Existence forms and distribution characteristics of radiocesium in marine sediments in the coastal area off Fukushima Prefecture

Soichiro Terasaki[1], Seiya Nagao[2], Naoteru Odano[3]

[1]*Graduate School of Natural Science and Technology, Kanazawa University: Kakuma, Kanazawa City, Ishikawa, 920-1192 and sot_row_27@ezweb.ne.jp*
[2]*Low Level Radioactivity Laboratory, Institute of Nature and Environmental Technology, Kanazawa University: o24 Wake, Nomi City, Ishikawa, 923-1224 and nagao37@staff.kanazawa-u.ac.jp*
[3]*National Maritime Research Institute: 6-38-1 Shinkawa, Mitaka City, Tokyo, 181-0004 and info2@nmri.go.jp*

This study investigates the distribution characteristics of ^{137}Cs and the existence forms by using selective extraction experiments. Sediment samples were collected in the coastal area off Fukushima and Miyagi Prefectures in 2014 and 2015. Water content is related to the variations of radiocesium concentration. Dissolution rate of ^{137}Cs from the bottom sediment to artificial seawater ranges from 0.021 to 0.083% for the sediments off the Abukuma River and from 0.17 to 7.4% for those off the Niida and FDNPP. Extraction rate of radiosecium by 1M ammonium acetate and 10% hydrogen peroxide are 0.56-1.2% for the Abukuma and 3.0-23.5% for the Niida and FDNPP. The results indicate that most of radiocesium is adsorbed to clay minerals in the sediments and the variation of ^{137}Cs is controlled by the movement of sediment particles.

I. INTRODUCTION

Radiocesium was released into the environment because of the accident of FDNPP caused by the Great East Japan Earthquake and tsunami on 11 March 2011. The 3.6 ± 0.7 PBq of ^{137}Cs was released directly from the nuclear power plant[1] and 12-15 PBq was transported as the atmospheric deposition.[2] A part of radiocesium was adsorbed to the bottom sediments and their radiocesium concentration varied spatially and tempotrally.[3,4] The movement of sedimentary radiocesium and/or the dissolution of radiocesium into the seawater appear to contribute their variation. Radiocesium associated with suspended solids also transfers from river to the seabed sediments. The purpose of this study is to understand the factors controlling the variation of ^{137}Cs inventory and its spatial distribution in coastal sediment off Fukushima and Miyagi Prefectures, Japan.

II. SAMPLING AND METHOD

Marine sediments were collected in the coastal area off Fukushima and Miyagi Prefectures in 2014 and 2015. We selected three sampling areas as shown in Fig.1. Sediment samples collected off the FDNPP in 2014 were cut into 0-1, 1-2, 2-4, 4-6, 6-10, 10-14, 14-20 and 20-26 cm layers. Other sediment samples were cut into 0-3, 3-10, 10-20, 20-30 and 30-40 cm layers.

The sediment samples were freeze-dried and water content of sediment was measured. The samples were sieved through a sieve of 2 mm mesh. After homogenization, the samples were filled in a plastic container. Gamma rays of ^{134}Cs (605 and 796 keV) and ^{137}Cs (662 keV) were measured for the samples by using a Ge detector at Low Level Radioactivity Laboratory and Ogoya Underground Laboratory at Kanazawa University.

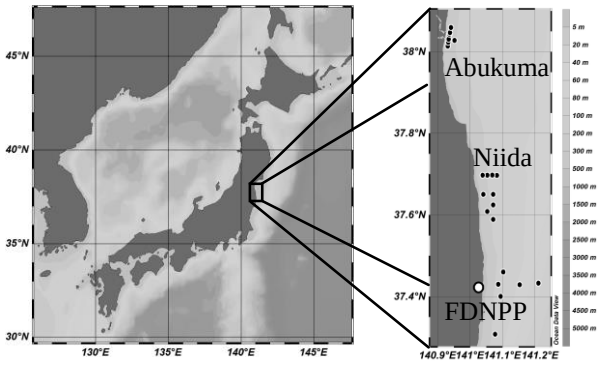

Fig. 1 Sampling location

Artificial seawater dissolution experiment was carried out to investigate dissolution of radiocesium from the bottom sediment to seawater. At this experiment, the sediment sample was shaken with artificial seawater at solid to solution ratio of 1:10 in a plastics bottle for two days at room temperature. After the dissolution experiment, extraction experiment of 1M ammonium acetate was carried out at the same solid-liquid ratio for two hours at 25℃ to investigate the exchangeable

fraction of radiocesium by ammonium ion. Extraction experiment using 10% hydrogen peroxide was also performed at solid-liquid ratio of 1:6 for six hours at 25℃ to study the association with organic matter.

III. RESULT AND DISCUSSION

^{137}Cs concentration in the surface sediments collected off the Abukuma ranges from 132 to 5880 Bq/kg-dry. The sediment samples off the Niida River and the FDNPP were 3.02 to 517 Bq/kg-dry and 14.7 to 2480 Bq/kg-dry, respectively. There is a good positive correlation between water content and ^{137}Cs concentration. ^{137}Cs inventory in the sediments at the depth of 0-10 cm layer is 12.1 to 333 kBq/m^2 for the Abukuma, 0.612 to 67.4 kBq/m^2 off the Niida, and 4.12 to 139 kBq/m^2 off the FDNPP.

The dissolution rate to artificial seawater ranges from 0.047 to 0.16% for the samples off the Abukuma River. The percentage of extracted radiocesium by 1M ammonium acetate ranges from 0.56 to 1.2%. The effect of the exchangeable radiocesium by of the ammonium ion is small. In the coastal sediments off the Niida River and the FDNPP, the dissolution rate to artificial seawater ranges from 0.38 to 14% and the percentage of extracted radiocesium by 1M ammonium acetate ranges from 1.7 to 24%. The major part of samples shows less than 10% extraction of radiocesium except for the four samples collected off the FDNPP (Fig. 2). The extraction rate by 10% hydrogen peroxide ranges from 0.079 to 0.72% for all sediment samples. The bonding of radiocesium to organic matter is small.

IV. CONCLUSIONS

Dissolution rate of radiocesium from the bottom sediment to artificial seawater and extraction rate by 1M ammonium acetate for the samples collected off the Abukuma River are very low values. The dissolution rate to artificial seawater and extraction rate by 1M ammonium acetate are less than 10% for the Niida and the FDNPP except for four samples. In addition, water content is related to the variations of radiocesium concentration. Therefore, the most of radiocesium in bottom sediment is adsorbed to clay minerals strongly and the transportation of sediment particles contributes to the variability of radiocesium in the surface sediment.

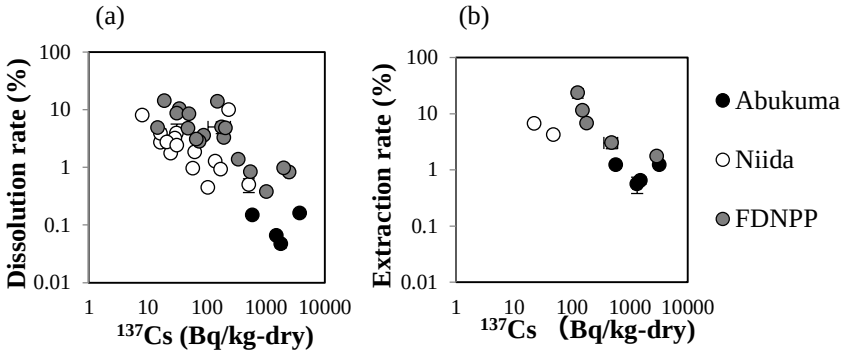

Fig. 2 Dissolution rate to artificial seawater (a) and extraction rate by 1M CH_3COONH_4 (b) as a function of ^{137}Cs radioactivity in the sediments.

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REFERENCES

1. Tsumune et al., "One-year, regional-scale simulation of ^{137}Cs radioactivity in the ocean following the Fukushima Dai-ichi Nuclear Power Plant accident", Biogeosciences, **10 (8)**, 5601 (2013).
2. Aoyama et al., "^{134}Cs and ^{137}Cs in the North Pacific Ocean derived from the March 2011 TEPCO Fukushima Dai-ichi Nuclear Power Plant accident, Japan. Part two: estimation of ^{134}Cs and ^{137}Cs inventories in the North Pacific Ocean", Journal of Oceanography, **72 (1)**, 67 (2016).
3. Kusakabe et al., "Spatiotemporal distributions of Fukushima-derived radionuclides in nearby marine surface sediments", Biogeosciences, **10**, 5019 (2013).
4. Otosaka and Kato, "Radiocesium derived from the Fukushima Daiichi Nuclear Power Plant accident in seabed sediments Initial deposition and inventories", Environmental Science: Processes Impacts, **16**, 978 (2014).