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An extensive study of the concentrations of particulate/dissolved radiocaesium derived from the Fukushima Dai-ichi Nuclear Power Plant accident in various river systems and their relationship with catchment inventory

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ABSTRACT

An extensive investigation of particulate radiocaesium in suspended solids and dissolved radiocaesium in river water was undertaken at 30 sites in Fukushima and Miyagi Prefectures in December 2012, and their relationships with catchment inventory and the solid/liquid distribution coefficient (K_d) were evaluated. Rivers located in the coastal region on the north side of the Fukushima Dai-ichi Nuclear Power Plant exhibited relatively higher particulate radiocaesium concentrations. Significant correlations were found between concentrations of particulate/dissolved radiocaesium and average catchment inventories, indicating that the concentrations of particulate/dissolved radiocaesium could be approximated from the catchment inventory. Particulate radiocaesium concentration was significantly correlated with dissolved radiocaesium concentration (with the exception of concentrations measured in estuaries), and the geometric mean K_d was calculated as 3.6×10^5 with a 95% confidence interval of $2.6–5.1 \times 10^5$.

1. Introduction

Researchers have reported the migration of radiocaesium derived from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident through river systems (Sakaguchi et al., 2012; Ueda et al., 2012; Nagao et al., 2013; Yamashiki et al., 2014). Yamashiki et al. (2014) estimated that 84–92% of total radiocaesium is transported in particulate form through the Abukuma River system, which is the largest river in Fukushima prefecture and was affected by radiocaesium deposition due to the FDNPP accident. Data about this migration have enabled predictions of changes in the contamination of land and downstream regions. Previous studies have reported the concentrations of particulate and dissolved radiocaesium in four river systems in Fukushima Prefecture (Ueda et al., 2012; Nagao et al., 2013), in which transport of radiocaesium was estimated based on data obtained by instantaneous water sampling. Fukushima Prefecture has many river systems with similar catchment characteristics and contamination levels including the Abukuma River, so an extensive investigation of radiocaesium in various river systems is needed.

To model the migration of radiocaesium through river systems, it is necessary to estimate radiocaesium concentrations in both particulate and dissolved forms. Matsunaga et al. (1998) suggested that particulate radiocaesium originating from the Chernobyl nuclear power plant accident was derived mainly from the erosion of surface soil. The concentration of radiocaesium in river water has been estimated using direct deposition to the ground for modelling of the dynamics of radiocaesium in the environment (Santschi et al., 1990; Smith et al., 2004, 2005). These studies indicate that a catchment inventory of radiocaesium is useful in determining the concentrations of radiocaesium in river water.

Suspended solids (SS) in river water display episodic characteristics including variations in concentration, particle size, and

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inputs of irregular matter, such as anthropogenic debris and terrestrial organisms, which can generate anomalous values in the evaluation of particulate radiocaesium. Additionally, it can be difficult to collect sufficient SS for analyses from low-turbidity water. Phillips et al. (2000) designed a time-integrated SS sampler that can remain under water for a certain duration and collect SS through sedimentation induced by ambient flow. They confirmed through field trials that the collected samples provided statistically representative values of the particle size and carbon content in ambient water based on weighted averages of SS concentration, indicating that the properties of SS during a flooding event are predominantly reflected in sediments trapped by the time-integrated SS sampler. Because this sampler can provide SS information that is time-integrated through the period of deployment and facilitates the collection of large amounts of SS (Laubel et al., 2002; Mizugaki et al., 2008), it has been widely used in investigations of the geochemical dynamics of SS, such as flux and fingerprinting studies to estimate the source of SS (Collins and Wallings, 2006; Walling et al., 2006; Fukuyama et al., 2010; Collins et al., 2010; Smith and Owens, in press). These characteristics make the sampler very useful for collecting and evaluating particulate radiocaesium at various sites, regardless of the SS concentration in the river water.

Radiocaesium migrates through river systems in both particulate and dissolved forms. Because each form plays a different role in the migration of radiocaesium in the water environment, solid/liquid partitioning of radiocaesium has been extensively investigated, particularly after the Chernobyl nuclear power plant accident (Ciffroy et al., 2003; Cha et al., 2006; IAEA, 2006 and references therein). The solid/liquid partitioning of radiocaesium is described in terms of a distribution coefficient (K_d). Although particulate/dissolved radiocaesium concentrations in field samples may not reach equilibrium and therefore provide an apparent K_d, the apparent K_d can represent actual solid/liquid partitioning in the field. Therefore, an evaluation of the apparent K_d of radiocaesium in river water provides crucially important information and can be used as a parameter in the prediction of future radiocaesium migration in water environments. The K_d is known to be affected by environmental factors such as the concentration of SS, mineral composition, ionic strength, and colloid aggregation (Aston and Duursma, 1973; Li et al., 1984a,b; Benes et al., 1992; Ciffroy et al., 2003; Cha et al., 2006; IAEA, 2006 and references therein; Nagao et al., 2013). Additionally, fine particle sizes can enhance radiocaesium concentrations in soil and sediment (Livens and Baxter, 1988; He and Walling, 1996), suggesting that the K_d is dependent on particle size composition. Coastal and inland regions of Fukushima Prefecture have different geologies, and many river systems have diverse hydrological characteristics and variations in water quality. Therefore, an extensive evaluation of the K_d is necessary to evaluate the solid/liquid partitioning of radiocaesium in river water affected by the FDNPP accident.

Because of the variations in geology, amounts of radiocaesium deposited, and catchment and river characteristics in Fukushima Prefecture, an extensive investigation was necessary to comprehensively evaluate the environmental dynamics of radiocaesium, in addition to the preliminary results previously reported at six monitoring sites (Yamashiki et al., 2014). This study investigated the concentrations of radiocaesium in SS in river water (hereafter particulate radiocaesium concentrations) and dissolved radiocaesium, as well as the relationships between radiocaesium concentrations and inventories in the river catchment area, with the goal of enabling estimations of the concentrations from inventories. Additionally, regional variation in the K_d was evaluated.

2. Materials and methods

2.1. Sampling and monitoring

To evaluate the K_d and the relationship between catchment inventory and particulate/dissolved radiocaesium concentrations, an extensive investigation was undertaken at the 30 sites shown in Fig. 1, during December 2012. The sites were located in the Abukuma River system (sites 1–6, 11–14, 16–22, 30), which runs through the centre of Fukushima Prefecture to Miyagi Prefecture, and in other river systems in coastal regions (sites 7–10, 15, 23–29). Table 1 lists the names of the sites, rivers, river systems, and the average radiocaesium inventories for the catchments corresponding to the numbers shown in Fig. 1. The average inventories in each catchment were obtained from the results of the Third Airborne Monitoring Survey by MEXT on 2 July 2011 (MEXT, 2011) excluding for the sites 18 and 19, because only 50–60% of the catchment areas of sites 18 and 19 are covered by the monitoring results while those of the other sites were covered more than 80%. The results of the Third Airborne Monitoring Survey were calibrated using soil core samples collected from 2200 points.

At each site, a sample of SS in river water was collected using a time-integrated SS sampler (Phillips et al., 2000), with the exception of Funaokaohashi (site 18) where the riverbed was too hard to fix the sampler. The samplers were installed from 4 to 8 December 2012, and kept *in situ* until 17–19 December 2012. The samplers were installed 0.2–5 m away from the edge of the riverbank and 5–20 cm above the riverbed, depending on the river conditions (i.e., width, depth, and structure of the riverbed) to keep the sampler submerged at base flow. A 20-L surface river water sample was also collected at each site, including Funaokaohashi (site 18), from 17 to 19 December 2012.

The SS samples were dried (105 °C, 24 h) and disaggregated using a mortar. Concentrations of particulate radiocaesium and particle size distributions were then measured. Each surface river water sample was filtered through a membrane filter (0.45-mm pore size, Millipore). Dissolved radiocaesium in the filtrate was concentrated by ammonium molybdophosphate (AMP), as described in Sakaguchi et al. (2012).

To monitor the concentration of SS in river water, turbidity was logged at 10-min intervals using a turbidity meter (ANALITE turbidity probe 3000-NTU, MacVan Instruments, USA) installed beside the SS sampler, and the output of the turbidity meter (mV) was converted into the SS concentration (mg L^{-1}).

2.2. Measurement of radiocaesium

Particulate radiocaesium in SS samples was measured using a high purity n–type Ge detector (EGC25–195–R, Canberra–Eurisys, USA.). Gamma-ray emissions at energies of 604 keV (^{134}Cs) and 662 keV (^{137}Cs) were measured. The measurement system was calibrated using standard gamma sources with different sample heights as reported by Kato et al. (2012), and a correction for the self-absorption of gamma-rays was established. Analytical accuracy was certified via a World-Wide Proficiency Test (IAEA, 2007) using standard soil samples available from IAEA. The counting error in the measurement of radiocaesium activity in the SS samples was less than 10%. The particulate radiocaesium concentration was expressed as Bq kg^{-1} of dried SS.

The 604 keV (^{134}Cs) and 662 keV (^{137}Cs) in the AMP samples were measured using gamma-ray spectrometry, with a low-background well type Ge detector (EGPC 150 P16, EURISYS) installed at the Low Level Radioactivity Laboratory, Kanazawa University. The detection limit for ^{137}Cs was estimated as 0.004 Bq after 2 d of measurement. The spectrometer was calibrated with a