



# Desorption of radioactive cesium by seawater from the suspended particles in river water



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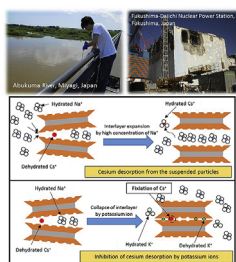
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## HIGHLIGHTS

- The presence of Na<sup>+</sup> and K<sup>+</sup> ions affects the cesium desorption behavior from suspended particles in river water.
- The amount of loaded cesium strongly correlates with cesium desorption from the suspended particles.
- Suspended particles have heterogeneous adsorption sites for cesium with a wide range of affinities.

## GRAPHICAL ABSTRACT



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## ABSTRACT

In 2011, the accident at the Fukushima-Daiichi nuclear power plant dispersed radioactive cesium throughout the environment, contaminating the land, rivers, and sea. Suspended particles containing clay minerals are the transportation medium for radioactive cesium from rivers to the ocean because cesium is strongly adsorbed between the layers of clay minerals, forming inner sphere complexes. In this study, the adsorption and desorption behaviors of radioactive cesium from suspended clay particles in river water have been investigated. The radioactive cesium adsorption and desorption experiments were performed with two kinds of suspended particulate using a batch method with <sup>137</sup>Cs tracers. In the cesium adsorption treatment performed before the desorption experiments, simulated river water having a total cesium concentration ( $[^{133+137}\text{Cs}^+]_{\text{total}}$ ) of 1.3 nM ( $10^{-9}$  mol/L) was used. The desorption experiments were mainly conducted at a solid-to-liquid ratio of 0.17 g/L. The desorption agents were natural seawater collected at 10 km north of the Fukushima-Daiichi nuclear power plant, artificial seawater, solutions of NaCl, KCl, NH<sub>4</sub>Cl, and <sup>133</sup>CsCl, and ultrapure water. The desorption behavior, which depends on the preloaded cesium concentration in the suspended particles, was also investigated. Based on the cesium desorption experiments using suspended particles, which contained about 1000 ng/g loaded cesium, the order of cesium desorption ratios for each desorption agent was determined as 1 M NaCl (80%) > 470 mM NaCl (65%) > 1 M KCl (30%) ≈ seawater (natural seawater and Daigo artificial seawater) > 1 M NH<sub>4</sub>Cl (20%) > 1 M <sup>133</sup>CsCl (15%) ≫ ultrapure water (2%). Moreover, an interesting result was obtained: The desorption ratio in the 470 mM NaCl solution was much higher than that in seawater, even though the Na<sup>+</sup> concentrations were identical. These results indicate that the cesium desorption mechanism is not a simple ion exchange reaction but is strongly related to structural changes in the clay minerals in the suspended particles. Hydrated Na<sup>+</sup> ions expand the interlayer distance of the clay minerals, resulting in the facile desorption of cesium; in contrast, dehydrated K<sup>+</sup> ions reduce the

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interlayer distance and inhibit the desorption of cesium. In conclusion, the desorption of cesium from the suspended particles is controlled by the presence of sodium and potassium ions and the preloaded cesium concentration in the suspended particles.

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## 1. Introduction

In 2011, radioactive cesium was dispersed throughout the environment following the Fukushima-Daiichi Nuclear Power Plant (1F NPP) accident, causing contamination of the land surface and ocean. The total atmospheric release of radioactive cesium ( $^{134+137}\text{Cs}$ ) has been estimated to be 20 PBq (from March 12 to 31, 2011) by TEPCO (TEPCO, 2012), which is 1.4% of the total radioactive cesium inventory (~1420 PBq) of units 1, 2, and 3 of the Fukushima Daiichi Nuclear Power Plant before the accident (Nishihara et al., 2012). Morino et al. has estimated that 80% of the cesium released into the atmosphere was deposited in the Pacific Ocean, while the remaining 20% was deposited on land (Morino et al., 2013). Although rivers occupy only 3% of the land surface of Fukushima Prefecture, their presence allows the transportation of radioactive materials from the land surface to the ocean. Yamashiki et al. reported that, between August 8, 2011 and May 11, 2012, 9.2 TBq of radioactive cesium flowed into the ocean from the estuary of the Abukuma River, which flows from the southern area of Fukushima prefecture to the southern area of Miyagi prefecture (Yamashiki et al., 2014). This migration of radioactive cesium is considered to be strongly related to the chemical interactions between the suspended particles and the radioactive cesium in river water, primarily because the abundance of particulate cesium is greater than that of ionic cesium species in the Abukuma River (Sakaguchi et al., 2015). Because the suspended particles contain clay minerals, cesium is strongly adsorbed on the suspended particles. For example, 2:1 phyllosilicates, especially illite, adsorb cesium strongly in the interlayer by forming inner sphere complexes (Qin et al., 2012) (Fan et al., 2014). In addition, Nagao et al. reported that the ratio of particulate radioactive cesium in rivers increases with increasing water turbidity (Nagao et al., 2013). Two interpretations of their observation are possible: First, the erosion of riverside soil containing radioactive cesium increases the cesium content of the river with increasing flow rate. Second, the concentration of suspended particles increases because of the disturbance of the river sediment by the high flow rate, resulting in an increase in the cesium adsorption capacity. In contrast, the abundance of free ionic cesium in the Dnieper River in Ukraine after the Chernobyl accident was greater than that of particle-adsorbed cesium (Sansone et al., 1996). In the Pripyat River that is a tributary of the Dnieper River, the surfaces of the clay particles were covered with natural organic matter, inhibiting the adsorption of cesium in the interlayer of the clay minerals (Suga et al., 2014). In summary, the transport of radioactive contaminants from river to ocean is complicated. Furthermore, there is the possibility that part of the adsorbed radioactive cesium on the suspended particles could be desorbed as ionic species on entering the sea, where the solution conditions are quite different from those of river water. The most dominant cations in Japanese river water are  $\text{Na}^+$  and  $\text{Ca}^{2+}$  with concentrations of 0.22 and 0.25 mM, respectively (Uchida et al., 2007), while the average  $\text{Cs}^+$  concentration of Japanese river water is about 0.3 nM. Here, “mM” and “nM” denote  $10^{-3}$  mol/L and  $10^{-9}$  mol/L, respectively. In contrast,  $\text{Na}^+$ ,  $\text{Mg}^{2+}$ , and  $\text{Ca}^{2+}$  are the dominant cations in seawater with concentrations of 470, 53, and 10 mM, respectively (Nozaki, 2001), while the  $\text{Cs}^+$  concentration in

seawater is 2.3 nM. The concentrations of the dominant cations change drastically between river water and seawater, whereas the change in  $\text{Cs}^+$  concentration is less than ten times, which is still very low even in seawater. This ion composition difference between river water and seawater could affect the desorption behavior of radioactive cesium adsorbed on the suspended river water particles. If some fraction of the radioactive cesium is desorbed from the suspended particles becoming free ionic species in the brackish zone, the behavior of radioactive cesium in ocean would be changed drastically because of the different chemical properties of particle-adsorbed cesium and free ionic cesium. In fact, Kakehi et al. reported that the ratio of ionic cesium increased with increasing salinity in the brackish area of estuaries (Kakehi et al., 2016). In contrast, the Nuclear Regulation Authority of Japan has reported that cesium adsorbed on the suspended particles of the Abukuma and Natsui Rivers was not desorbed on contact with artificial seawater (NRA, Japan, 2016). Thus, the understanding of the chemistry between the suspended particles of river water and radioactive cesium in seawater condition is still poor. To obtain accurate, long-term predictions of the distribution of radioactive cesium in rivers and oceans, we investigated the desorption behavior of radioactive cesium from suspended particles in the rivers of Fukushima Prefecture. As a possible basic scenario, we considered the adsorption of radioactive cesium on suspended particles occurred in river water, and designed a cesium pre-loading treatment for the suspended particles in simulated river water. Furthermore, as an optional scenario, it can be imagined that radioactive cesium was directly adsorbed on the clay particles in the ground via cesium aerosol released from 1F NPP, followed by the inflow of these clay particles into the river via erosion. The difference in the adsorption scenarios may affect the desorption behavior of cesium from the particles. Therefore, direct cesium adsorption on the suspended particles by cesium aerosol exposure was also attempted as an alternative cesium pre-loading treatment.

## 2. Materials and methods

### 2.1. Collection of the suspended particles from river water

Two kinds of natural particle suspensions were collected from two sites in the Fukushima area. The first sample was from the Abukuma River in Date-shi, about 60 km northwest of 1F NPP (Yanagawa-Ohashi, Yanagawa-cho, Date-shi, Fukushima, Japan), and is denoted as “Date Sample.” At the sampling time, the suspended particle concentration in the river water was 0.91 g/kg. The second sample was from the Natsui River in Iwaki-shi, located about 40 km south of 1F NPP (Iwaki-bashi, 24, Taira Shimohirakubo Magata, Iwaki-shi, Fukushima, Japan), and is denoted as “Iwaki Sample.” At the sampling time, the suspended particle concentration in the river water was 0.81 g/kg. The samples were collected from the two places on June 21 and 20, 2012, respectively. To increase the recovery efficiency of the suspended particles, we collected the samples on days just after typhoons. The river water samples containing suspended particles were collected using a Teflon container, about 20 L in total for each sample. The suspended particles were then collected on a filter with 0.45  $\mu\text{m}$  pores and

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