Temporal Variation and Source Analysis of Radiocesium in an Urban River after the 2011 Nuclear Accident in Fukushima, Japan

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ABSTRACT
The release of radiocesium from the Fukushima Dai-ichi Nuclear Power Plant caused environmental contamination. We analyzed sources, behavior, and temporal trends of radiocesium by 1-year monitoring in an urban river, the Ohori River. The concentrations of 137Cs in both particulate and dissolved phases decreased to ~ 16% within 1 year. The partition coefficient in dry weather was estimated to be 3.2 × 10^4 L/kg. 137Cs concentrations in the particulate phase were higher during wet weather than during dry weather on the basis of both suspended solids (SS) weight and liquid volume. The ratios of 137Cs concentration to deposited 137Cs in the Ohori River (e.g., composite: 0.26 m^2/kg-SS in June – July 2012) were higher than those in the Abukuma River, probably because of differences in land use. Source analysis by a chemical mass balance method showed that 22% of 137Cs came from river water in dry weather, 39% from river sediments, and 39% from road dust highlighting urban surface deposits such as road dust as major sources of 137Cs in the river water even 2 years after the accident, and that the wash-off of these deposits increased the 137Cs concentration in the river during wet weather.

Keywords: partition coefficient, radionuclides, urban road dust

INTRODUCTION
Radionuclides, including radiocesium (134Cs and 137Cs), released from the Tokyo Electric Power Company’s the Fukushima Dai-ichi Nuclear Power Plant following the earthquake and tsunami of 11 March 2011 have been deposited on surfaces mainly through rainfall and discharged into waters. Local hot-spots of accumulated radioactive contamination have been found in urban areas, including Kashiwa city, Chiba prefecture (MEXT, 2011). Around Kashiwa, which has a total area of 114.9 km^2 and a population of 406,973 (http://www.city.kashiwa.lg.jp/; April 2014), the major deposition occurred on 21 March 2011 (Kinoshita et al., 2011). Subsequently, high radiocesium concentrations (134Cs and 137Cs; 4,300 and 5,400 Bq/kg) have been detected in the sediment of the Ohori River, which runs through Kashiwa (MOE, 2011). The uptake and accumulation of radiocesium by fish are well documented (Rowan and Rasmussen,
1994), and the trade in several fishes (e.g., *Carassius auratus langsdorffii*) of the Ohori River has been suspended. In addition, radiocesium deposited on the ground increases ambient radiation levels. Therefore, understanding of the sources, behavior (e.g., distribution of dissolved and particulate concentrations), and long-term trends of radionuclides in freshwaters is necessary in order to take appropriate remediation and regulatory actions for minimizing exposure to radiation.

Research has been directed toward investigating and modeling the behavior and long-term trends of radionuclides following the 1986 accident in Chernobyl (Smith *et al*., 2002; Monte *et al*., 2004) and the 2011 accident in Fukushima (Sakaguchi *et al*., 2012; Nagao *et al*., 2013; Ueda *et al*., 2013; Mouri *et al*., 2014; Yamashiki *et al*., 2014). This research shows an initial fast increase in $^{137}$Cs concentrations in lake waters as a result of rapid wash-off, a slow decline as a result of soil fixation and redistribution, and a very long-term “equilibrium” situation, which were linked to concentrations in fish (Smith *et al*., 2005). The wash-off of radiocesium from forests and farmlands has also been well investigated (Bonnett, 1990; Teramage *et al*., 2014; Yoshikawa *et al*., 2014; Yoshimura *et al*., 2014). In contrast, although regional and temporal variations in ambient radioactivity levels in urban areas are available (Mueck and Steger, 1991; Erlandsson and Isaksson, 2006; NRA, 2014), research investigating runoff into urban rivers is surprisingly limited following both accidents and is thus required.

By 1-year field sampling, we analyzed sources, behavior, and temporal trends of radiocesium in the Ohori River. First, we evaluated the distributions of dissolved and particulate radiocesium, the partition coefficient ($K_d$), and the temporal trend of radiocesium concentrations. Second, we investigated the impact of rainfall on radiocesium concentrations, collecting grab samples in wet and dry weather and composite samples using a suspended solids (SS) sampler. Third, we evaluated river sediments, road dust, and road runoff in wet weather as sources of radiocesium in the river, which we identified by using source-specific hydrophobic markers of sewage.

**MATERIALS AND METHODS**

**Sample collection**

The Ohori River is located in Kashiwa city, Chiba prefecture (Fig. 1a). The catchment area is 31 km$^2$ and the average river discharge downstream is 1 m$^3$/s (Nihei *et al*., 2007). The land use is predominantly residential (housing, 53.2%; roads, 1.5% in 1997 (GSI, 1997)) (Fig 1b), unlike that in Fukushima prefecture, where 71% is forest in 2012 (Forestry Agency, 2013). During dry weather, 0.5 m$^3$/s of water was supplied to the Ohori River at st. g from the Tone River.

Temporal variations in radiocesium concentrations were investigated at Showa Bridge (st. d) from May 2012. This period corresponded to the start of the second phase of the survey of Smith *et al*. (2005). Grab and composite samples were collected every 2 to 3 weeks. Grab samples were also collected during wet weather. Composite samples were collected with an SS sampler, which is designed to collect time-integrated SS samples (Phillips *et al*., 2000). Collection rates are higher at higher flow rates (Koga *et al*., 2004), so the sampler preferentially stores SS samples collected during wet weather.
Fig. 1 - Sampling locations. (a) Location of the Ohori River in Kashiwa city. (b) Sampling locations in the Ohori River catchment. The river water samples were collected at st. d (WGS84, 35.872 N 139.969 E). The water was supplied to the Ohori River at st. g from the Tone River during dry weather.

Samples of potential sources were collected in the catchment. A water sample from the Tone River water transmission point was collected at st. g on 18 July 2012. River sediment samples were collected at st. a – f by Ekman-dredge sampler or by shovel on 3 October 2012, and the surface 2 cm or so was analyzed. Road dust was collected at st. 1 – 6 by sweeping the gutter with a pig-bristle brush on 17 October 2012. Road runoff was collected from the gutter at st. 7 on 19 September, 4 December 2012, and 6 February 2013.

Grab, composite, and road runoff samples were filtered through prebaked glass fiber filters (GF/F, pore size, 0.7 µm; Whatman, UK) to be consistent with the previous study (Isobe et al., 2002). SS concentrations were estimated from the filtrate volumes and the differences in dried filter weights between before and after filtration. Grab samples were further filtered through membrane filters (0.2 µm; Advantec, Japan) to obtain the dissolved phase. The solid samples (glass fiber filters, sediments, and road dust) were freeze-dried and stored at –30°C before analysis.

Radiocesium analysis
The glass fiber filters were placed in a polyethylene bag, which was fixed on an acrylic plate, and the other solid samples were injected into a stainless steel container. Radiocesium in the glass fiber filters and other solid samples was analyzed by germanium semiconductor detector (GEM-30195, Ortec, US; relative detection efficiency at 1,332 keV: 32.1%) and multi-channel analyzer (MCA 7600, Seiko EG&G Co., Ltd., Japan). The filtrate samples were injected into a 2-L Marinelli beaker and were measured by germanium semiconductor detector (GC4518, Canberra, USA; relative detection efficiency at 1,332 keV: 52.5%) and multi-channel analyzer (DSA 1000, Canberra, US). Gamma-ray emissions from $^{134}\text{Cs}$ and $^{137}\text{Cs}$ at energies of 0.1 – 2.0 MeV were counted. The spectra obtained were analyzed by Gamma Studio software (Seiko EG&G Co. Ltd., Japan) or Spectrum Explorer software (Canberra, US). The
The accuracy of analysis of the glass fiber filter and filtrate samples was calibrated against $^{134}$Cs and $^{137}$Cs standard solutions (Japan Radioisotope Association). The accuracy of analysis of the other solid samples was calibrated against U8-type radioactivity standard volume sources (Japan Radioisotope Association). The detection limit for glass fiber filters was 0.06 Bq measured over 80,000 s, which corresponded to 600 Bq/kg-SS for 100-mg samples. The detection limit for the other solid samples was 3 Bq measured over 3,000 s, which corresponded to 60 Bq/kg-SS for 50 g-samples. The detection limit for the filtrate was 25 mBq/L measured over 250,000 s.

The concentrations of radiocesium were calculated on both an SS-weight basis (Bq/kg-SS) and a liquid volume basis (Bq/L) for solid samples, and on a volume basis for the dissolved phase of river water samples. The radioactivity was decay-corrected for the collection date.

**PPCP and sterol analysis**

We analyzed two hydrophobic pharmaceutical and personal care products (PPCPs)—methyltriclosan and triclosan (with log octanol–water partition coefficients [log $K_{OW}$] of $\sim$ 4.76 (SRC, 2014)—and ten sterols—$5\beta$-cholestanone, coprostanone; $5\beta$-cholestan-3$\beta$-ol, coprostanol; $5\beta$-cholestan-3$\alpha$-ol, epicoprostanol; cholest-5-en-3$\beta$-ol, cholesterol; $5\beta$-cholestan-3$\beta$-ol, cholestanol; 24-methylcholest-5-en-3$\beta$-ol, campesterol; 24-ethylcholesta-5,22$E$-dien-3$\beta$-ol, stigmasterol; 24-ethylcholest-5-en-3$\beta$-ol, $\beta$-sitosterol; $\beta$-sitosterol; (3$\beta$,24$E$)-stigmasta-5,24(28)-dien-3-ol, fucosterol; 24-ethyl-5$\alpha$-cholestan-3$\beta$-ol, stigmastanol (log $K_{OW}$, 8.74 – 9.65 (SRC, 2014))—as sewage markers (Isobe et al., 2002; Nakada et al., 2008).

PPCPs and sterols were analyzed as described by Isobe et al. (2002) with a slight modification to extraction. PPCPs and sterols in solid samples (~ 1 g) were extracted by pressurized solvent extraction (ASE 200, Dionex, US) with acetone and dichloromethane (1:3, v/v) at 175°C and 10.5 MPa with a flush percentage of 100% in an 11-mL cell by static extraction for 5 min. Each sample was extracted twice and the extracts were combined. Surrogates ($^{13}$C$_{12}$-methyltriclosan, d$_3$-triclosan, d$_6$-cholesterol) were spiked into the extracts, which were then purified by 5% H$_2$O deactivated silica gel column chromatography. The first fraction containing polycyclic aromatic hydrocarbons and alkanes was eluted with dichloromethane and hexane (1:3, v/v) and then discarded. The second fraction, in dichloromethane, was collected, and the PPCPs and sterols were acetylated with pyridine and acetic anhydride. Concentrations were determined by gas chromatography – mass spectrometry (GC-MS; Hewlett Packard GC system HP 6890 series, Mass Selective 5973 Detector, US) equipped with an Agilent Technologies HP-5MS column (30 m × 0.250 mm i.d.; 0.25 µm film thickness, US).

The reproducibility was confirmed by using three aliquots of extracts from a sediment. Recovery rates were confirmed by spiking of standards into four aliquots of extracts. Relative standard deviations of individual analytes were < 10% and the recovery rates ranged from 81% to 121%. The detection limit was set at three times the amount detected in the operational blank.
Specific surface area analysis
Since radiocesium concentration depends on specific surface area (He and Walling, 1996), we also corrected concentrations for surface area. Particle size distributions were analyzed by laser diffraction particle size analyzer (SALD-3000S, Shimadzu, Japan). Specific surface areas were 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 solid samples were corrected as follows (He and Walling, 1996):

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

where $S$ is the specific surface area, ref indicates a reference sample (arithmetic mean of river water samples at st. d), $\nu = 0.65$ (He and Walling, 1996), $^{137}\text{Cs}$ is uncorrected concentration, and $^{137}\text{Cs}'$ is the concentration corrected for surface area. A similar value (0.76) for $\nu$ was found in Fukushima (JAEA, 2013a).

Source analysis
Estimated contributions of radiocesium from individual sources to composite river water samples were based on the chemical mass balance method (Moyo et al., 2013). River water during dry weather, river sediments, and road dust were considered as potential sources. We estimated the contributions of SS from individual sources as follows:

$$Y_{i,\text{comp}} = aX_{i,\text{rw(dry)}} + bX_{i,\text{rs}} + cX_{i,\text{rd}}$$

where $Y_{i,\text{comp}}$ is the estimated concentration of analyte in the composite samples, $i$ is the analyte (PPCP or sterol), $X$ is the measured concentration of analyte, rw(dry) is the river water sample in dry weather, rs is the river sediment sample, rd is the road dust sample, and $a$, $b$, and $c$ are the respective contribution ratios of SS from each source ($a + b + c = 1$).

The contribution ratios were determined to minimize the sum of the squares of ratios of differences between estimated and observed values to observed values of individual analytes ($\sum((Y_{i,\text{comp}} - X_{i,\text{comp}})/X_{i,\text{comp}})^2$). For the river sediment and road dust samples, the arithmetic mean values of analytes in samples collected at st. a – c (upstream of st. d) and st. 1 – 6 were used. For the river water samples in dry weather, the arithmetic mean of analytes in samples on the dates corresponding to the collection date of composite river water samples was used. Analyte profiles (compositional ratio of individual sterols to total sterols) were similarly used instead of analyte concentrations to support the results based on analyte concentration. In the use of profiles, the estimated values of $a$, $b$, and $c$ represent contributions of sterols.

The contributions of radiocesium concentrations from individual sources to composite samples were estimated as follows:

$$Y_{\text{Cs,comp}} = aX_{\text{Cs,rw(dry)}} + bX_{\text{Cs,rs}} + cX_{\text{Cs,rd}}$$

where $Y_{\text{Cs,comp}}$ is the estimated concentration of Cs in composite samples, and $X$ is the measured concentration of Cs.
Only $^{137}$Cs concentrations were used in the analysis, because $^{137}$Cs has a longer half life (30 y) than $^{134}$Cs (2.06 y) (ICRP, 1983).

RESULTS AND DISCUSSION

Dissolved and particulate radiocesium concentrations

The radioactivity ratios of $^{134}$Cs to $^{137}$Cs, decay-corrected to 21 March 2011, were 1.06 ± 0.08 (arithmetic mean ± standard error) for grab samples and 1.01 ± 0.04 for composite samples. The ratios of radioesium emitted from the Fukushima Dai-ichi Nuclear Power Plant were ~ 1 (Yamamoto et al., 2012; Matsunaga et al., 2013). These results indicate that the radioesium in our samples originated in the 2011 accident.

Dissolved and particulate concentrations of $^{137}$Cs in grab samples in dry weather in May 2012 were 0.24 Bq/L and 9,600 Bq/kg-SS (0.10 Bq/L). During the following 1-y sampling period they decreased to ~ 16% – 17% of the initial values (Fig. 2). $^{137}$Cs concentrations in composite samples in May and June 2012 were 9,700 – 15,900 Bq/kg-SS and subsequently also decreased to ~ 16%. Note that the long half-life (i.e., 30 y (ICRP, 1983)) means that decay contributed little to the decrease. While a similar decreasing trend has been observed in other rivers (Oura and Ebihara, 2012; Nagao et al., 2013; Ueda et al., 2013), this is the first study to provide evidence from an urban river based on frequent field samplings.

![Fig. 2](image-url)

Fig. 2 - $^{137}$Cs concentrations in grab and composite samples at st. d in the Ohori River. (a) Particulate (SS-weight basis), (b) particulate (liquid volume basis), (c) dissolved (liquid volume basis). Composite samples were collected with an SS sampler. Arrows indicate samples collected during wet weather. One of four samples was not analyzed for the dissolved phase. The open circle indicates that Cs was not detected, so half of the detection limit was plotted. Precipitation was monitored at Abiko City (JMA, 2013).
Tsuji et al. (2014) and Yoshimura et al. (2015) reported that $^{137}\text{Cs}$ concentrations (SS-weight basis) correlated strongly with average $^{137}\text{Cs}$ depositions in a catchment (area basis). Therefore we compared the ratios of $^{137}\text{Cs}$ concentration (SS-weight basis) to deposited $^{137}\text{Cs}$ between the Ohori River and the Abukuma River and other rivers in Fukushima prefecture (Fig. 3). As the values in the Ohori River we used the average deposited $^{137}\text{Cs}$ in Kashiwa (decay-corrected to June and December 2012) determined by airborne monitoring surveys in April – May and October – December 2012 (NRA, 2013). As the comparators we used the ratios of $^{137}\text{Cs}$ concentration to deposited $^{137}\text{Cs}$ in grab samples (Tsuji et al., 2014) and composite samples in the Abukuma River and other rivers (Yamashiki et al., 2014, Yoshimura et al., 2015). The ratios for both the grab and composite samples in the Ohori River (grab: median, 0.20 m$^2$/kg-SS in June – July 2012, 0.15 m$^2$/kg-SS in December 2012 – January 2013; arithmetic mean 0.19 m$^2$/kg-SS in June – July 2012, 0.15 m$^2$/kg-SS in December 2012 – January 2013; composite: median, 0.26 m$^2$/kg-SS in June – July 2012, 0.24 m$^2$/kg-SS in December 2012 – January 2013; arithmetic mean 0.29 m$^2$/kg-SS in June – July 2012, 0.28 m$^2$/kg-SS in December 2012 – January 2013) were much higher than those in the Abukuma River and other rivers. We attribute the difference to the differences in sources associated with different land use types between Kashiwa city (urban area) and Fukushima prefecture (forested area). This explanation is consistent with the finding by Tsuji et al. (2014) that the ratios were higher in locations neighboring urban areas. This result highlights that $^{137}\text{Cs}$ concentrations on an SS-weight basis in rivers in urban areas are higher at the same deposition of $^{137}\text{Cs}$ than those in forested areas.

Although not all increases in dissolved $^{137}\text{Cs}$ concentrations corresponded with rain events, during wet weather the particulate concentrations consistently increased on both an SS-weight and a liquid volume basis (Fig. 2). Similar results were obtained for $^{134}\text{Cs}$. These results are also consistent with the finding that ratios of particulate $^{137}\text{Cs}$ concentrations obtained by composite samples to those by grab samples collected only during dry weather were significantly higher than 1 (Fig. 4; $1.53 \pm 0.10$ [arithmetic mean ± standard error], $P < 0.001$ by 1-sample $t$-test). The sampling efficiency (SS concentration in the sampler divided by that in the channel) of the SS composite sampler increases with SS concentration and current velocity (Koga et al., 2004; Osanai et al., 2005), and thus the sampler accumulates more SS during wet weather. Therefore, our results suggest that particulate concentrations of radiocesium are higher in wet weather in this urban river. This contrasts with observations in rivers in mountainous areas in Fukushima prefecture (Ueda et al., 2013). Again, this difference is attributable to the different land uses.
Fig. 3 - Ratios of $^{137}$Cs in particulate phase (SS-weight basis) to deposited $^{137}$Cs in the Ohori River, the Abukuma River, and other rivers in Fukushima. (a) Grab samples in dry weather; (b) composite samples. a: Tsuji et al. (2014). The Abukuma River and two branch rivers (Kuchibuto River and Shakado River). Monitoring locations C and E, neighboring urban areas, were excluded. b: Yamashiki et al. (2014). The Abukuma River and one branch river (Kuchibuto River) in Fukushima. c: Yoshimura et al. (2015). The Abukuma River and seven branch rivers (Kuchibuto River, Hirose River, Syakado River, Suriage River, Ara River, Aise River, and Otakine River) in Fukushima (apart from two sites in Miyagi). d: Yoshimura et al. (2015). Six river systems (Mano River, Same River, Fujiwara River, Niida River, Odaka River, and Ukedo River) in Fukushima. Two data of 0.20 m$^2$/kg-SS overlapped in June – July 2012 in the Ohori River.

Fig. 4 - Ratios of $^{137}$Cs in particulate phase (SS-weight basis) between composite and grab samples at st. d. Concentrations in grab samples are arithmetic means of values in dry weather samples on the dates corresponding to the collection date of composite river water samples.
The ratios of the particulate phase of $^{137}$Cs to total concentrations (liquid volume basis) in grab samples ranged from 10% to 58% (arithmetic mean: 28%) in dry weather and from 59% to 84% (arithmetic mean: 72%) in wet weather, and increased with increased SS concentrations (Fig. 5). Samples in dry weather showed a linear relationship between particulate concentrations (SS-weight basis) and dissolved concentrations (liquid volume basis), indicating that $K_d$ was constant at $3.2 \times 10^4$ (95% confidence interval: $2.5 - 3.8 \times 10^4$; Fig. 6). This $K_d$ value is very close to the geometric mean of $2.9 \times 10^4$ obtained by a review of freshwater field measurements (range: $1.6 \times 10^3 - 5.2 \times 10^5$ (IAEA, 2010)). The estimate is generally 1 – 2 orders of magnitude lower than reported values in rivers in Fukushima prefecture (Nagao et al., 2013; Ueda et al., 2013). This distinction is also attributable to the difference in solid characteristics associated with land use. Interestingly, one wet-weather observation deviated remarkably from the regression line (Fig. 6). Further research would be needed to investigate the variation in $K_d$, particularly in wet weather.
Sources of radio cesium
To identify potential sources of radio cesium in the Ohori River, we compared concentrations among river water, river sediment, road dust, and road runoff samples (Fig. 7a). The water from the Tone River was ruled out as a source because the $^{137}$Cs concentrations (SS, 4.4 mg/L; $^{137}$Cs, 230 Bq/kg-SS; 18 July 2012 [dry weather] at st. g) were much lower than those in the Ohori River (SS, 7.4 mg/L; $^{137}$Cs, 7,900 Bq/kg-SS; on the same date at st. d). We considered the sources of radio cesium in the Ohori River to be surface deposits (represented by road dust) in the catchment and river sediments.

The highest concentrations of $^{137}$Cs were found in road dust and road runoff samples, followed by river water samples, and the lowest in the river sediment samples. We also compared $^{137}$Cs concentrations corrected by specific surface area among river water and source samples: the road dust sample again had the highest concentration, and concentrations in river sediment samples were similar to those in river water samples (Fig. 7b). These results indicate that road dust (in particular) and river sediments are sources of radio cesium in the Ohori River. Interestingly, road dust contained high concentrations of radio cesium even at 1.6 years after the accident. Asphalt materials showed the highest activity reduction among various surface materials (Mueck and Steger, 1991). Since radio cesium was present within ~ 3 mm of surface asphalt materials after the 2011 accident (JAEA, 2012, 2013b), radio cesium trapped within the road surface was released with asphalt wear and gradually washed off with rainfall. This wash-off of radio cesium in urban surface deposits such as road dust explained the increase in radio cesium concentrations in the river during wet weather.

![Fig. 7 - Comparison of $^{137}$Cs concentrations (weight basis) among river water samples at st. d and source samples. (a) Without correction for surface areas, (b) With correction for surface areas. Arithmetic mean values of specific surface areas were used to represent the values for river water samples (0.57 m$^2$/g for grab (dry) (30 July and 29 August 2013); 0.28 m$^2$/g for grab (wet) (26 June, 27 July, and 28 July 2013); 0.33 m$^2$/g for composite (29 August 2013)) and road runoff samples (0.62 m$^2$/g) (4 September 2013), because the data were not available. The value for the grab sample collected at dry weather was used as a reference.](image-url)
We performed source analysis using the hydrophobic PPCPs and sterols to roughly approximate the contributions of radiocesium from individual sources. We regarded the composite river water samples to be representative of river water and the potential sources to be river water in dry weather, river sediments, and road dust. River water in dry weather includes water from the Tone River supplied at st. g, but that in wet weather does not. Radiocesium concentrations depend on particle size (He and Walling, 1996), and concentrations showed strong positive correlations with ignition losses in river sediments in the same area (Nakata et al., 2014), as seen with sterols (Writer et al., 1995). The use of hydrophobic markers can therefore reduce the biases due to differences in particle size and organic matter concentration (Writer et al., 1995) and thereby correct contributions of radiocesium. We assumed that the radiocesium and markers showed conservative behavior (e.g., limited desorption) in waters in this assessment.

There were clear differences in PPCP and sterol concentrations and profiles among individual sources (Fig. 8). River water in dry weather contained high concentrations of these markers, reflecting contamination by sewage. Cholesterol and coprostanol were predominant in river water during dry weather and in river sediment samples, indicating the typical impact of urban sewage (Isobe et al., 2002). On the other hand, road dust contained a high concentration of β-sitosterol, which is typically predominant in rural sediments and bird droppings (Leeming et al., 1996; Isobe et al., 2002). The sterol concentrations and profiles in composite river water samples lay among the individual sources, indicating that the composite river water samples were derived from a mixture of these sources.

Fig. 8 - Comparison of PPCPs and sterols among river water samples and source samples. (a) Concentrations, (b) profile.
The source analysis showed that the estimated concentrations of $\sum$(PPCPs + sterols) (sum of 2 PPCPs and 10 sterols) agreed well with those observed (Fig. 9a, $r^2 = 0.91$), and gave arithmetic mean contributions of 25% of SS from river water in dry weather, 60% from river sediments, and 15% from road dust to the composite river water samples. The sterol profile showed contributions of 64%, 17%, and 19%, respectively, indicating a small contribution of road dust to both parameters.

The contributions of radiocesium from the three sources to the composite river water samples are shown in Fig. 9b. Even though road dust contributed only 15% of SS in the water, it contributed much larger proportions of $^{137}$Cs than the other sources: 22% of $^{137}$Cs (arithmetic mean) came from river water in dry weather, 39% from river sediments, and 39% from road dust, highlighting the latter two as major sources. There was no contribution from road dust on some dates; minor contributions of SS might not be captured, and therefore the contribution of $^{137}$Cs in road dust would be underestimated. The estimated $^{137}$Cs concentrations agreed well with those observed in general (although they were underestimated on some dates, probably because of the lack of capture of SS from road dust; Fig. 9c).

Estimation of the contributions of radiocesium from individual sources identified surface deposits such as road dust as major sources even 2 years after the accident. To more accurately apportion the contributions of road dust, other source-specific markers of road dust (e.g., benzothiazolamines (Spies et al., 1987), resin acids (Kumata et al., 2011), nitropolycyclic aromatic hydrocarbons (Murakami et al., 2008), and heavy metals (Censi et al., 2011)) may be useful.

![Fig. 9](attachment:fig9.png)

Fig. 9 - (a) Comparison between estimated and observed concentrations of $\sum$(PPCPs + sterols). (b) Estimated concentrations of $^{137}$Cs derived from each source. (c) Comparison between estimated and observed concentrations of $^{137}$Cs. $\sum$(PPCPs + sterols) represents sum of 2 PPCPs and 10 sterols.
CONCLUSIONS
We analyzed sources, behavior, and temporal trends of radiocesium in an urban river, the Ohori River, by collecting 1-year field samples.

1 Concentrations of $^{137}$Cs in particulate and dissolved phases decreased to ~ 16% of initial values within a year. The $K_d$ value in dry weather was estimated to be $3.2 \times 10^4$.

2 The ratios of $^{137}$Cs concentration (SS-weight basis) to deposited $^{137}$Cs in the Ohori River (urban area) (e.g., median in June – July 2012, grab: 0.20 m$^2$/kg-SS; composite: 0.26 m$^2$/kg-SS) were higher than those in the Abukuma River (forested area), probably because of differences in land use.

3 Concentrations of radiocesium in the particulate phase were higher in wet weather than in dry weather on both an SS-weight and a liquid volume basis.

4 Source analysis of composite samples collected between May 2012 and February 2013 showed that 22% (arithmetic mean) of $^{137}$Cs came from river water in dry weather, 39% from river sediments, and 39% from road dust. Surface deposits such as road dust were major sources of radiocesium in the river, even 2 years after the accident. The wash-off of surface deposits during wet weather increased the radiocesium concentrations in the river.

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