



Factors controlling radiocesium distribution in river sediments: Field and laboratory studies after the Fukushima Dai-ichi Nuclear Power Plant accident



Qiaohui Fan^{a,*}, Kazuya Tanaka^b, Aya Sakaguchi^{a,1}, Hiroaki Kondo^a, Naoko Watanabe^b, Yoshio Takahashi^{a,*}

^a Department of Earth and Planetary Systems Science, Graduate School of Science, Hiroshima University, 1-3-1 Kagamiyama, Higashi-Hiroshima, Hiroshima 739-8526, Japan

^b Institute for Sustainable Sciences and Development, Hiroshima University, 1-3-1 Kagamiyama, Higashi-Hiroshima, Hiroshima 739-8530, Japan

ARTICLE INFO

Article history:

Available online 12 July 2014

Editorial handling by M. Kersten

ABSTRACT

This study used laboratorial estimations and determined the distribution coefficient (K_d) of ^{137}Cs in river sediments sampled from the Abukuma River and the Kuchibuto River to investigate the particle size dependence of RCs distribution. Results show that the K_d patterns of ^{137}Cs (particle size-dependence of K_d) were not only related to the particle size of large particles (low clay mineral content) but also to the clay mineral content of small particles (high clay mineral content) and particularly cation concentration in aqueous phase. By contrast, the K_d patterns of stable Cs (^{133}Cs) exhibited no obvious particle size dependence. Adsorption species of Cs that was added to the river sediments at various particle sizes was almost the same at the molecular scale as determined by the extended X-ray absorption fine structure.

Our findings indicate that river sediments have high fixation ability to ^{137}Cs . Nevertheless, adsorbed ^{137}Cs can be extracted from sediments in the water phase when salinity becomes high, such as that in seawater. The distribution patterns of ^{137}Cs at various particle sizes can slowly down the equilibrium of the adsorption of ^{137}Cs during the transportation of river sediments, and then the behavior of ^{137}Cs should eventually be similar to that of stable Cs in the river system. Therefore, the particle size of sediments is an important factor in the distribution of RCs at the early stage of its deposition. In the end, RCs adsorbed on the sediments will be equilibrated with the stable Cs during its transportation in the river system. Controlling factors such as the mineralogy of sediments could possibly make the distribution patterns of RCs similar to that of stable Cs in the long run.

© 2014 Elsevier Ltd. All rights reserved.

1. Introduction

The massive earthquake on March 11, 2011 and the tsunami that followed inundated the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) with floodwater as high as 15 m. This catastrophe resulted in the loss of power and the eventual disruption of the controls of the cooling systems of the FDNPP. A series of core melt and hydrogen explosions occurred from March 13 to 15, 2011 at Units 1, 2, 3, and 4 of the power plant (Buessler et al., 2011; Tanaka et al., 2012, 2013). The event also led to the emission of a large amount of radioactive materials from the FDNPP to the

environment. The radioactive contaminants in the soil samples collected from the Fukushima Prefecture near the FDNPP after the accident included the following: $^{129\text{m}}\text{Te}$, ^{129}Te , ^{131}I , ^{132}Te , ^{132}I , ^{134}Cs , ^{136}Cs , ^{137}Cs , ^{140}Ba , and ^{140}La (Endo et al., 2012). Three volatile radionuclides, ^{134}Cs , ^{137}Cs , and ^{131}I were mainly responsible for the radioactivity in the contaminated zone surrounding the FDNPP (Yoshida and Takahashi, 2012; Yamaguchi et al., 2012). However, ^{131}I , which has a half-life of 8.02 days, decayed within half a year. A cause for concern is ^{137}Cs , which has a long half-life of 30.1 years and whose deleterious effects on agriculture, stock farming, and human life can last for the coming decades (Yasunari et al., 2011).

After the FDNPP accident, many studies have been conducted on the behavior and properties of radiocesium (RCs), including its distribution, redistribution, and transportation in aerosol, soil, river, and ocean environments. For example, the depth of RCs infiltration in the Fukushima area was reportedly deeper than that in the cultivated soil near the Chernobyl Nuclear Power Plant because of the relatively low clay content of Japanese soil (Kato et al., 2012). Less than

* Corresponding authors. Present address: Department of Earth and Planetary Science, Graduate School of Science, The University of Tokyo, Hongo, Bunkyo-ku, Tokyo 113-8654, Japan. Tel.: +81 82 424 7460; fax: +81 82 424 0735.

E-mail addresses: fanqiaohui@gmail.com (Q. Fan), ytakaha@eps.s.u-tokyo.ac.jp (Y. Takahashi).

¹ Present address: Graduate School of Pure and Applied Sciences, University of Tsukuba, Tennodai, Tsukuba, Ibaraki 305-8577, Japan.

1% of RCs in the topsoil from the Fukushima area can be extracted with water under various pH conditions, even with 2 M HCl solution; however, the extraction yield can increase up to 5–15% with 1.0 M NH_4Cl solution, which suggests that RCs is strongly adsorbed onto the frayed edge site (FES) and interlayer of clay minerals (Wauters et al., 1996; Tanaka et al., 2012, 2013). The extended X-ray absorption fine structure (EXAFS) has been successfully employed in exploring the adsorption species of Cs in clay minerals, Fukushima soils, and river sediments (Bostick et al., 2002; Qin et al., 2012; Fan et al., 2014a,b). Cs^+ can form both outer-sphere (OS) and inner-sphere (IS) complexes on clay minerals and soils, the ratio of which can be used to explain the strong fixation ability of Cs in a natural medium (Qin et al., 2012; Fan et al., 2014a,b).

He and Walling (1996) observed a quantitative relationship between the laboratorial distribution coefficient of ^{137}Cs (K_d^L) and the particle size of several types of homogenized mineral soil samples. The values of K_d^L in small particles were also found to be much larger than those in sandy soil with a particle size of over 63 μm . K_d^L was thus concluded to be a function of particle size, where the adsorption of RCs increases exponentially with the apparent surface area of each soil fraction. By contrast, no obvious dependence was found between the distribution of ^{137}Cs in the bottom sediments from Lake Michigan and its particle size (Alberts and Muller, 1979). After the FDNPP accident, the occurrence of ^{137}Cs in particle-size fractionated suspended particulates from the Abukuma River (AR) indicated that silt fraction (3–63 μm) exhibits maximum contribution to RCs. However, the concentration of ^{137}Cs (Bq/g) in each size fraction does not show any systematic change across various sampling sites (Sakaguchi et al., in press).

The distribution of Cs in a given environment is strongly related to the chemical and physical properties of the soil and other particulate matters, such as mineralogy, pH, cation exchange capacity, organic matter content, and coexisting cations, including Na^+ , K^+ , Ca^{2+} , and NH_4^+ in the aqueous phase (Poinssot et al., 1999; Kim et al., 2006; Konopleva et al., 2009; Fan et al., 2014a). Nevertheless, whether the adsorption of RCs onto sediments is mainly driven by particle size remains a controversy. Determining the distribution pattern of RCs in sediments with different particle sizes is important for several reasons. First, the physical movement of contaminated sediment particles is the most important transportation mechanism of RCs in a river system. Second, the re-suspension and inhalation of contaminated sediment particles has a relatively direct pathway to the biosphere, and the ease of the re-suspension of sediment particles related to their persistence and inhalation is dependent on size-related properties (Livens and Baxter, 1988). Finally, the distribution of RCs in sediments with different particle sizes is also crucial in estimating the migration behavior of RCs in the land-river-sediment-estuary system.

The present study investigates the distribution coefficient (K_d) of ^{137}Cs in river sediments at various particle sizes in the environs of Fukushima Prefecture and compares it with those measured by laboratory studies. The study also examines both the adsorption and desorption of RCs in river water and artificial seawater (ASW) to understand the behavior of RCs transported from rivers to estuaries. EXAFS and X-ray diffraction (XRD) analyses were used to clarify the factors controlling the fixation of RCs across different particle size fractions of river sediments.

2. Experimental

2.1. Sampling

Fig. 1 shows the sampling sites of river sediments from the AR and the Kuchibuto River (KR). The AR, the largest river in Fukushima Prefecture, runs through the Fukushima and Miyagi Prefectures

and discharges into the Pacific Ocean. It has a watershed area of 5390 km^2 and a population of about 1.2 million people along its basin (Takeda et al., 2001). The KR is one of tributaries of the AR and its source is situated in the Yamakiya district, Kawamata-town, within the evacuation zone (about 30 km from the FDNPP), and converges with the AR at Nihonmatsu City.

River sediments were collected at sites A, B, C, D, and E (estuary), as marked in the sampling map in Fig. 1. The precise northern latitude (N) and eastern longitude (E) of the sampling sites are shown in Table 1. The sample surface sediments were collected from a depth of 5 cm from the surface across a sampling area of 25 cm \times 25 cm. A plastic cup with a height of 6.5 cm and a diameter of 5 cm was used to collect the samples. The collected sediments were homogenized and then fractionated into several fractions with the following particle sizes: <2.0 μm , 2–10 μm , 10–20 μm , 20–40 μm , 40–63 μm , 63–125 μm , 125–250 μm , 250–500 μm , and 500–850 μm ; sedimentation and wet sieving methods were employed in the process (Livens and Baxter, 1988; Tanaka et al., in press). The samples were then dried at room temperature for further use in the sorption and desorption experiments.

The river water was filtered through a 0.45 μm membrane filter, and its ion composition was analyzed using ion chromatography (Thermo Scientific ICS-1100). For the stable cesium (^{133}Cs) analysis, the river sediments were decomposed with a mixed acid (HF and HClO_4) in a Teflon vessel with a screw cap via heating (ca. 180 $^\circ\text{C}$) for three days. The samples were then evaporated to dryness and re-dissolved into 2% HNO_3 solution (Takahashi et al., 2002). The concentration of ^{133}Cs in the solution was measured using inductively coupled plasma-mass spectrometry (Agilent Technologies, Agilent 7700). The concentrations of ^{133}Cs and RCs in both aqueous and solid phases allowed us to obtain the natural K_d values, indicated hereinafter as K_d^N .

2.2. Chemicals

A carrier-free tracer of ^{137}Cs in 0.1 mol/L HCl solution was purchased from Eckert & Ziegler Company, USA. ASW was synthesized strictly following the recipe of Tokunaga et al. (2013): 440 mmol/L of NaCl, 50 mmol/L of MgCl_2 , 27 mmol/L of Na_2SO_4 , 9.6 mmol/L of CaCl_2 , and 2.2 mmol/L of NaHCO_3 . All the other chemicals used in the experiments were of analytical grade and were not subjected to any purification or pretreatment.

2.3. Adsorption and desorption experiments with ^{137}Cs tracer

The adsorption of ^{137}Cs on the sediments was examined in polyethylene test tubes under ambient conditions. Then, 0.050 g of sediments with various particle sizes was equilibrated with 5.0 mL of river water from site C for 12 h at 20 $^\circ\text{C}$ and was filtrated using a membrane filter with a pore size of 0.45 μm . Then, 300 Bq of carrier-free tracer of ^{137}Cs was spiked into the suspension systems. After 24 h, the suspension was separated through centrifugation at 2000 rpm for 30 min, and the supernatant was filtered using a membrane filter with a pore size of 0.2 μm (polycarbonate, ADVANTEC). The radioactivity of ^{137}Cs in the filtrate was measured using a NaI detector. All the experimental data were measured thrice; their relative standard deviations were less than 5.0%.

After the adsorption experiments, sequential desorption experiments were conducted in the river water of site C and then in the ASW. The solid phases adsorbing ^{137}Cs were equilibrated with 5.0 mL of river water for 48 h and then separated. The separation process and the radioactivity analysis were strictly the same as those in the adsorption experiment described above. The solid phase was further spiked into 5.0 mL ASW. K_d (mL/g) was

Download English Version:

<https://daneshyari.com/en/article/4435789>

Download Persian Version:

<https://daneshyari.com/article/4435789>

[Daneshyari.com](https://daneshyari.com)