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Occurrence and partition ratios of radiocesium in an urban river during dry and wet weather after the 2011 nuclear accident in Fukushima

Michio Murakami [a, b, *], Nao Shibayama [c], Keisuke Sueki [d], Goro Mouri [a], Haechong O [e], Mihiro Nomura [f], Yukio Koibuchi [e], Taikan Oki [a]

[a] Institute of Industrial Science, The University of Tokyo, 4-6-1 Komaba, Meguro, Tokyo, 153-8505, Japan
[b] Department of Health Risk Communication, School of Medicine, Fukushima Medical University, 1 Hikarigaoka, Fukushima City, 960-1295, Japan
[c] Graduate School of Pure and Applied Sciences, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki, 305-8572, Japan
[d] Faculty of Pure and Applied Sciences, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki, 305-8572, Japan
[e] Graduate School of Frontier Sciences, The University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba, 277-8563, Japan
[f] Faculty of Arts and Sciences Programs in English at Komaba (PEAK), The International Program on Environmental Sciences, The University of Tokyo, 3-8-1 Komaba, Meguro, Tokyo, 153-8902, Japan

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A B S T R A C T

After the 2011 nuclear accident in Fukushima, radiocesium was released from the Fukushima Dai-ichi Nuclear Power Plant and contaminated waters in urban areas near Tokyo. By intensive field monitoring during 3 years, this study investigated the temporal trends and the occurrence of radiocesium during dry and wet weather, and analyzed the variations in radiocesium during rainfall events and factors controlling them. Concentrations of particulate radiocesium decreased rapidly from May 2012 to March 2013 and reached an equilibrium in 2014. Concentrations of particulate ^{137}Cs during wet weather were almost double those during dry weather in the same period. In contrast to the small variations in ^{137}Cs concentrations in the particulate phase on a suspended solids (SS) weight basis during events, those in the dissolved phase on a liquid-volume basis fluctuated greatly, resulting in variations in the partition coefficient (apparent K_d). The apparent K_d of ^{137}Cs during wet weather ranged from 30 000 to 150 000 L kg^{-1} and showed a significant negative correlation with SS concentrations during wet weather. Specific surface area in solids contributed to the variations in apparent K_d.

1. Introduction

After the Great East Japan Earthquake on 11 March 2011 and the subsequent nuclear accident in Fukushima, radionuclides such as radiocesium (^{134}Cs and ^{137}Cs) were released from the Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Plant. Radiocesium was deposited on surfaces during dry and wet weather (Gonze et al., 2014) and contaminated forests, fields, and urban areas, including local hot spots such as Kashiwa City, Chiba Prefecture, adjacent to Tokyo (Kinoshita et al., 2011). The radiocesium remained in surface soils and catchments, increasing ambient doses (Nuclear Regulation Authority, 2013). It has been gradually washed out into receiving waters (Yamashiki et al., 2014) and is still detected in both freshwater and sea fishes (Ministry of Health and Welfare, 2015). Although doses of radioiodine and radiocesium through dietary intake were very limited (Murakami and Oki, 2014), the consistent detection of radiocesium in fishes and suspension of the fish trade have caused economic damage to fisheries.

Since the 2011 accident, the behavior of radiocesium in waters has been investigated through intensive river monitoring in hydrological units ranging in size from small plots to large catchments, under normal and extreme flow conditions, over periods ranging from a few days to months or years (e.g., Ministry of Education Culture Sports Science and Technology, 2012b; Ministry of the Environment, 2015; Nuclear Regulation Authority, 2014; Tsuji et al., 2014; Ueda et al., 2013; Yamashiki et al., 2014; Yoshikawa et al., 2014). However, most studies have focused

* Corresponding author. Department of Health Risk Communication, School of Medicine, Fukushima Medical University, 1 Hikarigaoka, Fukushima City, 960-1295, Japan.
E-mail address: michio@fmu.ac.jp (M. Murakami).

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mainly on the wash-off of ^{137}Cs from forest and farmland, because Fukushima Prefecture is mostly rural (71% forest (Forestry Agency, 2013), 10.5% cultivated (Ministry of Internal Affairs and Communications, 2013), in 2012). In urban areas, previous studies focused on deposition amounts (Amano et al., 2012), regional variations and temporal changes in ambient radioactivity levels (Erlandsson and Isaksson, 2006), and removal efficiencies in water and wastewater treatment plants (Kosaka et al., 2011; Tsuhima et al., 2013) following both the 1986 accident in Chernobyl and the 2011 accident in Fukushima. Few studies, however, intensively investigated the wash-off behavior of radiocesium in urban rivers. Yamashita et al. (2015) and Koibuchi et al. (2015) reported the temporal variations, sources, stocks, and fluxes of radiocesium in an urban river, the Ohori River, which runs through Kashiwa City. The 1-year field sampling study (Yamashita et al., 2015) showed that radiocesium concentrations in the particulate phase were higher during wet weather than during dry weather on both SS weight and liquid volume bases, and that the partition ratios, apparent K_d (the ratio of radiocesium concentration in the particulate phase on a suspended solids [SS] weight basis to that in the dissolved phase on a liquid-volume basis) of radiocesium differed between wet and dry weather, although only four rainfall events were sampled. But only a very limited number of investigations have been based on dense monitoring of radiocesium concentrations and apparent K_d during wet weather. Therefore, intensive investigations of the effect of rainfall on radiocesium concentrations are required. In addition, although time-integrated SS samplers (Phillips et al., 2000) have proved useful in collecting composite samples (through their preferential collection of SS during wet weather owing to the higher efficiency of sampling of SS under those conditions; i.e., higher [SS] and flow velocity (Koga et al., 2004; Osanai et al., 2005)), their characteristics should be investigated by comparison of radiocesium concentration between grab samples and composite samples. Furthermore, variations in apparent K_d and factors responsible for the variations during wet weather should be fully investigated, because most fluxes of radiocesium in water are washed out during wet weather (Yamashiki et al., 2014).

The objectives of this study were fourfold. First, through intensive investigations of radiocesium in the Ohori River during 23 rainfall events over 3 years, in addition to dry-weather grab samples and composite samples, this study investigated radiocesium concentrations over time. Although radiocesium fluxes are also important to understand the behavior, the discussion will be reported in a further study. Second, it investigated the differences in concentrations between wet and dry weather. Third, it compared concentrations between grab samples and composite samples to assess the characteristics of the SS sampler. Fourth, it analyzed the variations in concentrations in the particulate and dissolved phases during rainfall events, and factors related to the variations in apparent K_d.

2. Materials and methods

2.1. Sample collection

The protocol used for the first stage of the investigations of radiocesium (May 2012–March 2013) (Yamashita et al., 2015) was used for the second stage also (April 2013–January 2015). The river water was collected at Showa Bridge (st. d, WGS84: 35.872°N 139.969°E; identified as the Ohori River in Yamashita et al. (2015)) (Fig. 1). The Ohori River is located in Kashiwa City, Chiba Prefecture. Kashiwa City has a total area of 114.9 km^2 and had a population of 409 043 in March 2015 (Kashiwa city, 2015), and is serviced mainly by a separate sewer system. The catchment area of the Ohori River is 31 km^2 and the average river discharge downstream is 1 m^3 s^{-1}

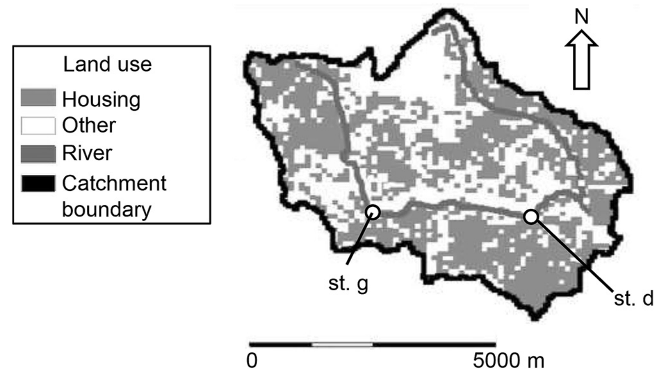

Fig. 1. Sampling locations in the Ohori River catchment. St. d is the sampling station for river water samples. St. g is the location for water supply from the Tone River during dry weather.

(Nihei et al., 2007). During dry weather, 0.5 m^3 s^{-1} of water is supplied to the Ohori River from the Tone River (st. g) upstream of this study's sampling point at st. d. The land use is predominantly residential (53.2% housing, 1.5% roads, in 1997 (Geographical Survey Institute, 1997)). The average deposition of radiocesium (sum of ^{134}Cs and ^{137}Cs) in the catchment of the Ohori River was 61 000 Bq/m^2 in June 2012 (Koibuchi et al., 2015) according to an airborne monitoring survey (Ministry of Education Culture Sports Science and Technology, 2012a).

Grab samples and composite samples were collected from May 2012. From the second stage of this survey (April 2013–January 2015), together with the collection of grab samples during dry weather and composite samples, time-series of grab samples during wet weather (up to 8 samples per event) were collected with an automatic sampler (Teledyne-Isco, USA). In total, 44 dry-weather grab samples, 122 wet-weather grab samples (23 events), and 39 composite samples were collected from May 2012 to January 2015 (Tables S1–S3 in Supplementary material). The water depth and electric conductivity (EC) were monitored on site with a 750 Area Velocity Flow Module (Teledyne-Isco) and a water quality multiprobe (Data Sonde 5, Hydrolab, USA), respectively.

The samples were filtered through prebaked glass fiber filters (GF/F, 0.7 μm pore size; Whatman, UK) for consistency with the previous study (Yamashita et al., 2015). Suspended solids concentration [SS] was determined from the filtrate volume and the differences in dried filter weights before and after filtration. Filtrates were further filtered through membrane filters (0.2 μm; Advantec, Japan) to obtain the dissolved phase. The 0.2 μm membrane filters were also used for consistency with the previous study (Yamashita et al., 2015).

2.2. Radiocesium analysis (Yamashita et al., 2015)

The glass fiber filters were placed in a polyethylene bag, fixed on an acrylic plate, and then analyzed by germanium semiconductor detector (GEM-30195, Ortec, USA; relative detection efficiency at 1332 keV, 32.1%) and multi-channel analyzer (MCA 7600, Seiko EG&G Co., Ltd., Japan). Filtrate samples (8 time-series samples collected on 26 June 2013 and the first sample collected in each event on 27 June, 27 July, 20, 27, and 29 August, 4 and 8 September, and 25 October 2013; Tables S1 and S2) were injected into a 2-L Marinelli beaker and analyzed by germanium semiconductor detector (GC4518, Canberra, USA; relative detection efficiency at 1332 keV, 52.5%) and multi-channel analyzer (DSA 1000, Canberra). Gamma-ray emissions from ^{134}Cs and ^{137}Cs at energies of 0.1–2.0 MeV were counted. The spectra obtained were analyzed by