount for each sediment layer and the highest fractions were obtained at NR1 and NR2, which are located northward from the river estuary. In contrast, fractions for granules and very coarse sand particle at NR4, which is located in an area of the same latitude as the river estuary, were relatively high and the total fraction for these particles ranged from 20-62 %. The ^{137}Cs activities for very coarse sand, coarse to very fine sand, and silt to clay particles were in the ranges of 2.8-14 Bq/kg-dry, 4.1-751 Bq/kg-dry, and 731-837 Bq/kg-dry, respectively, and these activity concentrations tended to be higher with decreasing grain size. However, the profile patterns for the sand particles and silt to clay particles fraction were similar. In this presentation, we also report the results of grain-size distributions of Pu activity concentration and Pu atom ratio. This work was partially supported by Grants-in-Aid for Scientific Research on Innovative Areas, the Ministry of Education Culture, Sports, Science and Technology (MEXT), Japan (Nos. 24110004, 24110005), the JSPS KAKENHI (grant number JP17k00537) and Research and Development to Radiological Sciences in Fukushima Prefecture.

Keywords: radiocaesium, pulutonium, off the Niida River, sediment

