Journal of Environmental Radioactivity 195 (2018) 40–53

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Radiocesium and ^{40}K distribution of river sediments and floodplain deposits in the Fukushima exclusion zone

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ARTICLE INFO

Keywords:
Fukushima accident
Particle size distribution
Radiocesium
^{40}K
Riverbed sediment
Floodplain deposit

ABSTRACT

In this study, radiocesium and ^{40}K analysis were accomplished for samples of riverbed sediments and floodplain deposits collected from five rivers in the vicinity (< 20 km) of the damaged Fukushima Daiichi Nuclear Power Plant after seven years of the accident. Sediment particle size distribution and major oxide content were determined also for six selected samples to understand the retention and migration process of radiocesium in river environments. The radiocesium activity concentration varied from 103 ± 6 Bq·kg^{-1} to 22,000 ± 500 Bq·kg^{-1} in riverbed sediments and from 92 ± 5 Bq·kg^{-1} to 117,000 ± 2000 Bq·kg^{-1} in floodplain deposits. The ^{134}Cs/^{137}Cs ratio (decay corrected to 15 March 2011) was 1.02 in the both samples. Compared to monitoring results in 2011, it was proved that the radiocesium distribution pattern had been changed remarkably during seven years. The radiocesium was primarily attached to fine clay particles but its sorption on sand and coarse sand particles was also considerable. The sorption process of radiocesium was not affected by the presence of water and moreover, after seven years of the Fukushima accident, a significant radiocesium migration cannot be expected without particle migration. Consequently, radiocesium will remain for a long time in the river environments and its redistribution is mainly affected by the erosion process of the sediments. The average ^{40}K activity concentration of riverbed sediment and floodplain deposit samples was 640 ± 152 Bq·kg^{-1} changing from 319 ± 18 Bq·kg^{-1} to 916 ± 41 Bq·kg^{-1}. In the river estuary zones, significant activity concentration decrements were observed for both radionuclides. This suggests that seawater intrusion has a decreasing effect on both natural and artificial radionuclides via wash-out of particulate radiocesium and ^{40}K, and desorption of these radionuclides, but to reveal the detail of this process further investigations are required. The analysis of ^{40}K can help in a simple and easy way to reveal the mineral composition differences of sediment samples.

1. Introduction

On 11 March 2011 a huge tsunami, triggered by the most powerful earthquake ever recorded in the modern history of Japan, hit the country's eastern coastal region (Baba M., 2013) The tsunami caused thousands of deaths and damaged facilities of the Fukushima Daiichi Nuclear Power Plant (FDNPP) which finally led to a level 7 nuclear accident on the International Nuclear and Radiological Event Scale (INES) (IAEA, 2011). During the accident, huge amounts (more than 10^{17} Bq) of nuclear fission products, such as ^{129m}Te, ^{131}I, ^{134}Cs, ^{137}Cs, ^{90}Sr, etc., were released into the atmosphere and the Pacific Ocean (METI, 2011). Seven years after the emission, ^{137}Cs (half-life 30.17 y) remains as the major concern from a radiological safety viewpoint whereas ^{134}Cs (half-life 2.06 y) decays more rapidly due to its shorter half-life (Kakehi et al., 2016). Significant radioactive contamination affected mountain forest areas as well as urban and rural residential and agricultural zones (Hosoda et al., 2013; IAEA, 2011).

To minimize the radiological health risks of the Fukushima accident, intensive clean-up efforts have been successfully accomplished in contaminated urban areas (Yasutaka et al., 2016; Fujiwara et al., 2017; Mori et al., 2017). By contrast, decontamination activities in forest districts are much more difficult to carry out, and ^{137}Cs (hereafter referred to as radiocesium) will remain there for a long time.

The mobility of accidentally released radiocesium is influenced by the ratio of its chemical forms in fallout and site-specific environmental features regulating the rates of wash-out, fixation-remobilization, as

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https://doi.org/10.1016/j.jenvrad.2018.09.003
Received 5 July 2018; Received in revised form 4 September 2018; Accepted 5 September 2018

well as sorption-desorption of the mobile fraction (Konoplev et al., 2016). The Fukushima-released radiocesium had an aerosol form in the atmosphere and was deposited on the ground as directdry deposition and by precipitation as wet deposition. After the deposition it was rapidly attached to soil and sediment particles containing micaceous clay minerals (illite, vermiculite etc.) (Kozai et al., 2012; Guzman et al., 2013; Kaneko et al., 2015). In the course of time, the radiocesium has migrated vertically, as colloids with water or bounded to soil particles, in the soil profile (Konoplev et al., 2016; Bossew and Kirchner, 2004; Mishra et al., 2016). Heavy rainfall had some impact on the vertical distribution of radiocesium immediately following the Fukushima accident but in general, radiocesium is strongly bonded to soil particles, thus, deep layer soil contaminations have not been reported from Fukushima (Matsunaga et al., 2013). Typically, the 80–90% of the radiocesium has been found in the upper 5 cm layer of soils in Fukushima Prefecture (Mishra et al., 2014; Konoplev et al., 2018).

The explanation for contamination of floodplain deposit is more complex compared to soil because of the intensive sedimentation-erosion processes (Konoplev et al., 2016), especially in the Fukushima area as it has a monsoon climate with an annual precipitation of up to around 2000 mm according to the Japan Meteorological Agency database (JMA, 2018). Under this climate the summers are hot and rainy while the winters are mild (monthly average above zero) without soil freezing. The maximum precipitation and floods usually occur during the rainy season (late May - mid-July) and typhoon season (mid-August - October) in the exclusion zone (Konoplev et al., 2016, 2018). Fine particles on the surface soil have later been mobilized by rainfall events and transported gradually to mountain streams, rivers and floodplain areas (Kurikami et al., 2014). This lateral transport redistributes radiocesium contamination resulting decrements or enrichments (secondary contamination) in the amount of radiocesium in the affected area.

Depending on the water stream flowrate which influences the sediment deposition rate, the particulate radiocesium can be accumulated into river sediment layers or moved over great distances from upstream to downstream (Sakaguchi et al., 2015; Tanaka et al., 2015; Yamada et al., 2015). Water supplies from rivers are used for agricultural purposes and human consumption. Consequently, the radiocesium contamination in rivers and riverbed sediments have direct impact on aquatic plant and animal species and an indirect impact on humans (Tsuboi, 2015).

In Fukushima prefecture, the Abakuma river is the largest with the biggest radioactive fallout-affected catchment, hence, most research studies have concentrated on investigating the radiocesium transfer of this area, however this river is relatively far ($>$ 50 km) from the FDNPP (Evrard et al., 2015). Related to river basins in the exclusion zone around the FDNPP, limited data have been reported (Koibuchi et al., 2015). Therefore, it is important to acquire more information about radiocesium distribution patterns and mobility in river systems in the Fukushima exclusion zone due to long-time presence of radiocesium in the environment.

Cesium is a member of alkali metals in Group 1 of the periodic table. Amongst alkali metals, potassium is the most relevant from a natural radioactivity perspective, since its extremely long-lived ^{40}K isotope (1.3×10^9 y) which is a naturally occurring primordial isotope has existed since the formation of the Earth. Potassium, along with ^{40}K, is widely distributed in the environment and it is the eighth most common element on Earth and comprises around 1.8% of the its crust. ^{40}K emits high energy gamma radiation (1461 keV), therefore, it is detectable together with radiocesium (661 keV) on the same gamma spectra of environmental samples.

Since alkali metals show chemical similarity and ^{40}K data is easily available during radiocesium measurement, a decision was made in this study to examine ^{40}K activity concentration to gain some additional information to better understand radiocesium redistribution and mobility.

According to a large-scale natural radioactivity study wherein hundreds of soil samples from the central region of Japan were analyzed, the ^{40}K activity concentrations in soils was directly influenced by the mineral composition of the soil. It was found that the lower level of ^{40}K (75–150 Bq·kg^{-1}) typically attributes to soil samples originated chiefly from weathering of ultrabasic rocks (primarily composed by quartz, amphibole, olivine and chlorite), while higher level of ^{40}K (450–1400 Bq·kg^{-1}) typically presents in granite weathered soil samples (mainly composed by quartz, microcline, albite and biotite) (Megumi et al., 1988). In the Fukushima area, the soil type is dominated by weathered granite (Takahashi, 2017), and higher ^{40}K concentrations were confirmed by a previous study after the Fukushima accident in December 2011. In this previous study, wherein 28 soil samples were collected within 30 km radius from the FDNPP, the average of ^{40}K activity concentration was 650 Bq·kg^{-1} in the range from 130 to 1000 Bq·kg^{-1}. Excluding the sample with the minimum value, 130 Bq·kg^{-1}, it was reported that every sample has higher ^{40}K activity concentration than 400 Bq·kg^{-1} (Taira et al., 2012).

The aim of this study was to investigate the radiocesium activity concentrations and ^{40}K additionally, and monitoring their distribution along rivers in riverbed sediments and floodplain deposits from the Fukushima exclusion zone. For this purpose, sampling points were selected along five river systems, namely the Ukedo River, Takase River, Maeda River, Kuma River and Tomioka River, all within 20 km distance from the FDNPP. In this research, every sample was measured by gamma spectrometry and some selected samples were analyzed for particle size distribution and major oxide content.

2. Materials and methods

2.1. Study area and sampling methods

In this study five river areas and 20 sampling points were selected for sample collection. Three rivers, Ukedo (3 points) and its tributary Takase (4 points) and Maeda (5 points) are in the Northwest direction from the FDNNP and two others, Kuma River (3 points) and Tomioka River (5 points) are in the Southwest direction. All these rivers are located in the 20-km exclusion zone around the FDNPP. The headwaters of the rivers are based in the Abukuma highland, which consists of Cretaceous granite, and the mouths of the rivers are in the Hamadori plain at the Pacific coast. The slope gradients of the rivers from the Abakuma highland to the Hamadori plain is around 1.5% (1.2–1.7%) but in the Hamadori plain the slope gradients become very gentle with an average value of 0.1%. The Maeda river is the shortest with 17 km length and the Ukedo is the longest with 45 km. The others have similar length, around 30 km (Saegusa et al., 2016; Teranaka et al., 1990).

At each sampling point one river bottom sediments (BS) and one floodplain deposits (FD) sample was collected during July and August 2017. This field study resulted in a total of 40 samples. Sampling points were chosen according to the accessibility of the river but consideration was taken to avoid possible disturbance from road dust and drainage pipes. For each river, samples were collected from the upper, middle stream and from the estuary area to cover different slope gradient zones.

The BS samples were collected from the top $\sim$5 cm layer of river bottom sediments at the edge of river using a 5 cm diameter Wildco® hand core sediment sampler. At each point, five samples were taken and mixed from an area of about 1 m^2, four from four vertices and one from the centre point. The wet sediment samples were stored in polyethylene bags and transported to laboratory for processing. Top layer FD samples were collected using a Daiki soil sampler up to 5 cm depth, stored in polyethylene bags and transported to laboratory. At each sampling point, distance between BS sample (edge of river) and FD sample varied between 5 and 10 m. All samplings were carried out by a single member of the field staff in order to avoid sampling biases. For FD samples, the depth profiles of radiocesium may differ from BS because of the natural