Introduction

A nuclear accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP) occurred after the 2011 Tohoku Earthquake and Tsunami. Radioactive materials with short to long half-life were released from the FDNPP. The radionuclides with short half-life, such as a few seconds or minutes, decayed after a few months after the accident. To estimate the impact of the radiation dose or human health, it is important to understand the dynamics of radionuclides, especially 134Cs and 137Cs, in aquatic environments because of large releases, relatively long half-life (134Cs 2.06 years, 137Cs 30.17 years) and higher contribution to external exposure. Monitoring surveys of radionuclides radioactivity have been performed at river and lake watersheds and coastal marine environments in Japan since the FDNPP accident on March 11, 2011. The river systems running through the Hamodori area, which is located in the eastern part of Fukushima Prefecture, have been playing a role of the transport of radionuclides deposited on the river watershed from land to ocean during rain and snow-melting. The coastal land and marine region in the Hamadori area is important for agricultural and fishery industries. Therefore, research on the behavior of radioactivity is needed for understanding the spatial distribution, migration, and effects of radioactivity on human health and ecosystems.

Prefectural governments have been monitoring the amount of radiation in coastal land and marine environments. However, a few research studies have been conducted on the behavior of radionuclides on sand beaches. Harada et al. reported spatial and vertical distribution of radioactivity and radiation loses in sand from beaches in Iwaki City, over tens of kilometers away from the FDNPP. Sanial et al. found the highest 137Cs radioactivity up to 23 Bq l⁻¹ in groundwater beneath sand beaches in Fukushima Prefecture, though the radiation dose in the ground surface was almost similar to that found in surrounding land area. The sand at deeper layers is considered the source of radioactivity in groundwater. Sanial et al. postulated that the sand on the beaches was contaminated in
2011 by wave- and tide-driven exchange and sorption of highly radioactive Cs from seawater.

Sorption and desorption experiments for $^{137}$Cs are important for understanding the existence forms of radiocesium released from the FDNPP accident and the migration behavior of radiocesium in terrestrial environments. The estuary environment is important for the desorption of Cs from soil, sediments and suspended solids by freshwater-seawater. Sakaguchi et al. showed that more than 15% of total $^{137}$Cs in the surface soil sample was desorbed by seawater leaching. Takata et al. reported that 0.75 to 6.6% of $^{137}$Cs were desorbed from the sieved particles ($<74 \mu m$) of the riverbank soils and river-bottom sediments from four rivers in Fukushima, Ibaraki and Chiba Prefecture by using filtered natural seawater. Harada et al. showed lower desorption of $^{137}$Cs from sand, which had adsorbed $^{137}$Cs in contaminated water, by using tap water. Sanial et al. carried out experiments for sand collected from two beaches by using seawater spiked $^{137}$Cs standard. The 1.4 - 11% of $^{137}$Cs adsorbed on the sand was removed from the sands packed with a column using seawater. Onodera et al. reported that the sorption capacity for radiocesium is affected by the sorption and desorption behavior of riverine suspended solids. Our group has reported lower desorption of $^{137}$Cs for the riverine suspended solids than those of $^{137}$Cs spiked to the suspended solids sample by artificial seawater. This study reports the results of direct desorption experiments for $^{134}$Cs and $^{137}$Cs from beach sand collected in 2016 at the same coastal beach, Yotsukura beach, which was used for groundwater analysis by Sanial et al. Desorption experiments for $^{134}$Cs and $^{137}$Cs by seawater were performed for four selected sand samples by batch experiments, and repeated at least 12 times to understand the degree of desorption by seawater and the weak and strong sorption of $^{134}$Cs and $^{137}$Cs. Various desorption reagents, such as ultrapure water, seawater, artificial seawater and NaCl solution, were also used for the desorption experiment to understand the desorption by freshwater-seawater, and the role of sodium and potassium ions in the desorption of $^{134}$Cs and $^{137}$Cs from the sand samples in land-coastal environments.

**Experimental**

**Sampling collection**

Yotsukura Beach is located in Iwaki City in Fukushima Prefecture, Japan (Fig. 1). The beach is located at a distance of 32 km from the FDNPP. Beach sand samples were collected vertically at 5 cm intervals at four sites (St.1, St.3 - 5) on November 14 and 16, 2016 (Fig. 1). Digital map data were based on the Fundamental Geospatial Data (Geospatial Information Authority of Japan) and National Land Numerical Information (Ministry of Land, Infrastructure, Transport and Tourism, Japan) databases. Groundwater was present beneath the deepest sand depth at each sampling site. Groundwater collection was conducted at St.2 and St.3. The groundwater results have been reported by Sanial et al. and exhibited higher $^{137}$Cs concentration with maximum of 23 Bq l$^{-1}$ at deeper layers below the ground surface and toward the end of the beach. At the initial sampling of groundwater, wet sand was collected from a depth of 42 - 46 cm at St.2 and a depth of 65 - 70 cm at St.3. The sand samples were dried in an oven at 40°C and then sieved with 2 mm mesh size sieve.

**Desorption experiment**

The desorption experiments were performed on four sand samples (St.1 75 - 80 cm, St.4 35 - 40 cm, St.5 0 - 5 cm, and St.5 100 - 105 cm) with $^{134}$Cs and $^{137}$Cs by using seawater in a
batch experimental system to obtain basic information about the desorption behavior of radio cesium in sand samples with different $^{134}$Cs and $^{137}$Cs concentrations. Sanial et al.\textsuperscript{12} used surface sand samples from two sites at Funatsuke and one site from Yotsukura beach. But this study used the sand samples collected from different sites and depth intervals. Approximately 30 g of dry sand was put into a 500-mL container with 300-mL seawater collected at Tsukumo Bay on the Noto Peninsula in April and August 2017. The salinity was 33.12 – 33.36 and the radioactivity levels of $^{134}$Cs and $^{137}$Cs were less than the detection limit and 1 – 2 mBq $\text{L}^{-1}$, respectively.\textsuperscript{20} The radioactivity level is negligible for the desorption experiment. The seawater sample was filtered with a 0.45-$\mu$m membrane filter before the experiment. The desorption experiments were carried out by using a rotation system at 60 rpm for two days, based on a modified method of Otsuka and Kobayashi.\textsuperscript{21} After the desorption experiment, the solution was centrifuged at 3000 rpm and then filtered with a 0.45-$\mu$m membrane filter. After drying each sand sample at 40°C in an oven, the experiment was carried out under the same experimental conditions 12 – 16 times. The recovered solutions containing the radio cesium desorbed from the sand samples were concentrated for the $^{134}$Cs and $^{137}$Cs by ammonium molybdatephosphate (AMP)/Cs compound.\textsuperscript{11} The percentage of radio cesium desorption was calculated as follows: Desorption(%) $= \left( \frac{C_{\text{solution}} \times V_{\text{solution volume}}}{C_{\text{solid}} \times W_{\text{solid weight}}} \times 100 \right)$; $C_{\text{solution}} =$ $^{134}$Cs and $^{137}$Cs radioactivity (Bq $L^{-1}$) extracted in the solution, $C_{\text{solid}} =$ $^{134}$Cs and $^{137}$Cs radioactivity (Bq $kg^{-1}$) in the sand before the desorption experiment.

The desorption experiments with various solutions (ultrapure water, 470 mM NaCl, 470 mM NaCl + 8 mM KCl, artificial seawater) were also carried out for sand samples with the highest radioactivity of $^{134}$Cs and $^{137}$Cs collected from 100 – 105 cm at St. 5. The concentration of 470 mM of Na\textsuperscript{+} and 8 mM of K\textsuperscript{+} are almost similar in seawater. The chemical composition of artificial seawater used in this study was as follows: NaCl 453 mM, MgSO\textsubscript{4}·7H\textsubscript{2}O 27.5 mM, MgCl\textsubscript{2}·6H\textsubscript{2}O 25.4 mM, CaCl\textsubscript{2}·2H\textsubscript{2}O 10.0 mM, KCl 9.76 mM, NaHCO\textsubscript{3} 2.38 mM, NaBr 0.628 mM, H\textsubscript{2}BO\textsubscript{3} 0.419 mM, SrCl\textsubscript{2}·6H\textsubscript{2}O 7.98 $\times$ 10$^{-2}$ mM, NaF 6.82 $\times$ 10$^{-3}$ mM, KI 4.76 $\times$ 10$^{-4}$ mM. The artificial seawater did not contain stable Cs so the comparison with the result from the extraction of seawater is useful for understanding the effects of stable Cs for the desorption of $^{134}$Cs and $^{137}$Cs. Sand to solution ratio was 1:10 in the unit of weight. The recovered solutions containing the radio cesium desorbed from the sand were filtered with a membrane filter (a pore size of 0.45$\mu$m) and then the $^{134}$Cs and $^{137}$Cs were concentrated by ammonium molybdatephosphate (AMP)/Cs compound.\textsuperscript{11} The percentage of radio cesium desorption was calculated as follows: Desorption(%) $= \left( \frac{C_{\text{solution}} \times V_{\text{solution volume}}}{C_{\text{solid}} \times W_{\text{solid weight}}} \times 100 \right)$; $C_{\text{solution}} =$ $^{134}$Cs and $^{137}$Cs radioactivity (Bq $L^{-1}$) extracted in the solution, $C_{\text{solid}} =$ $^{134}$Cs and $^{137}$Cs radioactivity (Bq $kg^{-1}$) in the sand before the desorption experiment.

The grain size fraction was prepared by sieving with a 63-$\mu$m mesh size sieve, corresponding to the difference in grain size between clay-silt and sand fraction. After drying and determination of weight and radioactivity of $^{134}$Cs and $^{137}$Cs, the sand fraction was performed for the radio cesium desorption experiment under the same conditions. The percentage of desorption was calculated by the above method.

Analysis

The soil sample was packed into a container. Gamma emission peaks were used for calculating activity at 605 keV for $^{134}$Cs and 661 keV for $^{137}$Cs. The radioactivity of $^{134}$Cs and $^{137}$Cs in the desorption solution was measured for the AMS/Cs compound using gamma-ray spectrometry. The compound was dried at room temperature, weighed to determine the weight yield (>94%), and packed into a polyethylene bag (3.5 $\times$ 7.0 cm). The $^{134}$Cs and $^{137}$Cs desorbed from the sand samples were measured by gamma-ray spectrometry using a low BKG Ge semiconductor detector (EGPC250-P21, CAMBERRA, France) at the Low Level Radioactivity Laboratory and the Ogoya Underground Laboratory, Kanazawa University.\textsuperscript{22} The cascade summing effect was corrected for $^{134}$Cs using a contaminated soil sample from Fukushima. Decay correction for radioactivity for $^{134}$Cs and $^{137}$Cs was performed on each sampling date. X-ray diffraction patterns of silt + clay fraction of beach sand from St. 4 (35 – 40 cm depth) and St. 5 (0 – 5 cm depth) were analyzed from 2 to 65° 2θ every 0.02 2θ on a Rigaku D8 Ultima IV diffractometer with Cu Kα radiation.

Results and Discussion

Vertical distribution of $^{134}$Cs and $^{137}$Cs radioactivity

Figure 2 shows the vertical distribution of $^{134}$Cs and $^{137}$Cs radioactivity of the beach sand collected at five sampling sites.\textsuperscript{12} Cs radioactivity was previously reported by Sanial et al.\textsuperscript{12} The radioactivity data are given in Table S1. The results are plotted as a function of depth and $^{134}$Cs and $^{137}$Cs radioactivity. At St.1, radio cesium concentration is almost constant with depth from 0 to 60 cm, but below at a depth of 60 cm, the concentration increases with depth. The radioactivity of sand samples from St.3 is almost constant at depths of 0 – 30 cm. At St.4 radio cesium concentration is almost constant at depths from 0 to 25 cm and then increases at deeper layers. The $^{134}$Cs profile for sand samples at St.5 increases with depth from 4.6 Bq $kg^{-1}$ at a depth of 65 – 70 cm to 167 Bq $kg^{-1}$ at depths of 100 – 105 cm. The $^{137}$Cs profile is almost similar in variation and $^{134}$Cs/$^{137}$Cs activity ratio decay corrected to March 11, 2011 ranges from 0.84 to 1.07 (average of 0.98 ± 0.06). This indicates a major part of radio cesium is derived from the FDNPP accident.\textsuperscript{1} The radioactivity of $^{134}$Cs and $^{137}$Cs in the sand collected from a depth of 42 – 46 cm at St.2 is 2.8 and 17.6 Bq $kg^{-1}$, respectively. The radioactivity of sand collected at a depth of 67 – 71 cm at St.3 is 2.7 Bq $kg^{-1}$ for $^{134}$Cs and 17.5 Bq $kg^{-1}$ for $^{137}$Cs. The highest radioactivity of $^{134}$Cs and $^{137}$Cs was observed at deeper sand layers just above groundwater at a depth of 100 – 105 cm at St.5. Water content of the sand samples ranged from 21.5 to 25.0%, and is almost constant with depth. Higher radiation doses were observed for sand below 60 cm in depth from the ground surface at two study sites on Yotsukura Beach in January 2012.\textsuperscript{11} Therefore, the levels of radio cesium radioactivity remained high at deeper layers of beach sand in January 2012.

Desorption of radio cesium from sand by seawater sequential extraction

To understand the desorption behavior of radio cesium in the sand, the desorption experiments were performed by seawater sequentially for the four sand samples with $^{137}$Cs radioactivity from 16 to 1077 Bq $kg^{-1}$. The results are shown in Fig. 3. The percentage of $^{137}$Cs desorption decreases exponentially with increasing number of desorption experiments. A 0% $^{134}$Cs desorption indicates less than the detection limit of its radioactivity. Total desorption percentage ranged from 19 to 58% in 12 desorption experiments (Fig. 3a) 58%, (b) 19%, (c) 37%, and (d) 51%) and the values exhibited the maximum desorption for each sand sample. The $^{137}$Cs radioactivity before the desorption experiments is in the order of Fig. 3(c) < (b) < (a) < (d) and the order is not in agreement with the order of total
The total percentage of $^{137}$Cs from the sand collected at 0 – 5 cm depth is lower than that of the deeper layer (100 – 105 cm depth) at St. 5. The deeper layers at St.1 and St.5 correspond to the close to groundwater level and show higher desorption percentages of 51 – 58%. Therefore, the sand is considered possible sources of higher radioactivity of $^{137}$Cs in groundwater beneath the sand beach, which was reported by Sanial et al.\textsuperscript{12} The results also indicate that the strong and weak sorption sites for $^{137}$Cs are present in the Yotsukura beach sand. The differences in desorption percentage among the sand samples is possibly related to the difference in clay mineral content and/or the effects of weathering of clay minerals in the presence of groundwater at the deeper layers.

Radioesium desorption by ultrapure water, NaCl solution and seawater

Figure 4 shows the results of radioesium desorption from the sand by ultrapure water, 1/2 seawater (diluted seawater with same volume of ultrapure water) and seawater. The desorption percentage of $^{137}$Cs by seawater increases in the order of St.4...
35 - 40 cm (3.1%) < St.5 0 - 5 cm (7.5%) < St.5 100 - 105 cm (11.8%) < St.5 75 - 80 cm (14.9%). The desorption values are slightly higher than those (0.75 - 6.6%) of the sieved particles from the river bank soil samples and river-bottom sediments by seawater.18 The desorption percentage of $^{137}$Cs by 1/2 seawater increases in the order of St.4 35 - 40 cm (1.6%) < St.5 0 - 5 cm (4.4%) < St.5 100 - 105 cm (7.1%) < St.5 75 - 80 cm (9.3%). The desorption by ultrapure water ranges from 0.11 to 0.39% and is lower. The desorption depends on the salinity of seawater, that is, major cation concentration. The artificial seawater does not contain stable Cs so the effect of stable Cs in seawater on the desorption is small.

Ammonium and potassium ions have been used as desorption reagents of radioesium for soils and sediments.21 To understand the effects of sodium and potassium ions on the desorption, radioesium desorption experiments were carried out for the sand collected at 100 - 105 cm depth at St.5 using various solutions. The results are shown in Table 1. The $^{137}$Cs desorption from deeper sand with 1052 ± 25 Bq kg$^{-1}$ (1021 - 1086 Bq kg$^{-1}$) is 0.1% by ultrapure water, 3.7% by 1/4 seawater and 7.1% by 1/2 seawater, and 12% by seawater. The artificial seawater and 470 mM NaCl + 8 mM KCl solution seawater and 7.1% by 1/2 seawater, and 12% by seawater. The desorption of radiocesium derived from the FDNPP accident in coastal beach sand in the presence of seawater.

### Factors controlling desorption behavior of radioesium by seawater

The sand (63 μm<) and silt + clay grain size (<63 μm) were separated for three sand samples with $^{137}$Cs radioactivity of 17 – 33 Bq kg$^{-1}$ by using a wet sieving method. Figure 5 shows the weight percentage of each grain size fraction, $^{137}$Cs radioactivity and its percentage to total radioactivity of a bulk sample. The weight percentage of silt and clay fraction ranged from 1.8 to 4.0%. The sand size fraction occupied >95% of the bulk sand sample. However, $^{137}$Cs radioactivity is 391 - 599 Bq kg$^{-1}$ for the silt-clay fraction and 8.1 - 9.5 Bq kg$^{-1}$ for the sand fraction. The radioactivity ratio of $^{134}$Cs/$^{137}$Cs decay corrected to the Fukushima accident on March 11, 2011 ranged from 0.80 to 1.11 and an average of 0.97 ± 0.11. The percentage of $^{137}$Cs in the silt-clay fraction is 41 - 74% of the total radioactivity. The higher percentage of $^{137}$Cs in a smaller size fraction of sand, soil and sediment has been reported.14,28-30

The desorption experiments using seawater were carried out for the sand samples >63 μm grain size separated by wet sieve method. The $^{137}$Cs desorption was 3.1 - 7.5% for the bulk sand and 8.5 - 9.5% for larger grain size fraction of the sand. The desorption experiment for the silt + clay fraction did not perform due to the low weight of the samples. But we estimated the contribution of desorption from the silt-clay fraction. The difference in $^{137}$Cs radioactivity in solution from bulk sand and the sand grain size fraction is considered as desorption from the silt-clay fraction. The solution radioactivity was divided by the

### Table 1 Percentage of $^{134}$Cs and $^{137}$Cs desorption from beach sand samples by using various solutions

| Desorption solution | Sand $^{137}$Cs Bq kg$^{-1}$ | Extracted solution $^{137}$Cs Bq | $^{137}$Cs % | Sand $^{137}$Cs Bq kg$^{-1}$ | Extracted solution $^{137}$Cs Bq | $^{137}$Cs % |
|---------------------|-------------------------------|---------------------------------|--------------|-------------------------------|---------------------------------|--------------|
| Seawater            | 1077 ± 21                     | 3.81 ± 0.06                     | 11.8 ± 0.3   | 176 ± 9                       | 0.62 ± 0.04                     | 11.7 ± 0.9   |
| Seawater/1/2        | 1066 ± 13                     | 2.28 ± 0.01                     | 7.1 ± 0.1    | 173 ± 6                       | 0.37 ± 0.01                     | 7.1 ± 0.3    |
| Seawater/1/4        | 1038 ± 4                      | 1.15 ± 0.02                     | 3.7 ± 0.1    | 178 ± 2                       | 0.21 ± 0.02                     | 3.8 ± 0.3    |
| Ultrapure water     | 1037 ± 17                     | 0.04 ± 0.00001                 | 0.1 ± 0.004  | 178 ± 8                       | 0.004 ± 0.001                   | 0.1 ± 0.02   |
| 470 mM NaCl         | 1021 ± 11                     | 0.68 ± 0.01                     | 2.2 ± 0.01   | 184 ± 6                       | 0.11 ± 0.01                     | 2.0 ± 0.2    |
| 470 mM NaCl + 8 mM KCl | 1036 ± 11                 | 3.19 ± 0.04                     | 10.2 ± 0.04  | 168 ± 5                       | 0.52 ± 0.03                     | 10.4 ± 0.7   |
| Artificial seawater | 1086 ± 11                     | 3.98 ± 0.06                     | 12.2 ± 0.2   | 193 ± 6                       | 0.59 ± 0.05                     | 10.1 ± 0.9   |

The error of radioactivity is counting error.
radioactivity of sand samples less than 63 μm and then denoted as the percentage (Table S2). The desorption percentage for the silt + clay fraction is 0.4 – 4.4%, and is almost similar with bulk sand, and sand grain size fractions, though the solid-solution ratio (250 – 570) of silt-clay fraction is one order of magnitude higher than that of the bulk and sand size fraction. The higher solid-solution ratio of the desorption experiment indicates a higher amount of 137Cs released from the suspended materials. Clay mineral composition was analyzed for the silt-clay fraction by X-ray diffraction method. As shown in Fig. 6, 7 and 10 Å clay minerals were detected for the silt-clay fractions and are possibly kaolinite, illite, and montmorillonite. The selective sorption of 137Cs to the clay minerals have the ability for the sorption of radiocesium.

Onodera et al. showed higher desorption of 137Cs from the riverine suspended solids with medium grain size of ca. 50 μm using 470 mM and 1 M NaCl solution after the 137Cs adsorption experiment. The 137Cs radioactivity after the adsorption experiment was ca. 2.64 × 10^8 Bq kg⁻¹ and was three to four orders of magnitude higher than that of sand samples in this study. Our group performed the desorption experiment for the riverine suspended solids (137Cs radioactivity of 2390 Bq kg⁻¹) concentrated from the Abukuma River water by continuous flow centrifugation. The desorption percentage of 137Cs was 0.13% by artificial seawater. Onodera et al. showed that after the sorption experiments, the desorption percentage from the Abukuma River suspended solids (137Cs radioactivity of 2445 Bq kg⁻¹ before the sorption) was about 65% by 470 mM NaCl and about 72% by 1 M NaCl solution at a solid to solution ratio of 0.17 g l⁻¹. They also have reported the strong and weaker sorption sites, and negative relationship in distribution coefficient for 137Cs with equilibrium concentration of cesium ion. The strong adsorption sites are occupied first followed by the weaker affinity sites, when the cesium concentration in the bulk solution is increased. The riverine suspended solids consist mainly of silt + clay size fraction but the sand samples used in this study, silt + clay size fraction is 1.8 – 4.0%. The strong sorption sites are lower than those of the riverine suspended solids. Sanial et al. showed the desorption experimental results for the beach sand by ultrapure water and seawater with salinity of 4 to 30 after the 137Cs adsorption. The 137Cs radioactivity is estimated as 1785 Bq kg⁻¹ and no more than twice its radioactivity of sand samples collected from the beach. The desorption fractions ranged from 1.4 to 11.4% and were almost similar with our batch desorption experiment using 1/2, 1/4 and natural seawater (1.6 – 14.9% in Table 1 and Fig. 4). Therefore, the clay mineral content, that is, the capacity of clay minerals with strong and weaker sorption sites to radiocesium adsorption is related to the desorption behavior of 137Cs.

Conclusions

The desorption experiments were carried out for beach sand samples collected from four sites on Yotsukura Beach in Fukushima Prefecture using seawater, artificial seawater and NaCl solution. The radioactivity of 134Cs and 137Cs in the sand samples ranged from 16 to 1077 Bq kg⁻¹. The continuous seawater desorption experiments showed an exponential decrease with an increasing number of experiments. The cumulative desorption percentage was 58% for the sand (75 – 80 cm depth) at St.1, 19% for the sand (35 – 40 cm depth) at St.4, 37% for the sand (0 – 5 cm depth) at St.5 and 51% for the sand (100 – 105 cm depth) at St.5. The higher desorption was observed for sands from deeper layers. Additional desorption experiments were also carried out for sand collected from deeper layers with 1052 ± 25 Bq kg⁻¹. The desorption was 0.1% by ultrapure water, 3.7% by 1/4 seawater and 7.1% by 1/2 seawater, 2.2% by 470 mM NaCl solution (corresponding to Na⁺ concentration in seawater) and 10 – 12% by seawater, artificial seawater and 470 mM NaCl + 8 mM KCl solution. The difference in the desorption was observed for the leaching with 470 mM NaCl solution in the presence and absence of potassium ion. It was surmised that a major part of 137Cs released from the FDNPP accident strongly adsorbs on the silt + clay minerals.
clay fraction of sand, based on grain size separation with 63 μm mesh size sieve. The results indicate that the presence of potassium ion increases the desorption of radiocesium from the sand with larger grain size (sand size) fraction.

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Supporting Information

Supplementary material contains supporting data of the radiocesium concentration in sand samples and the desorption results of sieved sand samples. This material is available free of charge on the Web at http://www.jsac.or.jp/analsci/

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