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Assessment of the caesium-137 flux adsorbed to suspended sediment in a reservoir in the contaminated Fukushima region in Japan

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ABSTRACT

We estimated the flux of caesium-137 adsorbed to suspended sediment in the Kusaki Dam reservoir in the Fukushima region of eastern Japan, which was contaminated by the Fukushima Nuclear Power Plant accident. The amount and rate of reservoir sedimentation and the caesium-137 concentration were validated based on the mixed-particle distribution and a sediment transport equation. The caesium-137 and sediment flux data suggested that wash load, suspended load sediment, and caesium-137 were deposited and the discharge and transport processes generated acute pollution, especially during extreme rainfall-runoff events. Additionally, we qualitatively assessed future changes in caesium-137 and sediment fluxes in the reservoir. The higher deposition and discharge at the start of the projection compared to the 2090s are most likely explained by the radioactive decay of caesium-137 and the effects of reservoir sedimentation. Predictions of the impacts of future climate on sediment and caesium-137 fluxes are crucial for environmental planning and management.

1. Introduction

Reservoir sedimentation and caesium discharge are dependent on particulate behaviour and the effects of extreme events and long-term river flows on suspended sediment within different environmental contexts. Sedimentation occurs in a variety of reservoir operations, including those conducted during extreme events and to flush sediment. Most recent models of reservoir sedimentation have considered a mixed particle distribution and emanation from reservoirs. The wash load and suspended load are the dominant components of deposited sediment in reservoirs and are discharged over a wide area around the source, including as delta formations at dam sites (Kreznor et al., 1990; Borretzen and Salbu, 2002; Hou et al., 2003; Ciffroy et al., 2005; Korobova, 2010; Bagarello et al., 2010; Gómez-Guzmán et al., 2011; Mouri et al., 2011a, 2011b, 2013a, 2013b, 2013c, 2013e). The occurrence and abundance of spherical particles have been used to identify sediments transported to reservoirs and river channels during flow (Wik, 1953; Kocher, 1981; Ciffroy et al., 2000; Mouri and Oki, 2010; Mouri et al., 2011a, 2011b, 2013a, 2013f).

Fine soil minerals, usually embedded in the turbulent flow, offer a convenient way to separate spherical particles for identification and analysis. Temporal variations in reservoir sedimentation and discharge have also been studied, including since the 1940s when many reservoirs began operating and river channel construction projects began (Lane and Kalinske, 1941). Fine sediment transport can also be traced from a stream channel into a reservoir, and fine sediment levels increase in streams where channel construction has taken place. In addition, fine sediment containing radioactive caesium may come from streams in which turbine power sources are operated (Harvey and Ruch, 1986; Locke and Bertine, 1986; Thiessen et al., 1999; Ivanova et al., 2000; Santschi and Schwehr, 2004; Golosov and Panin, 2006; Pinder et al., 2011).

Reservoirs and river channels can act as diffusors of radioactive material, with fine sediments containing radioactive caesium being transferred through a river channel to the ocean. Motivated by concerns regarding water supplies and caesium levels, this study used a general circulation model and climate change scenarios to project suspended sediment yield in the 21st century in a reservoir located in a region of Japan contaminated by the Fukushima Nuclear Power Plant accident (Messiha, 1984; Olson et al., 2003, 2006; Golosov and Panin, 2006; Pinder et al., 2005; Mouri et al., 2013b, 2013c; Rajkumar et al., 2013). The fine particles in sediment deposited in the reservoir can be used to identify sediment and

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radioactive caesium accumulation following the Fukushima accident and to interpret the hydrological circulation of sediments and radioactive substances. Katata et al. (2012) found that fine soils and large amounts of atmospheric spherical particles near the study area, and in other hotspots in the southern Fukushima region, contained radioactive caesium. Also in the Fukushima area, Tagami et al. (2011) found hotspots with higher levels of radioactive caesium than in the major contaminated territories in the Tokyo region, to the south of Fukushima. They also found that the concentration of radioactive caesium-137 in surface soils was >5000 Bq/kg near Fukushima and <100 Bq/kg near Tokyo in 2011.

Fine sediment has proven to be a better indicator of radioactive caesium than have organic carbon, magnetic minerals, or magnetic susceptibility. Olson et al. (2002) used the content of fine suspended particles (such as fly ash), magnetic susceptibility, magnetic minerals, and organic carbon as indicators of soil erosion at a site near Moscow, Russia, the entire catchment contained 12% less fly ash than a reforested site. Artificial caesium-137 Isot. can appear in the natural environment by hydrological circulation and sediment transfer. The accelerated development of such transfers was noted after the Chernobyl accident in 1986 in Russia and adjacent countries. In that case, in which widespread fallout occurred, researchers investigated the presence of caesium-137 in surface soil and its spatial variability in catchments (Ritchie and McHenry, 1990; Belyaev et al., 2003; Golosov et al., 1999; Golosov, 2000; Ivanova et al., 2000; Kuznetsov and Demidov, 2002; Bolsunovsky and Dementyev, 2011; Giussani and Risica, 2012).

Soil loss and deposition in a river channel can be estimated using caesium-137. Caesium-137 was not present in soils prior to the atmospheric detonation of fissionable weapons, which largely occurred in the late 1950s and early 1960s. During that time, caesium-137 was emitted into the atmosphere from the frequent testing of nuclear devices by the US and USSR, including at testing grounds in the South Pacific. Hence, there is a specific time frame associated with caesium-137 that can be used as a stratigraphic marker. The behaviour of radioactive caesium in sediment is largely controlled by adsorption to the surface of fine particles, and subsequently the migration by soil erosion and transfer; such movement can be estimated by technogenic magnetic tracer methods in slightly eroded chernozems (Huffman and Huggins, 1986; Jones and Olson, 1990; Loughran, 1994; Walling and Quine, 1992; Gennadiyev et al., 2005).

As noted above, the Chernobyl accident in 1986 resulted in caesium-137 fallout across vast areas of Russia. Two component markers were used in subsequent studies and differed not only in their origin and age, but also in their form of occurrence in the fine soils. The initial migration capacity of caesium-137 was greater than that of the magnetic spheres, which is explained by the partial translocation of caesium in the ionic form and its predominant sorption to suspended sediments associated with the wash and suspended loads, whereas magnetic spheres were larger in size (from 1 to 25 μm) (Schaeffer, 1975; Soil Survey Staff, 1999; Flury et al., 2004; Gennadiyev et al., 2005). The loss of caesium-137 always exceeded the loss of magnetic spheres. Melin et al. (1994), Shcheglov et al. (2001), and Geraskin et al. (2011) showed that a reservoir can contain higher caesium-137 levels than a forest or a mountainous area as a result of tree intercepts. Du and Walling (2011) used caesium-137 measurements to investigate the influence of erosion and soil particle redistribution on fine soil properties.

The objectives of the present study were to (1) determine the period of deposition of fine sediment with radioactive caesium (caesium-137), (2) confirm the current variability in the fine sediment and radioactive caesium deposition and discharge, and (3) predict long-term (100-year) sediment and caesium-137 fluxes in a reservoir in a contaminated territory using a general circulation model (GCM) based on radiocaesium migration and hydrodynamic approaches (Schaeffer, 1975; Whitehead, 1984; Golosov et al., 1999, Golosov, 2000; Hou et al., 2003; Kheiashy et al., 2010; Mouri et al., 2011a, 2011b, 2011c, 2011d, 2013e, 2013f; Viparelli et al., 2013).

2. Methods

The field site displayed aquatic sedimentation, and two parallel transects were selected for sampling and *in situ* measurements of caesium-137. The transects were spaced about 1000 m apart. Measurement points were established at the dam site and the upstream ends of the transects. We particularly focused on choosing reference locations at which caesium-137 inventories would be representative of the total inflow input from upstream. The topography of the field site was considered when deciding on sampling points. For sampling, a core tube was inserted to 30-cm depth on slopes and interfluve areas at points in each transect. Three cores were taken at each point to minimise the effects of spatial variability. In addition, incremental samples from layers at depths of 0–30 and 30–40 cm were taken at some sampling points within the interfluve area. In order to study the distribution of fallout radionuclides with depth in surface soils, sectioned soil samples were collected using a scraper plate, with a sampling area of 450 cm^2 (15 cm × 30 cm). The scraper plate has two components: a metal frame that is placed on the ground and an adjustable metal plate that can scrape or remove fixed increments of soil within the frame. Advantages of the device include that it is robust, can collect a large volume of material from a large surface area, has few moving parts, and is simple in construction (Loughran, 1994). Samples were taken in 0.5-cm increments for the depth range of 0–5 cm, 1.0-cm increments for the depth range of 5–10 cm, and 2.0-cm increments for the depth range of 10–30 cm. In order to prevent contamination from surface soils that might fall down from the wall of the sampling hole, spray glue was used to fix the wall. The collected soil samples were shipped to the laboratory in a sealed plastic container without oven drying or sieving to prevent the release of caesium-137 from the samples into the atmosphere. After the soils samples were analysed for radionuclides, they were dried at 105 °C for 12 h and lightly ground to pass through a 2-mm sieve; the weight percentages of soil (<2 mm, in diameter) and water content were then determined. The particle size distribution of topsoil (<10 cm depth) was analysed using a sieve method (>450 mm) and a laser diffraction particle size analyser (SALD-3100, Shimadzu Co., Ltd., Kyoto, Japan). These samples were used to evaluate the possible temporal migration of Fukushima-derived caesium-137. Simultaneously with the collection of the bulk samples, *in situ* measurements of caesium-137 activity were made adjacent to most of the sampling points (Govorun et al., 1994; Chesnokov et al., 1997). The viability of using a portable detector in areas with high levels of radionuclide contamination is discussed in detail elsewhere (Golosov, 2000). A detailed topographic survey of the study area, including the sampling and measurement points, was provided by the Japan Water Agency (JWA) and the Ministry of Land Infrastructure, Transport and Tourism (MLIT). All activities were corrected for the radioactive decay since 11 March 2011 when the Fukushima accident occurred. Information about reservoir operation, sedimentation, caesium-137 levels, and precipitation in the Dec 2011–Dec 2012 period was collected from the JWA and MLIT. All water and soil samples (dried and sieved to <2 mm) as well as standard reference materials and laboratory standards prepared from standard solutions were analysed for gamma-ray emissions at energies of 662 keV (caesium-137) using a high-purity n-type germanium coaxial gamma-ray detector (EGC25-195-R, Canberra-Eurisys, Meriden, CT, U.S.A.) with an amplifier (PSC822, Canberra, Meriden, CT, U.S.A) and multichannel analyser (DSA1000, Canberra). Count times were sufficient to provide a typical analytical precision of ±4–5%. The instrument was calibrated using standard gamma sources with different sample heights, and self-absorption corrections were made (Cutshall et al., 1983). Details of the standard gamma sources are as follows: (1) lead-210 and caesium-137, with total activity of 2.3–23 kBq (EG-CUSTOM; Isotope Products Laboratories, Valencia, CA, USA), and (2) americium-241, cadmium-109, and europium-152, with a total activity of 7.1 kBq (EG-CUSTOM; Eckert and Ziegler Isotope Products, Valencia, CA, USA). Measurement accuracy was certified by determination of caesium-137 and lead-210 in spiked soil samples (Soil sample 01; the IAEA-CU-2006-03 World-Wide Open Proficiency Test on the Determination of Gamma Emitting Radionuclides, IAEA/AL/171, organised by the International Atomic Agency in 2006). The sediment transfer and caesium-137 levels at the study site were validated using the Catchment Simulator model (Mouri and Oki, 2010a; Mouri et al., 2010, 2011a, 2011b, 2012a, 2012b, 2013a, 2013b) and a modified version of the Total Runoff Integrating Pathways (TRIP) model developed by Oki and Sud (1998). The modelling components considered were suspended sediment transport, deposition and discharge, the effects on geographical features of the Kusaki Dam, and the impact of future climate changes projected by the GCMs. One-dimensional (1-D) future simulations, based on meteorological forces, showed reasonable sediment and caesium-137 fluxes (Mouri et al., 2013a, 2013b, 2013f).

2.1. Study site description

The Kusaki Dam is located in eastern Japan and exerts an important topographical effect on the predominant south-easterly winds, which were the major