

Desorption Behavior of Fukushima-derived Radiocesium in Sand Collected from Yotsukura Beach in Fukushima Prefecture

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Beach sand samples were collected along a coastal area 32 km south of the Fukushima Daiichi Nuclear Power Plant (FDNPP) in Fukushima Prefecture, Japan, 5 years after the FDNPP accident. Desorption experiments were performed on the sand samples using seawater in a batch experimental system to understand the forms of existence of radiocesium in sand and their desorption behavior in a coastal environment. The percentage of radiocesium desorption decreased exponentially with an increase in the number of desorption experiments for the four sand samples, with ¹³⁷Cs radioactivity from 16 to 1077 Bq kg⁻¹ at surface and deeper layers from three sites. Total desorption percentage ranged from 19 to 58% in 12 desorption experiments. The results indicate that the weak adsorption varies with the sampling sites and their depth layer. To understand the desorption behavior of radiocesium in the sand samples, the desorption experiments were performed for a sand sample by using natural and artificial seawater, and NaCl solution in the presence and absence of KCl. The ¹³⁷Cs desorption from the sand collected at a depth of 100 – 105 cm from the ground surface (¹³⁷Cs radioactivity 1052 ± 25 Bq kg⁻¹) 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 a similar concentration of seawater) and 10 – 12% by seawater, artificial seawater and 470 mM NaCl + 8 mM KCl solution. These results indicate that about 10% of radiocesium adsorbed on the sand is mainly desorbed by ion exchange of potassium ion in seawater, though the concentration of major cation, or sodium ion, in seawater makes a small contribution on ¹³⁷Cs desorption from the sand samples.

Keywords Cs, beach sand, ion exchange, potassium ion, groundwater, Fukushima Daiichi Nuclear Power Plant accident

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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.¹ The radionuclides with short half-life, such as a few seconds or minutes, decayed after a few months after the accident.^{2,3} 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, 2011.¹ The river systems running through the Hamadori area, which is located in the eastern part of

Fukushima Prefecture, have been playing a role of the transport of radionuclide deposited on the river watershed from land to ocean during rain and snow-melting.⁴⁻⁶ The coastal land and marine region in the Hamadori area is important for agricultural and fishery industries.⁷ Therefore, research on the behavior of radiocesium is needed for understanding the spatial distribution, migration, and effects of radiocesium on human health and ecosystems.

Prefectural governments have been monitoring the amount of radiation in coastal land and marine environments.⁸⁻¹⁰ However, a few research studies have been conducted on the behavior of radionuclides on sand beaches. Harada *et al.*¹¹ reported spatial and vertical distribution of radiocesium and radiation loses in sand from beaches in Iwaki City, over tens of kilometers away from the FDNPP. Sanial *et al.*¹² 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 radiocesium in groundwater. Sanial *et al.*¹² postulated that the sand on the beaches was contaminated in

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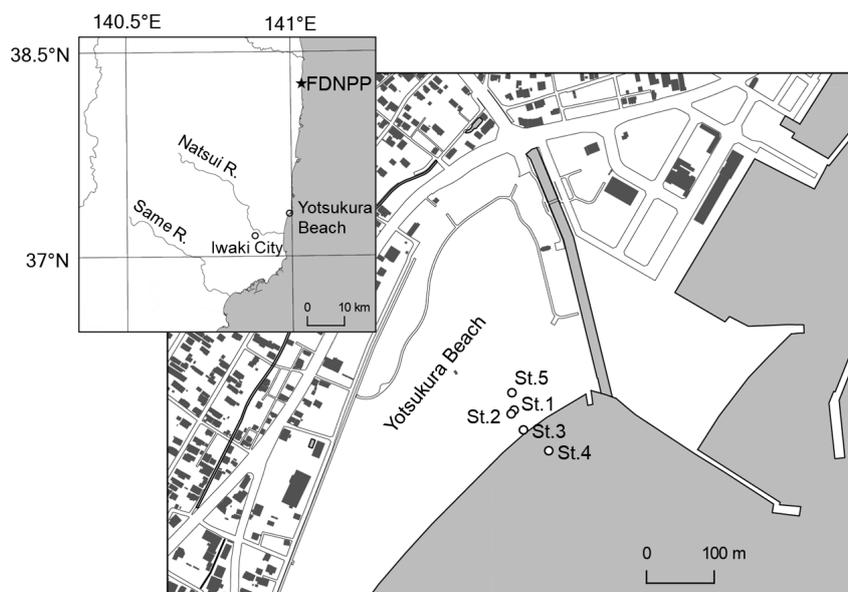


Fig. 1 Sampling location of this study. This map is modified using GIS data from the Fundamental Geospatial Data (Geospatial Information Authority of Japan) and National Land Numerical Information (Ministry of Land, Infrastructure, Transport and Tourism, Japan) databases. As the shoreline is based on the high tide condition under the topographical map, St.4 is indicated at a point off the shoreline. The sand core sample at St.4 was collected during low tide conditions.

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.¹³⁻¹⁶ The estuary environment is important for the desorption of Cs from soil, sediments and suspended solids by freshwater-seawater. Sakaguchi *et al.*¹⁷ showed that more than 15% of total ^{137}Cs in the surface soil sample was desorbed by seawater leaching. Takata *et al.*¹⁸ reported that 0.75 to 6.6% of ^{137}Cs were desorbed from the sieved particles (<74 μm) 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.*¹¹ showed lower desorption of ^{137}Cs from sand, which had adsorbed ^{137}Cs in contaminated water, by using tap water. Sanial *et al.*¹² 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.*¹⁶ 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.¹⁹ The discrepancy in the desorption of ^{137}Cs was observed for the riverine suspended solids collected from river waters and after the sorption experiments. Direct desorption experiment without adsorption operation for ^{134}Cs and ^{137}Cs is needed to understand the presence of radiocesium beneath sand beaches.

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.*¹² 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, 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.*¹² 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 radiocesium in sand samples with different ^{134}Cs and ^{137}Cs concentrations. Sanial *et al.*¹² used surface sand samples from two sites at Funatsuke and one site from Yotuskura 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 of 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 l⁻¹, respectively.²⁰ The radioactivity level is negligible for the desorption experiment. The seawater sample was filtered with a 0.45- μm 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.²¹ After the desorption experiment, the solution was centrifuged at 3000 rpm and then filtered with a 0.45- μm 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 radiocesium desorbed from the sand samples were concentrated for the ^{134}Cs and ^{137}Cs by ammonium molybdophosphate (AMP)/Cs compound.¹¹ The percentage of radiocesium desorption was calculated as follows: $\text{Desorption (\%)} = (C_{\text{solution}} \times V_{\text{solution volume}}) / (C_{\text{sand}} \times W_{\text{sand weight}}) \times 100$; $C_{\text{solution}} = ^{134}\text{Cs}$ and ^{137}Cs radioactivity (Bq l⁻¹) extracted in the solution, $C_{\text{sand}} = ^{134}\text{Cs}$ 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, 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⁺ and 8 mM of K⁺ are almost similar in seawater. The chemical composition of artificial seawater used in this study was as follows: NaCl 453 mM, MgSO₄·7H₂O 27.5 mM, MgCl₂·6H₂O 25.4 mM, CaCl₂·2H₂O 10.0 mM, KCl 9.76 mM, NaHCO₃ 2.38 mM, NaBr 0.628 mM, H₃BO₃ 0.419 mM, SrCl₂·6H₂O 7.98 × 10⁻² mM, NaF 6.82 × 10⁻² mM, KI 4.76 × 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 radiocesium desorbed from the sand were filtered with a membrane filter (a pore size of 0.45 μm) and then the ^{134}Cs and ^{137}Cs were concentrated by ammonium molybdophosphate (AMP)/Cs compound.¹¹ The percentage of radiocesium desorption was calculated as follows: $\text{Desorption (\%)} = (C_{\text{solution}} \times V_{\text{solution volume}}) / (C_{\text{sand}} \times W_{\text{sand weight}}) \times 100$; $C_{\text{solution}} = ^{134}\text{Cs}$ and ^{137}Cs radioactivity (Bq l⁻¹) extracted in the solution, $C_{\text{sand}} = ^{134}\text{Cs}$ 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- μm 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 radiocesium 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 × 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, CAMBERA, France) at the Low Level Radioactivity Laboratory and the Ogoya Underground Laboratory, Kanazawa University.²² The cascade summing effect was corrected for ^{134}Cs using a contaminated soil sample from Fukushima. Decay correction of 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° 2 θ every 0.02 2 θ on a Rigaku D8 Ultima IV diffractometer with Cu K α 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.¹³⁷ Cs radioactivity was previously reported by Sanial *et al.*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, radiocesium 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 radiocesium 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 $^{134}\text{Cs}/^{137}\text{Cs}$ 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 radiocesium is derived from the FDNPP accident.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 2012.¹¹ Therefore, the levels of radiocesium radioactivity remained high at deeper layers of beach sand in January 2012.

Desorption of radiocesium from sand by seawater sequential extraction

To understand the desorption behavior of radiocesium 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. 3(a) 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

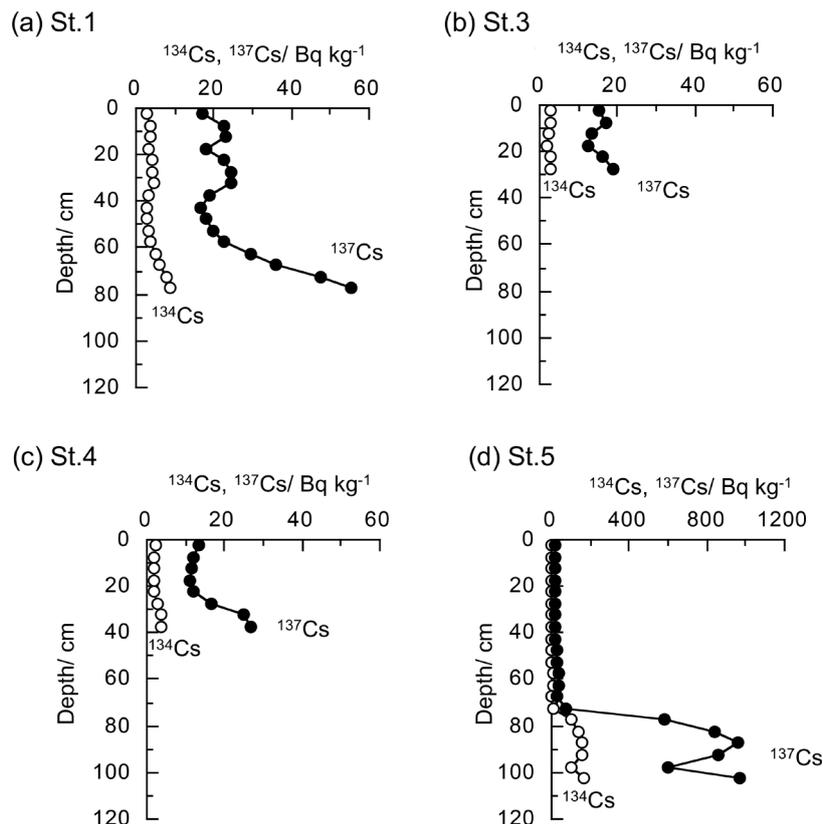


Fig. 2 Vertical distribution of ^{134}Cs (open circles) and ^{137}Cs (closed circles) radioactivity of sand samples from four sites on Yotsukura Beach.

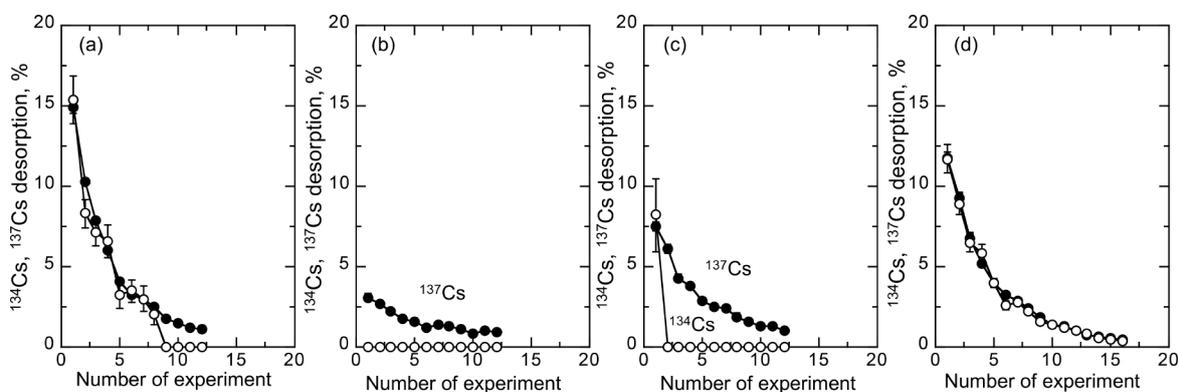


Fig. 3 Percentage of desorption ^{134}Cs (open circles) and ^{137}Cs (closed circles) from sand samples collected from the surface and deeper layers with various radioactivity determined by using seawater: (a) St.1 75 – 80 cm depth, (b) St.4 35 – 40 cm depth, (c) St.5 0 – 5 cm depth, (d) St.5 100 – 105 cm depth. The depth indicates the distance from the ground surface.

desorption percentage. 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.*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.

Radiocesium desorption by ultrapure water, NaCl solution and seawater

Figure 4 shows the results of radiocesium 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 ¹³⁷Cs 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 radiocesium for soils and sediments.²³ To understand the effects of sodium and potassium ions on the desorption, radiocesium 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 ¹³⁷Cs desorption from deeper sand with $1052 \pm 25 \text{ Bq kg}^{-1}$ ($1021 - 1086 \text{ Bq kg}^{-1}$) 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 show 10 – 12% of radiocesium desorption. There is a positive correlation between the desorption percentage and salinity for desorption solution of ultrapure water, 1/4 seawater, 1/2 seawater

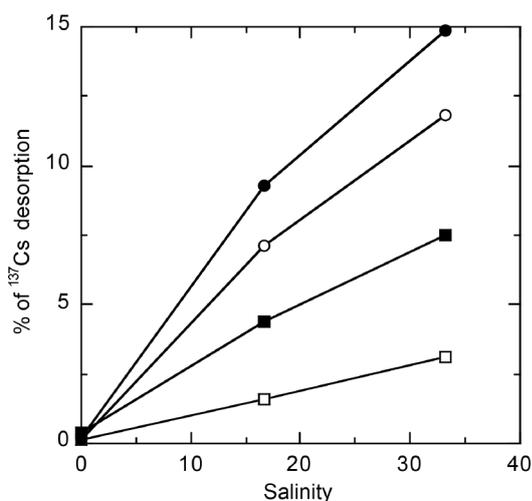


Fig. 4 Percentage of ¹³⁷Cs desorption from sand samples (closed circles: St.1 75 – 80 cm depth, open square: St.4 35 – 40 cm depth, closed square: St.5 0 – 5 cm depth, open circles: St.5 100 – 105 cm depth) by using 1/2 diluted seawater, seawater and ultrapure water.

and original seawater (correlation factor of 0.99). On the other hand, 470 mM NaCl solution is 2.2% of radiocesium desorption and is about 10 points lower than that of 470 mM NaCl + 8 mM KCl solution. Therefore, ionic strength and major cation, sodium ion in these solutions, are not major desorption reagents for ¹³⁴Cs and ¹³⁷Cs from the sand. The percentage of ¹³⁷Cs desorption is plotted as a function of potassium concentration. The seawater potassium concentration is referred from Nozaki.²⁴ There is a positive correlation, correlation factor of 0.99, between the ¹³⁷Cs desorption percentage and KCl concentration in various reagents. A similar result was also obtained for ¹³⁴Cs desorption (correlation factor of 0.96). The sorption of ¹³⁴Cs and ¹³⁷Cs onto clay minerals (illite, muscovite, weathered biotite, and vermiculite) has been reported for soils from the Fukushima area.^{15,25,26} Fukushi *et al.*²⁷ showed that desorption of Cs in smectite increased with potassium ion concentrations of less than 20 mM. These results indicate that radiocesium associated with the sand is replaced by potassium ion, that is, an ion-exchange reaction, in the concentration range of 2.5 to 10 mM. The potassium concentration is an important factor for the desorption of radiocesium derived from the FDNPP accident in coastal beach sand in the presence of seawater.

Factors controlling desorption behavior of radiocesium by seawater

The sand (63 μm<) and silt + clay grain size (<63 μm) were separated for three sand samples with ¹³⁷Cs radioactivity of 17 – 33 Bq kg⁻¹ by using a wet sieving method. Figure 5 shows the weight percentage of each grain size fraction, ¹³⁷Cs 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, ¹³⁷Cs 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 ¹³⁴Cs/¹³⁷Cs 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 ¹³⁷Cs in the silt-clay fraction is 41 – 74% of the total radioactivity. The higher percentage of ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁷Cs 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 ¹³⁴Cs and ¹³⁷Cs desorption from beach sand samples by using various solutions

Desorption solution	Sand ¹³⁷ Cs/ Bq kg ⁻¹	Extracted solution ¹³⁷ Cs/Bq	Desorption ¹³⁷ Cs, %	Sand ¹³⁴ Cs/ Bq kg ⁻¹	Extracted solution ¹³⁴ Cs/Bq	Desorption ¹³⁴ 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	1068 ± 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 ± 0.001	0.1 ± 0.004	178 ± 8	0.004 ± 0.001	0.1 ± 0.02
470 mM NaClaq	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 KClaq	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.

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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