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Research article

Do forests represent a long-term source of contaminated particulate matter in the Fukushima Prefecture?

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A B S T R A C T

The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident resulted in radiocesium fallout contaminating coastal catchments of the Fukushima Prefecture. As the decontamination effort progresses, the potential downstream migration of radiocesium contaminated particulate matter from forests, which cover over 65% of the most contaminated region, requires investigation. Carbon and nitrogen elemental concentrations and stable isotope ratios are thus used to model the relative contributions of forest, cultivated and subsoil sources to deposited particulate matter in three contaminated coastal catchments. Samples were taken from the main identified sources: cultivated ($n = 28$), forest ($n = 46$), and subsoils ($n = 25$). Deposited particulate matter ($n = 82$) was sampled during four fieldwork campaigns from November 2012 to November 2014. A distribution modelling approach quantified relative source contributions with multiple combinations of element parameters (carbon only, nitrogen only, and four parameters) for two particle size fractions (<63 μm and <2 mm). Although there was significant particle size enrichment for the particulate matter parameters, these differences only resulted in a 6% (SD 3%) mean difference in relative source contributions. Further, the three different modelling approaches only resulted in a 4% (SD 3%) difference between relative source contributions. For each particulate matter sample, six models (i.e. <63 μm and <2 mm from the three modelling approaches) were used to incorporate a broader definition of potential uncertainty into model results. Forest sources were modelled to contribute 17% (SD 10%) of particulate matter indicating they present a long term potential source of radiocesium contaminated material in fallout impacted catchments. Subsoils contributed 45% (SD 26%) of particulate matter and cultivated sources contributed 38% (SD 19%). The reservoir of radiocesium in forested landscapes in the Fukushima region represents a potential long-term source of particulate contaminated matter that will require diligent management for the foreseeable future.

1. Introduction

The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident deposited a significant quantity of radionuclides over Japanese soils (Chino et al., 2011; Evrard et al., 2015; Groëll et al., 2014). Of these radionuclides, radiocesium represents the most serious threat for the foreseeable future (Kitamura et al., 2014; Saito and Onda, 2015). Radiocesium is quickly and almost irreversibly bound to fine soil particles (Saito et al., 2014; Sawhiney, 1972) and predominantly stored within the top five centimetres of undisturbed soil profiles (Lepage et al., 2015; Matsuda et al., 2015). As the majority (~66%) of the landscape receiving high levels of radiocesium fallout (i.e. >1000 kBq m^{-2}) were a mixture of evergreen and deciduous forests (Hashimoto et al., 2012), a significant quantity of radiocesium is also stored within the forest canopy, leaf litter, and soil organic matter (Hashimoto et al., 2013; Koarashi et al., 2012; Loffredo et al., 2014).

Japanese authorities have conducted an extensive decontamination program targeting mainly rural residential and cultivated landscapes, including paddy fields (Sakai et al., 2014; Yasutaka and Naito, 2016). Among other activities, decontamination removes

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vegetation and replaces topsoil (~5 cm). As these rural residential areas are progressively decontaminated, forests will become the main reservoir of radiocesium contamination. Importantly, these radiocesium contaminated forests are typically upslope, or upstream, of decontaminated landscapes.

In this region, researchers have demonstrated that rice paddy fields are a major sediment source (Chartin et al., 2013; Lepage et al., 2016). Conversely, soil erosion and radiocesium export from forest environments is thought to be limited (Shinomiya et al., 2014; Yoshimura et al., 2015) as a thick litter layer of organic matter covers the soil surface, minimizing rainfall driven soil erosion. A key question for ongoing radiocesium management is whether contaminated forest landscapes represent a potential source of radiocesium to downstream and downslope environments.

One approach to examining the potential of forested landscapes to contribute contaminated particulate matter downstream is to directly trace the properties of this material back to their potential sources. Sediment tracing techniques provide a direct method of determining sediment provenance (for reviews, please see: Collins and Walling, 2004; Davis and Fox, 2009; Guzmán et al., 2013; Haddadchi et al., 2013; Koiter et al., 2013). Sediment tracing research has demonstrated that large dams reduce downstream migration of contaminated sediments (Lepage et al., 2016), that decontamination reduces downstream radiocesium transfers (Evrard et al., in review), and that alluvial soils, and the rice paddies occupying them, contribute disproportionately more sediment downstream relative to their spatial extent in the Fukushima region (Lepage et al., 2016).

In this study, the composition of particle borne organic matter is used to test the hypothesis that forested landscapes are a potential source of contaminated matter in three coastal catchments of the Fukushima Prefecture. In particular, source contributions (i.e. cultivated, subsoil, forest) are quantified with the analyses and modelling of total organic carbon (TOC), total nitrogen (TN) and carbon and nitrogen stable isotope ratios (δ^{13}C and δ^{15}N). These parameters have been effectively used to trace the source of sediment and particulate matter (Fox and Papanicolaou, 2007; Laceby et al., 2015b; McConnachie and Petticrew, 2006). Soil δ^{15}N increases with depth in the soil profile (Amundson et al., 2003; Natelhoffer and Fry, 1988) whereas TOC and TN decrease (Blake et al., 2006; Owens et al., 2006). Tillage and harvesting also reduce TOC and TN concentrations, potentially allowing for cultivated sources to have intermediate concentrations relative to surface and subsoils (Juracek and Ziegler, 2009; Walling and Woodward, 1995). δ^{13}C discriminates between particulate material derived from landscapes with C_3 vegetation (e.g. the majority of tree or temperate grass species) compared to C_4 vegetation (e.g. several grass and cropping species predominantly found in warmer climates) (Fry, 2006; Mariotti and Peterschmitt, 1994; Schimel, 1993). The combination of these organic matter parameters will be used in three contaminated coastal catchments of the Fukushima Prefecture to investigate the potential of forest sources to supply contaminated matter downstream to recently decontaminated landscapes or landscapes less contaminated by the FDNPP accident.

2. Material and methods

2.1. Study site

Sampling occurred in the Mano (171 km^2), Niida (265 km^2) and Ota (77 km^2) catchments (Fig. 1). The main relief features of these catchments include an upstream coastal mountain range and a more densely populated coastal plain. The upstream catchment areas have total radiocesium (^{134}Cs + ^{137}Cs) inventories ranging from approximately 20 kBq kg^{-1} to 150 kBq kg^{-1} compared to inventories of <20 kBq kg^{-1} on the coastal plain (Fig. 1) (Chartin et al., 2013). The mean land use for these catchments, classified with data the Biodiversity Center of Japan (BCJ (2016)) and satellite imagery from Google Earth (version 7.1.5.1557) is 75% forest (SD 3%), 16% cultivated (SD 1%), 5% rural farmland (SD 1%) and 2% urban (SD 1%) (Fig. 2). Coniferous forests comprise the majority of forests, and paddy fields are the main cultivated land use. Mean annual rainfall, for the region within 100 km from the FDNPP, is 1420 mm y^{-1} (Laceby et al., 2016) and the rainfall regime drives the downstream migration of radiocesium (Chartin et al., in review; Evrard et al., 2014).

2.2. Field sampling

In-stream samples of particle borne organic matter were obtained during four fieldwork campaigns between November 2012 and November 2014. Sampling occurred after each typhoon season in early November and after the spring snow melt, in May 2013. These time periods represent the major hydro-sedimentary events in this region. Deposited fine particulate material (n = 82) was sampled during each campaign at multiple sites (Fig. 1) with surface scrapes using a plastic spatula (e.g. the top 1 cm of deposited material). Each sample was comprised of ~10 subsamples (~5 g per subsample) of deposited particle borne organic matter collected along a 5 m river reach. For the remainder of the text, particle borne organic matter will be referred to as particulate matter. This material includes sediment and mineral-bound organic matter trapped in soil micro-aggregates released by soil erosion (Lützow et al., 2006; Mikutta et al., 2006; Remusat et al., 2012), adsorbed or occluded in clay minerals (Vogel et al., 2014) and non-mineral bound organic matter (e.g. litterfalls, roots and other vegetation debris) (Feller and Beare, 1997; Gregorich et al., 2006).

Soil samples were collected from the dominant sources identified through a literature review (Evrard et al., 2015), multiple field campaigns, and stakeholder consultation. Sources included 46 forest soil samples (15 deciduous, 16 coniferous, and 15 mixed), 28 cultivated soil samples (14 rice paddy fields and 14 other fields), and 25 subsoil samples (15 decontaminated soil and 10 channel bank and subsoil erosion scars) (Fig. 2). A small plastic trowel was used to sample surface sources, compositing 10 sidewall scrape subsamples (top 1–2 cm, ~5 g) into one sample with decontaminated soils being sampled similarly to surface soils. Subsoil erosion scars were sampled by first scraping away the exposed surface sidewall of channel banks and hillslope erosion scars and then subsampling the manually exposed subsoil with 10 plastic trowel grabs that were composited into one sample.

2.3. Sample processing and analyses

Samples were dried in a ventilated oven at low temperature (40 °C) to avoid possible desorption of organic matter bound to clay minerals with swelling layers (Bailey, 1980; Remusat et al., 2012), thoroughly homogenized and dry-sieved to <2 mm to remove stones and coarse vegetation debris. A subsample of this material was wet-sieved to <63 μm with deionized water (Zirbser et al., 2001). The recovered <63 μm size fraction was oven-dried at 40 °C. In post FDNPP accident research, reports indicated that radiocesium activities were evident in both fine and coarse particle size fractions. For example, Tanaka et al. (2014) found that the silt and sand fractions contained 95% of the total ^{137}Cs activities in suspended sediment. Furthermore, radiocesium is contained within the tree canopy, leaf litter and soil organic matter in contaminated forests (Evrard et al., 2015; Kato et al., 2012; Loffredo