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Quantification of dissolved and particulate radiocesium fluxes in two rivers draining the main radioactive pollution plume in Fukushima, Japan (2013–2016)

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ABSTRACT

Radionuclides released by the accident at the Fukushima Daiichi Nuclear Power Plant included ^{137}Cs (Cesium-137) and ^{134}Cs (Cesium-134), which were deposited on land as a result of fallout and concentrated in the uppermost 2–5 cm of the soil. In this study, river monitoring was conducted for 3.5 years to quantify dissolved and particulate radiocesium fluxes in the Mano and Hiso Rivers in Fukushima Prefecture from 2013 to 2016. Total ^{137}Cs export was estimated to be 191 GBq from the Hiso River and 26 GBq from the Mano River during the 3.5-year study. Annual particulate ^{137}Cs export during rainfall events accounted for 90–98% of total ^{137}Cs export. Annual dissolved ^{137}Cs export during rainfall events accounted for 1.5–3.3% of total export. The annual sum of particulate and dissolved ^{137}Cs exports during low-flow periods accounted for 0.6–6.5% of total export. Hence, radiocesium was redistributed mainly by transport of particulate radiocesium bound to suspended sediments. During the study period, 0.7% and 0.6% of the ^{137}Cs initially deposited on soil were exported by the Hiso River and Mano River, respectively. The slopes of regression lines relating fine suspended sediment concentrations and particulate ^{137}Cs concentrations in river water declined from 2013 to 2016 by 79% for the Hiso River and 83% for the Mano River. Especially noteworthy was a sharp decrease of the annual mean ^{137}Cs concentration of SS in 2016. This decrease was much greater than the decrease that would have been expected from the physical decay of ^{137}Cs and was probably related to changes of soil erosion processes due to heavy rainfall and the effects of decontamination efforts in 2015.

1. Introduction

Large amounts of radionuclides, especially radiocesium (Cesium-137, ^{137}Cs, and Cesium-134, ^{134}Cs) and radioiodine (Iodine-131, ^{131}I), were released into the atmosphere by the accident that occurred at the Fukushima Daiichi Nuclear Power Plant (FDNPP) in March 2011 (e.g., Chino et al., 2011; Terada et al., 2012; Yamamoto, 2015). The FDNPP accident resulted in considerable deposition of radiocesium on the soil in and around Fukushima Prefecture (official monitoring data are available from Ministry of Education, Culture, Sports, Science and Technology, Japan (MEXT, 2012). Residents within the seriously contaminated area were forced to move. Intensive decontamination efforts, including the removal of vegetation and topsoil around houses and agricultural lands, were conducted between April 2013 and March 2017 (Ministry of the Environment Government of Japan (MOE, 2017; Evrard et al., 2016). Six years after the accident, in April 2017, evacuation orders were lifted for the area, with the exception of specified zones referred to as “Returning is Difficult” where annual dose is greater than 50 mSv y^{-1} (Ministry of the Environment Government of Japan (MOE, 2017). However, a large amount of radiocesium remains in upland forests on the plateau, which were not the target of decontamination efforts. The remaining radiocesium has been redistributed with water and sediment transport (many reports are reviewed in Evrard et al., 2015).

Radiocesium transfer involves two main forms of radiocesium: dissolved and particulate. Dissolved radiocesium exists as cesium ions and cesium ion hydrates, which are easily transported by flowing water (Tsuji et al., 2014). Shiozawa (2013) has pointed out that radiocesium is strongly bound to clay particles within

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2–3 months after initial deposition. Radiocesium bound to particles, referred to as particulate radiocesium, may then be detached by raindrops and surface water flow. Evrard et al. (2015) have summarized the fluxes of particulate ^{137}Cs transported by several rivers in Fukushima from 2011 to 2013 based on studies published in the literature (e.g., Nagao et al., 2015; Shinomiya et al., 2014; Yoshikawa et al., 2014). These results showed that the particulate fraction was higher dominated than the dissolved during rainfall events, though it varied with time and between catchments. Evrard et al. (2015) have also indicated that the particulate phase accounts for the majority of radiocesium transfer.

Particulate radiocesium enters river water via erosion from hillslopes, the riverbed, and riverbank, and it is then transported with the suspended sediment. Many reports have been published concerning radiocesium dynamics in rivers and catchments. Shinomiya et al. (2014), Matsunaga et al. (2016), and Iwagami et al. (2017) have conducted field monitoring in a small catchment in Fukushima Prefecture (0.012–0.6 km^2). Those studies have enabled quantification of radiocesium export via erosion. Shinomiya et al. (2014) concluded that the flux of ^{137}Cs from forested headwaters was negligible compared to the ^{137}Cs inventory deposited on soils. Iwagami et al. (2017) likewise concluded that ^{137}Cs export was only 0.02–0.3% of the ^{137}Cs inventory. Matsunaga et al. (2016) found a decline of ^{137}Cs concentrations of suspended sediment during 2011–2013 in a forested catchment. The latter two studies used a time-integrated suspended sediment sampler (Phillips et al., 2000). Such a sampler is well designed to collect suspended sediment, but it cannot represent a time series of ^{137}Cs concentrations in sediment or river water.

Nagao et al. (2013), Tsuji et al. (2014), Sakaguchi et al. (2015), Yoshimura et al. (2015), and Eyrolle-Boyer et al. (2016) conducted field monitoring in a large catchment in and around Fukushima Prefecture. Those studies often involved multiple observation points in a river system or in multiple river systems. Tsuji et al. (2014) and Eyrolle-Boyer et al. (2016) focused on the relationships between deposited ^{137}Cs or its inventory and dissolved/particulate ^{137}Cs concentrations from multipoint sampling during low-flow periods. Nagao et al. (2013) roughly estimated ^{137}Cs export from two catchments in 2011 on the basis of 12 water samples. Yoshimura et al. (2015) conducted multipoint suspended sediment sampling during a single rainfall event for evaluation of the solid/liquid distribution coefficient, K_d. Sakaguchi et al. (2015) focused on the size distribution of ^{137}Cs concentrations in suspended sediment based on multipoint sampling. These studies have enhanced understanding of the mechanisms that account for the form of ^{137}Cs in runoff and the spatial variations of ^{137}Cs dynamics. As a further study, quantification of annual radiocesium fluxes in rivers will be required for analysis of ^{137}Cs dynamics based on mass balance considerations.

Monitoring results were also used to simulate the redistribution of radiocesium in a watershed and/or the riverine transfer of radiocesium to the Pacific Ocean. Models of radiocesium transfer developed by Kinouchi et al. (2015), Wei et al. (2017), and Sakuma et al. (2017) were used to numerically simulate the total amount of radiocesium runoff. The calculated results were then compared with the radiocesium inventory. Further development of a numerical model will require that monitoring data be accumulated to calibrate and validate the model.

In this study, river monitoring was conducted to quantify dissolved and particulate radiocesium fluxes in two similar watersheds in Fukushima Prefecture. A distinctive feature of this study is that continuous river monitoring was conducted for 3.5 years, from 2013 to 2016, a time interval that included numerous rainfall events. The data collected during rainfall events and/or low-flow periods were used to investigate the relationships between particulate radiocesium concentrations and suspended sediment concentrations, turbidity, sediment particle sizes, and dissolved radiocesium concentrations. We also investigated the inter-annual variation of radiocesium export, which has been rarely studied using continuous-monitoring data.

2. Materials and methods

2.1. Study sites

The study sites were two watersheds in Iitate Village, Fukushima, Japan (Fig. 1). This village is located about 27–45 km northwest of the Fukushima Daiichi Nuclear Power Plant in the main radioactive pollution plume. The southern observation watershed was the Hiso River watershed (37°36′N to 37°38′N, 140°40′E to 140°48′E), which has a catchment area of 25.6 km^2. The Hiso River is a branch of the Niida River system. Another northern observation watershed was the Mano River watershed (37°43′N to 37°45′N, 140°40′E to 140°44′E), which has a catchment area of 10.8 km^2. The radiation levels were higher in the Hiso River watershed than in the northern Mano River watershed. The average ^{137}Cs inventory in the surface soil on 11 March 2013 was 1021 kBq m^{-2} and 422 kBq m^{-2} for the Hiso River watershed and Mano River watershed, respectively (original data from Ministry of Education, Culture, Sports, Science and Technology, Japan (MEXT, 2013).

Land uses in the Hiso River and Mano River watersheds are 74% and 75% forest, 14% and 18% upland fields (grasslands, vegetables, etc.), and 10% and 6% paddy fields, respectively (original data from MLIT 2018a). From the FDNPP accident in March 2011 until December 2016, all of the agricultural lands in this village were abandoned, and weeds covered the surface all of the time, except during a temporary period of decontamination work. In 2014, efforts to decontaminate residential lands and agricultural lands in these areas began with removal of the vegetation and topsoil. The decontamination efforts for agricultural lands were conducted from March or April 2015 to December 2016 in the Hiso area and from April 2014 to March 2016 in the Mano area, with the exception of specified zones referred to as “Returning is Difficult” in the Hiso area.

The soil types in the Hiso River watershed include Cambisol (52%), Andosol (44%), and Gleysol (4%); in the Mano River watershed the soil types are Cambisol (48%), Andosol (37%), Gleysol (9%), and Histosol (6%) (original data from Ministry of Land, Infrastructure, Transport and Tourism, Japan (MILT, 2018b). Forests and upland fields are located mainly on the Cambisol and Andosol. Paddy fields are located mainly on Andosol and Gleysol.

2.2. Continuous water and sediment monitoring

The monitoring systems we used in both rivers consisted of a rain gauge, water level sensor, water velocimeter, turbidity sensor, and water sampler (Fig. 2). A rain gauge (RG3-M, Onset Computer Corporation, Bourne, MA, USA) was attached to the top of a steel pipe on the river bank to measure precipitation. A water level sensor (U20-001-04, Onset Computer Corporation, Bourne, MA, USA) was attached to the steel pipe which was stabbed into the riverbed (Fig. 2). The water level sensor recorded pressure, which could be converted into water depth. A similar sensor placed in the air above the level of the river was used to compensate the first sensor for changes of barometric pressure. A two-dimension electromagnetic water velocimeter (Compact-EM, JFE Advantech Co. Ltd., Nishinomiya, Hyogo, Japan) was attached to the submerged pipe to measure the flow velocity (Fig. 2). Flow rate was determined by using the flow cross-sectional area calculated from water depth and transversal river bed topography; the velocity profile derived from the measured flow velocity was assumed to be logarithmic. An