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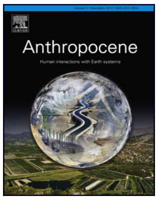

Investigating the source of radiocesium contaminated sediment in two Fukushima coastal catchments with sediment tracing techniques

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ABSTRACT

The Fukushima Dai-ichi nuclear power plant accident resulted in the fallout of significant quantities of radiocesium. After deposition on the soil surface, rainfall and spring flood events transfer radiocesium downstream. Identifying the source of contaminated sediment is important for managing potential downstream radiocesium contamination.

Soil ($n=37$) and sediment ($n=211$) were sampled from November 2011 to May 2014 in the Mano and the Niida coastal catchments. Two sediment fingerprinting approaches quantified the source of radiocesium contaminated sediment. First, cesium-137 activities in surface soil and sediment samples were modelled to determine the contribution of upstream, more contaminated areas, to sediment transiting the more densely populated coastal plain. Second, elemental geochemistry of three major soil types (Andosols, Cambisols and Fluvisols) was used to model the relative contribution of these soils to sediment sampled throughout the catchments.

In the Niida catchment, 47% (σ 19%) of sediment sampled in the coastal plain was modelled to be derived from the upstream area whereas, it was only 19% (σ 19%) in the Mano catchment. The main factor driving this difference is a large dam on the main stem of the Mano River. Geochemical modelling results indicated that Fluvisols, an alluvial soil type on which paddy fields are typically cultivated, supply the majority of sediment (μ 76, σ 14%).

The results confirm that the management of dams is fundamental to radiocesium migration. Moreover, this research indicates that Fluvisols and concomitantly, rice paddies on this soil type, supply a disproportionate amount of sediment. Managing sediment derived from Fluvisols, while incorporating potential impacts from major dams, should help mitigate the downstream migration of radiocesium contaminated sediment.

1. Introduction

The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident on March 11, 2011 resulted in the deposition of vast quantities of radionuclides over Japanese soils (for a review see: Evrard et al. (2015)). Among these radionuclides, cesium-137 (^{137}Cs $t_{1/2}=30.17$ y) will be the most serious health risk to the local population for the foreseeable future (Kitamura et al., 2014).

Cesium is rapidly and almost irreversibly fixated to fine soil particles, particularly clay minerals (Sawhney, 1972; Kamei-Ishikawa et al., 2008). Owing to this rapid fixation, ^{137}Cs is predominantly bound to fine particles in the top 0–5 cm of undisturbed soil profiles (Lepage et al., 2015). Importantly, these contaminated fine particles are preferentially eroded (Walling and Woodward, 1992; Motha and Wallbrink, 2002).

The Fukushima region has an erosive climate, particularly during the typhoon season (July–October)(Laceby et al., 2016). During typhoon events, significant volumes of contaminated sediment are transported downstream (Evrard et al., 2013; Lepage et al., 2014b). These major events result in elevated ^{137}Cs concentrations in suspended riverine material (Yoshikawa et al.,

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2014). For example, Typhoon Roke (September 2011) transferred 61% (6 TBq) of the total radiocesium load in the Abukuma catchment between August 2011 to May 2012 (Yamashiki et al., 2014). Understanding the climatic influence on radiocesium and sediment fluxes is important.

Only the largest river draining the main radioactive pollution plume, the Abukuma River, has been continuously monitored since the accident (Chartin et al., 2013). Alternative sample-based approaches are therefore required to examine sources of ^{137}Cs-contaminated sediment in the other coastal rivers (Chartin et al., 2013; Lepage et al., 2014a). In lieu of monitoring, sediment fingerprinting techniques provide a direct method to identify and quantify sediment contributions from different areas through analyzing and modelling source soil and sediment properties (Collins and Walling, 2002; Haddadchi et al., 2013).

In this post-accident context, sediment source contributions are quantified with two sediment fingerprinting techniques. First, a ^{137}Cs-based approach is used to investigate the downstream migration of ^{137}Cs-contaminated sediment. Here, ^{137}Cs activities are quantified in two distinct catchment areas: the upstream, highly contaminated region and the coastal plains that received low levels of ^{137}Cs fallout. The objective of this ^{137}Cs-based fingerprinting is to examine the source of ^{137}Cs-contaminated sediment transiting the more densely populated coastal plain.

Second, elemental geochemistry of sediment and three main soil types (Andosols, Cambisols and Fluvisols) is analysed to determine the relative contribution of these soils to ^{137}Cs-contaminated sediment. Conservative elements are selected for modelling with the Kruskal Wallis *H*-test and discriminant function analysis (Collins et al., 1997; Wilkinson et al., 2013). Distribution models are used to identify sediment sources for both fingerprinting approaches (Laceby and Olley, 2015).

For most of the Fukushima-impacted catchments, the downstream, more densely populated coastal plains were less-affected by the initial fallout. Therefore, it is important to understand the sources of contaminated sediment transiting these coastal plains. This application of sediment fingerprinting techniques improves our knowledge of the sources of ^{137}Cs-contaminated sediment transiting these coastal plains. This improved understanding will assist the long-term management of radioactive contamination in the Fukushima Prefecture.

2. Materials and methods

2.1. Study site description

This research was conducted in the Mano (175 km^2) and Niida (275 km^2) catchments (Fig. 1). The main features of these catchments include an upstream coastal mountain range (<900 m) and a broad, more densely inhabited, coastal plain (Geospatial Information Authority of Japan, 2015). Soils in the upstream areas of these catchments were heavily contaminated, with soil radiocesium (^{134}Cs + ^{137}Cs) inventories ranging from 20 kBq kg^{-1} to 150 kBq kg^{-1} (Fig. 1) (Chartin et al., 2013). In contrast, soil radiocesium inventories in the lowland coastal plains were less than 20 kBq kg^{-1}.

An important hydrological distinction between these catchments is the presence of a major dam on the main stem of the Mano River. The only major dam in the Niida River catchment is situated on a tributary (Fig. 1). Catchment land use mainly consists of forest (72%) and cropland (12%) (Land Conservation Research, 2005). Paddy fields constitute the majority of the cropland agriculture, typically cultivated on alluvial soils. Cambisols comprise 59% of the soil types for these two catchments, followed by Andosols (22%) and Fluvisols (7%) (Economic Planning Agency, 1972). Cambisols and Andosols are predominantly located in the upper catchment, whereas the Fluvisols are mainly located near the river channel in upstream reaches and are ubiquitous throughout the coastal plain (Fig. 4).

2.2. Soil and sediment sampling

Six sampling campaigns were conducted between November 2011 and May 2014. Sediment sampling occurred bi-annually: in November after the typhoon season, and in spring, after the snowmelt runoff (Fig. 2). The goal was to sample deposited sediment that was transferred during these main erosive periods. Cumulative rainfall reached 3300 mm at a gauging station in the Niida catchment monitored between March 2011 and May 2014. This monitoring period included three typhoons and one tropical storm. Mean annual rainfall in the Fukushima region is ~1400 mm (Laceby et al., 2016).

Fine sediment samples (n = 211) were taken from material deposited after the last major event at the same sites, during each of the six campaigns (Figs. 3 and 4). These lag deposit samples are comprised of fine particulate material that settled during the falling limb of the last significant hydro-sedimentary event. Lag deposit samples have proven to be comparable to *in-situ* suspended sediment samples in sediment fingerprinting research (Olley et al., 2013a). Ten subsamples (~5 g per subsample) of recent lag deposit material were taken with a plastic spatula over a 5 m reach, down to the underlying coarser cobble or gravel layer and composited into one sample. Lag deposit samples are referred to as sediment for the remainder of the text.

To investigate the source of ^{137}Cs-contaminated sediment transiting the coastal plain, the catchments were subdivided into their two inherent, dominant features (the upstream mountainous area and the coastal plain). The fallout from the contamination plume exhibits a somewhat similar upland/lowland pattern, with a radiocesium threshold inventory of 20 kBq kg^{-1} delineating these two distinct catchment features (Fig. 3). An elevation threshold of ~100 m provides a similar spatial delineation between these upland/lowland features (Fig. 1). As the goal is to trace the movement of ^{137}Cs-contaminated sediment, the threshold of 20 kBq kg^{-1} of radiocesium is used to delineate these two sources.

Two approaches were used to develop a ^{137}Cs source dataset for modelling. First, soil samples (n = 37) were collected in locations reported by Chartin et al. (2013) to be highly connected to the stream network. At each of these locations, ten subsamples (~5 g per subsample) were scraped from the soil surface randomly in a 10 m^2 quadrant using a non-metallic trowel and composited into one sample. Second, ^{137}Cs activities were included for 99 soil samples collected in the upper 5 cm of the soils in June and July 2011 in these two catchments by the Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT, 2012) (Fig. 3). Their ^{137}Cs activities remained in the same range as those measured in soil samples collected for this study.

To determine the relative contribution of the different soil types to ^{137}Cs-contaminated sediment, the highly connected soil samples from soil types that covered >5% of the catchment area were selected for elemental analyses. This 5% threshold excludes areas downstream from the lowest sediment sampling point as these soils do not contribute to sediment sampled. In addition, three soil samples were collected in the Ota catchment, south of the Niida catchment (Fig. 4). In total, 15 soil samples were analysed from Andosols, 12 from Cambisols and 7 from Gleyic Fluvisols (Fig. 4). Gleyic Fluvisols are here after referred to as Fluvisols.

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