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Modeling radiocesium transport from a river catchment based on a physically-based distributed hydrological and sediment erosion model

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ABSTRACT

The accident at the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) in March 2011 resulted in the deposition of large quantities of radionuclides, such as ¹³⁴Cs and ¹³⁷Cs, over parts of eastern Japan. Since then high levels of radioactive contamination have been detected in large areas, including forests, agricultural land, and residential areas. Due to the strong adsorption capability of radiocesium to soil particles, radiocesium migrates with eroded sediments, follows the surface flow paths, and is delivered to more populated downstream regions and eventually to the Pacific Ocean. It is therefore important to understand the transport of contaminated sediments in the hydrological system and to predict changes in the spatial distribution of radiocesium concentrations by taking the land-surface processes related to sediment migration into consideration. In this study, we developed a distributed model to simulate the transport of water and contaminated sediment in a watershed hydrological system, and applied this model to a partially forested mountain catchment located in an area highly contaminated by the radioactive fallout. Observed discharge, sediment concentration, and cesium concentration measured from June 2011 until December 2012 were used for calibration of model parameters. The simulated discharge and sediment concentration both agreed well with observed values, while the cesium concentration was underestimated in the initial period following the accident. This result suggests that the leaching of radiocesium from the forest canopy, which was not considered in the model, played a significant role in its transport from the catchment. Based on the simulation results, we quantified the longterm fate of radiocesium over the study area and estimated that the effective half-life of ¹³⁷Cs deposited in the study area will be approximately 22 y due to the export of contaminated sediment by land-surface processes, and the amount of ¹³⁷Cs remaining in the catchment will be reduced to 39% of the initial total within 30 y after contamination. This study provides a perspective on the transport of suspended sediments and radiocesium in catchments with similar land use and radiocesium contamination.

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1. Introduction

Following the accident at the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) in March 2011, enormous quantities of radionuclides, including ¹³⁷Cs and ¹³⁴Cs, were deposited over eastern parts of Japan. These radionuclides were further transported and diffused through atmospheric flow, watershed hydrological processes, and terrestrial ecosystem functions. The third airborne monitoring survey of ¹³⁴Cs and ¹³⁷Cs conducted after the accident (MEXT, 2011)

http://dx.doi.org/10.1016/j.jenvrad.2014.07.022 0265-931X/© 2014 Elsevier Ltd. All rights reserved. detected large amounts of radiocesium deposited over forested areas, and other land use types, such as agricultural land and builtup areas, were also contaminated. In forested land, most of the fallout was initially trapped by the tree canopy, some of which was then washed out by rainfall and gradually migrated into ground surfaces by throughfall and stemflow (Kato et al., 2012a). Atmospheric winds also contributed to the relocation of radiocesium deposited on the canopy and ground surfaces. In agricultural areas, the land surface typically shows seasonal changes in vegetative cover. When the accident occurred, in early spring, most of the agricultural land was probably not planted or vegetated, which resulted in enhanced direct deposition of radiocesium onto the soil surface and adsorption of large amounts of radiocesium by soil particles. In addition to the radiocesium deposited on vegetated surfaces and its adsorption into soil particles, more radiocesium

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was likely to be deposited on paved and rooftop surfaces in built-up areas, and the contaminants were delivered to drainage systems and rivers by surface runoff during rainfall events.

Once radiocesium reaches the ground surface and is sorbed onto the soil, especially finer soil particles such as clay and silt, the rainfall-runoff process plays an important role in its redistribution. In forested areas, short grasses and plant litter layers covering the surface act as a buffer controlling the transport of radiocesium by reducing soil erosion and temporarily storing the radiocesium within the layer. In agricultural land, soil erodibility decreases in the growing season as vegetated cover increases. In paddy fields, although complicated irrigation and drainage systems make it difficult to understand the behavior of sediment erosion and runoff, irrigation water and rainfall typically fill the land surface during the irrigation period, which is normally from April to August, and less sediment is eroded due to ponded water, while more surface runoff can occur. During the non-irrigated period, more erosion may be possible, although storage in surface depressions can reduce surface runoff with higher sediment concentrations. As a result, the different responses in runoff and erosion associated with each type of land use complicate the integrated behavior of contaminated sediments transported from mixed land use watersheds. Therefore, the modeling and prediction of radiocesium transport through the watershed hydrological system must take into consideration the characteristic responses that depend on the surface conditions of individual land use types. Surface runoff is a dominant component in the transport of soil particles from catchments, and large amounts of eroded soil particles with adsorbed radiocesium are transported during flood events. The surface runoff directly contributes to the transport of sediments and adsorbed radiocesium, while subsurface and groundwater flows play significant roles in forested catchments in generating relatively slow runoff components. These hydrological components, together with surface runoff, influence the concentration of suspended sediments, which further affects the erosion and deposition in overland flow and channel flow. Therefore, the relative magnitude of surface runoff and conditions of land use and land cover are both important in the transport of soil particles and the migration of adsorbed radiocesium.

On the other hand, dissolved radiocesium has been detected in water samples collected from rivers flowing through areas contaminated by radioactive fallout derived from the FDNPP. Sakaguchi et al. (2014) analyzed the water samples taken from the Kuchibuto River, which is the target river of this study, and found that dissolved ¹³⁷Cs in normal flow conditions was on average less than 10% of the total ¹³⁷Cs at the downstream site and about 20% at the upstream site. Ueda et al. (2013) found that dissolved radiocesium of streamflow in two small rivers located in Fukushima Prefecture was approximately 60% during normal flow conditions, while particulate radiocesium was the dominant form of radiocesium in flooding periods (90% of total radiocesium). Although the dissolved radiocesium occupies a certain ratio to the total, concentration of dissolved radiocesium (Bq/L) in the normal flow condition is much smaller than that during periods of flooding, resulting in a limited contribution to the total amount of radiocesium delivered to downstream regions.

In many previous studies, either conceptual, physical or system model was used to assess the fate of particulate and dissolved radionuclide in watersheds by involving many complex processes into the model (Monte et al., 2004; Garcia-Sanchez, 2008; Johnson and Dortch, 2014). For mountainous catchments with steep topography and unevenly distributed radiocesium deposition over various kinds of land surfaces, which is the situation found in our study area, it is particularly important to model rainfall-runoff processes and accompanying soil erosion and transport considering detailed condition of land use and hydrological conditions with fine spatial and temporal resolutions.

Our final objective is to predict long-term changes in the discharge of radiocesium to rivers and oceans, and the spatial distributions of radiocesium remaining on the land surface in accordance with various important factors and processes in the watershed hydrological system. In this study, we attempted to construct a watershed-scale hydrological model that takes into consideration the fundamental characteristics of rainfall-runoff processes in mountainous catchments with mixed land use to simulate the transport of eroded sediments and adsorbed radiocesium. We coupled a physically-based distributed hydrological model with modules for the erosion and transport of sediments and adsorbed radiocesium, and applied the coupled model to the Abukuma River watershed, which is located in the area of high radiocesium deposition from the FDNPP accident. The model takes into account the actual conditions of mixed land use and land cover, as well as the hydrological processes related to human activities, such as irrigation and drainage in agricultural fields. A continuous simulation was conducted to reproduce observed streamflow, concentration of suspended sediments, and the concentration of radiocesium included in sediments during the 19-month period from June 2011 to December 2012. The total amounts of sediments and radiocesium transported from the study area were quantified, and the relative significance of individual flood events was determined.

2. Study area

We studied the transport of water, sediments, and radiocesium in the Kuchibuto River catchment (catchment area $\approx 140 \text{ km}^2$), a sub-catchment of the Abukuma River watershed (Fig. 1), to develop a simulation model for predicting long-term changes in radiocesium transport through the watershed hydrological system, and the spatial distribution of radiocesium inventory on the ground surface. The Kuchibuto River catchment is located 33-49 km northwest of the FDNPP, and its upstream area has been studied extensively with regard to the migration of radiocesium (e.g., Kato et al., 2012b). According to 250-m resolution elevation data published by the Geospatial Information Authority of Japan (GSI), the altitude of the catchment ranges from about 200 to 1010 m a.s.l. The dominant land use types in the catchment are forest, cropland, and paddy fields (Fig. 1b, Table 1), covering 55%, 29%, and 11% of the catchment, respectively. The upstream areas are dominated by evergreen coniferous trees and deciduous broad-leaved trees. The natural topsoil in the catchment is classified as mainly either forest soil or Andosol (also known as Kuroboku, which is black soil rich in humus content). In paddy fields, clay soil forms the base layer that minimizes water infiltration into the ground. Results from the airborne survey (MEXT, 2011) showed deposition of relatively large amounts of radiocesium in upstream areas of the catchment (Fig. 1a).

Since June 2011, water level and turbidity have been monitored at 10-min intervals at a hydrological station downstream in the main channel (Fig. 1b, Yamashiki et al., 2014). The time series of the water level monitored by a pressure-transducer gauge were converted to those of flow rate using a rating curve obtained for the downstream site. Turbidity was monitored using an optical sensor (MacVan Analite turbidity meter, 3000-NTU) and the data were converted to the concentration of suspended sediments using the relationship between monitored turbidity and suspended sediments in collected stream water samples determined in laboratory experiments. In addition, suspended sediments were collected using a time-integrated sediment sampler (Phillips et al., 2000), which is made from a 1-m length polyvinylchloride pipe (internal

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