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

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28 **Abstract**

29 After the 2011 nuclear accident in Fukushima, radiocesium was released from the Fukushima
30 Dai-ichi Nuclear Power Plant and contaminated waters in urban areas near Tokyo. By
31 intensive field monitoring during 3 years, this study investigated the temporal trends and the
32 occurrence of radiocesium during dry and wet weather, and analyzed the variations in
33 radiocesium during rainfall events and factors controlling them. Concentrations of particulate
34 radiocesium decreased rapidly from May 2012 to March 2013 and reached an equilibrium in
35 2014. Concentrations of particulate ^{137}Cs during wet weather were almost double those during
36 dry weather in the same period. In contrast to the small variations in ^{137}Cs concentrations in
37 the particulate phase on a suspended solids (SS) weight basis during events, those in the
38 dissolved phase on a liquid-volume basis fluctuated greatly, resulting in variations in the
39 partition coefficient (apparent K_d). The apparent K_d of ^{137}Cs during wet weather ranged from
40 30 000 to 150 000 L kg^{-1} and showed a significant negative correlation with SS
41 concentrations during wet weather. Specific surface area in solids contributed to the variations
42 in apparent K_d .

43

44 **Keywords:**

45 Coefficient of partition, radionuclides, rainfall, specific surface areas, temporal trend, urban
46 areas

47

48 **1. Introduction**

49 After the Great East Japan Earthquake on 11 March 2011 and the subsequent nuclear accident
50 in Fukushima, radionuclides such as radiocesium (^{134}Cs and ^{137}Cs) were released from the
51 Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Plant. Radiocesium

52 was deposited on surfaces during dry and wet weather (Gonze et al., 2014) and contaminated
53 forests, fields, and urban areas, including local hot spots such as Kashiwa City, Chiba
54 Prefecture, adjacent to Tokyo (Kinoshita et al., 2011). The radiocesium remained in surface
55 soils and catchments, increasing ambient doses (Nuclear Regulation Authority, 2013). It has
56 been gradually washed out into receiving waters (Yamashiki et al., 2014) and is still detected
57 in both freshwater and sea fishes (Ministry of Health and Welfare, 2015). Although doses of
58 radioiodine and radiocesium through dietary intake were very limited (Murakami and Oki,
59 2014), the consistent detection of radiocesium in fishes and suspension of the fish trade have
60 caused economic damage to fisheries.

61 Since the 2011 accident, the behavior of radiocesium in waters has been investigated
62 through intensive river monitoring in hydrological units ranging in size from small plots to
63 large catchments, under normal and extreme flow conditions, over periods ranging from a few
64 days to months or years (e.g., Ministry of Education Culture Sports Science and Technology,
65 2012b; Ministry of the Environment, 2015; Nuclear Regulation Authority, 2014; Tsuji et al.,
66 2014; Ueda et al., 2013; Yamashiki et al., 2014; Yoshikawa et al., 2014). However, most
67 studies have focused mainly on the wash-off of ^{137}Cs from forest and farmland, because
68 Fukushima Prefecture is mostly rural (71% forest (Forestry Agency, 2013), 10.5% cultivated
69 (Ministry of Internal Affairs and Communications, 2013), in 2012). In urban areas, previous
70 studies focused on deposition amounts (Amano et al., 2012), regional variations and temporal
71 changes in ambient radioactivity levels (Erlandsson and Isaksson, 2006), and removal
72 efficiencies in water and wastewater treatment plants (Kosaka et al., 2011; Tsumura et al.,
73 2013) following both the 1986 accident in Chernobyl and the 2011 accident in Fukushima.
74 Few studies, however, intensively investigated the wash-off behavior of radiocesium in urban
75 rivers. Yamashita et al. (2015) and Koibuchi et al. (2015) reported the temporal variations,
76 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 km² 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 km² and the average river discharge downstream is 1 m³ s⁻¹ (Nihei et al., 2007). During dry weather, 0.5 m³ 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 ¹³⁴Cs and ¹³⁷Cs) in the catchment of the Ohori River was 61 000 Bq/m² 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 multi-probe (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, 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 Gamma Studio software (Seiko EG&G Co.) or Spectrum Explorer software (Canberra). The detection limit for the filters was 0.06 Bq measured over 80 000 s, which corresponded to 600 Bq kg-SS⁻¹ for 100-mg samples. The detection limit for the filtrate was 25 mBq L⁻¹ measured over 250 000 s. The concentrations of radiocesium were calculated on both an SS-weight basis (Bq kg-SS⁻¹) and a liquid volume basis (Bq L⁻¹) for the particulate phase, and on a volume basis for the dissolved phase. The radioactivity was decay-corrected to the collection date.

2.3. Specific surface area analysis

The radiocesium concentration in the particulate phase depends on specific surface area (He and Walling, 1996; Tanaka et al., 2015). To remove variations in the apparent K_d due to specific surface area, concentrations were corrected for surface area. Particle size distributions in samples collected during wet weather in 2013 (the first sample collected in each event on 20 and 27 August, 4 and 8 September, and 25 October) were analyzed by laser diffraction particle size analyzer (SALD-3000S, Shimadzu, Japan), and distributions of 8 time-series samples collected on 26 June and the first sample collected in an event on 27 July (Yamashita et al., 2015) were also used (Table S2). Specific surface areas were then estimated on the assumptions that the particles have a uniform spherical form and their density is 2.6 g cm^{-3} . The radiocesium concentrations in the particulate phase were corrected as follows (He and Walling, 1996; Ministry of Education Culture Sports Science and Technology, 2012b):

$$^{137}\text{Cs}' = ^{137}\text{Cs}/(S_{\text{sample}}/S_{\text{ref}})^{\nu}$$

where $^{137}\text{Cs}'$ is the concentration corrected for surface area, ^{137}Cs is the uncorrected concentration, S is the specific surface area of the sample or reference (ref), and $\nu = 0.65$, which was estimated from an adsorption test with a cultivated soil in the UK (He and Walling, 1996). A river water sample collected on 15 September 2013 during wet weather was regarded as the reference sample because of high [SS]. Similar values for ν in situ were found in Fukushima (0.79 for a river bed sediment; 0.76 for a pasture soil) (Japan Atomic Energy Agency, 2013).

3. Results and discussion

3.1. Temporal trends and occurrence of ^{137}Cs during wet and dry weather

The particulate concentrations of radiocesium were $7000 \text{ Bq kg-SS}^{-1}$ ^{137}Cs and $9600 \text{ Bq kg-SS}^{-1}$ ^{137}Cs in the grab sample collected during dry weather in May 2012 (Fig. 2). They

decreased sharply to 13% and 16% of those values in March 2013, increased again in summer 2013, and then gradually reduced to an equilibrium ($\sim 3000 \text{ Bq kg-SS}^{-1} \text{ }^{137}\text{Cs}$). $[^{137}\text{Cs}]$ was expressed as an exponential function of elapsed time (see Fig. 2 caption). Estimated decay rates (including physical decay) based on the whole period were 0.00067 d^{-1} for dry-weather grab samples, 0.0012 d^{-1} for wet-weather grab samples, and 0.0011 d^{-1} for composite samples. Estimated decay rates based on the first stage were 0.0058 d^{-1} , 0.0052 d^{-1} , and 0.0047 d^{-1} , respectively. Grab samples collected during wet weather and composite samples showed similar decay rates and trends (Fig. 2). Although the shorter half-life of ^{134}Cs (2.06 y) than that of ^{137}Cs (30 y; (ICRP, 1983)) contributed to its slightly faster rate of reduction, most of the decrease might be attributable to dilution with less-contaminated soils newly generated in the catchment. The increase during summer 2013 might not be related to seasonal changes, because similar trends were not found during summer 2014, despite similar precipitation between the two years (i.e., 302.5 mm in July to September 2013; 286.0 mm in July to September 2014 at Abiko City (Japan Meteorological Agency, 2015)). Further investigations are necessary to identify the reason for such variation..

To compare the concentrations of radiocesium in the particulate phase on an SS-weight basis among samples, the ratios of ^{137}Cs concentrations in grab samples to those in composite samples in the same period were estimated. The arithmetic mean ratio of wet-weather grab samples to composite samples was 2.0 (95% confidential interval, 1.6–2.3); that of dry-weather grab samples to composite samples was 0.91 (0.71–1.1) (Fig. 3). Thus, radiocesium concentrations during wet weather were double those during dry weather. This intensive monitoring by 3-year field sampling supports the previous 1-year result of higher radiocesium concentrations during wet weather than during dry weather (Yamashita et al., 2015). ^{137}Cs concentrations in composite samples were comparable to or slightly higher than those in dry-weather grab samples, even though the SS sampler preferentially collects coarser particles

(Phillips et al., 2000), which hold less radiocesium than finer particles (He and Walling, 1996; Japan Atomic Energy Agency, 2013; Tanaka et al., 2015). This can be explained by the facts that composite sampling favors SS during wet weather and that ^{137}Cs concentrations during wet weather were higher than those during dry weather. Composite samples can be used to estimate the ^{137}Cs flux (e.g., Yamashiki (2014)), because the SS sampler can collect large amounts of SS, which can partially reflect ^{137}Cs concentrations during wet weather. However, since ^{137}Cs concentrations in wet-weather grab samples were double those in composite samples, the use of the ^{137}Cs concentration in composite samples possibly underestimates the flux of ^{137}Cs in urban rivers.

3.2. Variations in [^{137}Cs] during rainfall events and factors related to apparent K_d

Figure 4 shows representative results of variations in water depth, [SS], particulate [^{137}Cs], dissolved [^{137}Cs], and apparent K_d during a rainfall event on 26 June 2013 and during dry weather the following day. Whereas [SS] and dissolved [^{137}Cs] reduced with time and with falling water depth (relative standard deviation [RSD] during the event, 47% for SS, 33% for dissolved ^{137}Cs), particulate ^{137}Cs varied little (RSD, 9%). Other events also showed small variations in particulate ^{137}Cs , with an arithmetic mean RSD of 15% (Fig. 5). The apparent K_d of ^{137}Cs during the event on 26 June varied by a factor of 2.5 (RSD, 38%). Although a constant value of apparent K_d is often used in model simulations (e.g., Ishikawa et al., 2014; Lake Biwa Environmental Research Institute, 2013) which represent the wash-off behavior of ^{137}Cs in the particulate and dissolved phases separately, these observations suggest that the variations in apparent K_d during wet weather are not negligible.

The particulate distribution of [^{137}Cs] increased with [SS] (Fig. S1). It ranged from 10% to 58% during dry weather and from 59% to 93% during wet weather. The dominant distribution of particulate radiocesium during wet weather is similar to findings in Fukushima (Ueda et al., 2013). Since most fluxes of radiocesium in waters are washed out during wet

weather (Yamashiki et al., 2014), it is important to understand the factors related to the variations in apparent K_d during wet weather. The apparent K_d of radiocesium depends on characteristics of solids (e.g., specific surface area) (He and Walling, 1996; Japan Atomic Energy Agency, 2013; Tanaka et al., 2015) and competing ions in the water (e.g., EC as an indicator) (Wilflinger et al., 2005). The apparent K_d of ^{137}Cs during wet weather (30 000–150 000 L kg^{-1}) was significantly negatively correlated with [SS] ($P < 0.05$; Fig. 6), but not with EC ($P > 0.10$). Thus, regression against [SS] is useful to correct the apparent K_d and therefore to estimate particulate and dissolved ^{137}Cs separately in model simulations. The absence of a correlation in dry weather might be attributable to the difference in the characteristics of solids in the water owing to water input from the Tone River only during dry weather.

The negative correlation of apparent K_d with [SS] during wet weather could be explained by the increase in particle size with increasing [SS] during heavy rain (Shigeta et al., 2008). Without correction for surface area, the relationship between dissolved [^{137}Cs] (on a liquid volume basis) and particulate [^{137}Cs] (on an SS-weight basis) was weak (Fig. 7a, $r = 0.34$, $P > 0.10$, slope for log–log plots = 0.19); with correction, it strengthened (Fig. 7b, $r = 0.53$, $P = 0.05$, slope for log–log plots = 0.46). The slope for log–log plots also became closer to 1 after the correction for surface area, also indicating the improvement of linearity ($Y = aX^b \Leftrightarrow \log Y = b \log X + \log a$). Therefore, the difference in specific surface areas of solids during events contributed to the variation in the apparent K_d of ^{137}Cs during wet weather. Since the relationship between dissolved and particulate [^{137}Cs] was still weak even after correction for surface area, other factors (such as competing ions or other soil characteristics) might influence the variations in the apparent K_d . Another possible reason is that the ν value was based on the data from cultivated soil in the UK, which might differ from the SS in the Ohori River. Further studies will be necessary to estimate ν and to identify the factors influencing

the variations in the apparent K_d . Nevertheless, this study highlights that SS concentrations are useful for correcting the apparent K_d of radiocesium during wet weather, and that specific surface area plays an important role in the variations in the apparent K_d .

4. Conclusions

Concentrations of radiocesium were investigated by intensive field monitoring over 3 years in an urban river during wet and dry weather. Variations in the concentrations and apparent K_d of radiocesium during rainfall events and contributing factors were analyzed.

1. The concentrations of particulate radiocesium decreased rapidly from 9600 Bq kg-SS⁻¹ ¹³⁷Cs in dry-weather grab samples in May 2012 to 16% of that level in March 2013 and reached an equilibrium (~3000 Bq kg-SS⁻¹) in 2014.
2. The concentrations of particulate radiocesium in wet-weather grab samples were higher than those in dry-weather grab samples and in composite samples. The ratio of ¹³⁷Cs in wet-weather grab samples to composite samples was 2.0 (95% confidential interval, 1.6–2.3). That of dry-weather grab samples to composite samples was 0.91 (0.71–1.1). The SS sampler possibly underestimates average concentrations, and thus fluxes of radiocesium in urban rivers.
3. Whereas radiocesium concentrations in the dissolved phase on a liquid-volume basis fluctuated during rainfall events (e.g., RSD = 33% during an event on 26 June 2013), those in the particulate phase on an SS weight basis were almost constant (e.g., 9%).
4. The apparent K_d of radiocesium was negatively correlated with SS concentrations during wet weather. The use of SS concentrations could therefore be used to correct the apparent K_d of radiocesium in model simulations.
5. The specific surface area of solids influenced the variations in the apparent K_d of radiocesium. Correction for the specific surface area strengthened linearity between dissolved [¹³⁷Cs] on a liquid-volume basis and particulate [¹³⁷Cs] on an SS-weight basis

(i.e., the slope of log–log plots was 0.19–0.46; $r = 0.34$ –0.53).

Acknowledgements

This study was supported by a CREST grant from the Japan Science and Technology Agency and "Wisdom of Water" (Suntory), Corporate Sponsored Research Program, Organization for Interdisciplinary Research Projects, The University of Tokyo.

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399

400 **Figure captions**

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

403 Fig. 2. Concentrations of (a) ^{134}Cs and (b) ^{137}Cs in grab and composite samples. The first
404 stage data come from a previous study (Yamashita et al., 2015). Concentrations were not
405 corrected for surface area. Median values are plotted for wet-weather grab samples.

406 Precipitation was monitored at Abiko City (Japan Meteorological Agency, 2015). [^{137}Cs] (Y)
407 was expressed as exponential functions of the elapsed days (X , where $X = 0$ on 11 May 2012).

408 For the whole period: dry-weather grab samples, $Y = 4800e^{-0.00067X}$, $r^2 = 0.11$; wet-weather
409 grab samples, $Y = 11000e^{-0.0012X}$, $r^2 = 0.44$; composite samples, $Y = 7000e^{-0.0011X}$, $r^2 = 0.41$.

410 For the first stage: dry-weather grab samples, $Y = 9800e^{-0.0058X}$, $r^2 = 0.78$; wet-weather grab
411 samples, $Y = 27000e^{-0.0052X}$, $r^2 = 0.87$; composite samples, $Y = 12000e^{-0.0047X}$, $r^2 = 0.75$.

412 Fig. 3. Comparison of ^{137}Cs between grab and composite samples. The first stage data come
413 from a previous study (Yamashita et al., 2015). Concentrations were not corrected for surface
414 area. Arithmetic means of concentrations in grab samples during the collection period of the
415 SS sampler were plotted.

416 Fig. 4. Variations in water depth, [SS], dissolved and particulate [^{137}Cs], and apparent K_d of
417 ^{137}Cs during a rainfall event on 26 June 2013 and during dry weather the following day.

418 Radiocesium concentrations were not corrected for surface area.

419 Fig. 5. Variations in particulate [^{137}Cs] during rainfall events ($n = 8$ samples in each event).

420 Concentrations were not corrected for surface area. RSD, relative standard deviation.

421 Fig. 6. Relation between [SS] and apparent K_d of ^{137}Cs . Apparent K_d was not corrected for
422 surface area. For wet weather samples, $r = -0.51$, $\log Y = -0.0014X + 5.00$, $P < 0.05$. The

423 first stage data come from a previous study (Yamashita et al., 2015).

424 Fig. 7. Relation between dissolved and particulate [^{137}Cs] in grab samples during wet weather

425 in 2013. (a) Without correction for surface area; $r = 0.34$, $\log Y = 0.19\log X + 3.47$, $P > 0.10$.

426 (b) With correction for surface area; $r = 0.53$, $\log Y = 0.46\log X + 2.81$, $P = 0.05$.

Fig. 1.

(a)

(b)

Fig. 2.

Fig. 4.

Fig. 5.

Fig. 6.

Fig. 7.