Analysis of Temporal Changes in Terrestrial Cesium-137 Using Publicly Available Monitoring Data in Japan

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Abstract

Since the Fukushima Daiichi Nuclear Power Plant accident, vast amounts of radionuclide monitoring data obtained from various governmental and institutional surveys have been released to the public. In this study, we analyzed these large amounts of data to evaluate the long-term, large-scale fate of Cesium-137 in eastern Japan. Analysis of *in situ* measurements showed that temporal changes in the *virtual* amount of Cs-137 deposited, which were estimated from measured air dose rate data, varied greatly among measurement points, depending on air dose level and land use. The decrease in *virtual* amounts deposited was comparable to the decay rate of radioactive cesium, but was large in areas where the air dose rate had been relatively high and human activities had occurred. An analysis of Cs-137 concentrations in river and lake sediments showed Cs-137 levels to decrease at a fast rate, one order of magnitude faster than the radioactive decay rate. The present study highlights the importance of the effects of vertical and/or horizontal migrations of Cs-137 within the terrestrial area (including decontamination activities), direct deposition of Cs-137 into surface water, behavior of Cs-137 in built-up areas, and Cs-137 behavior in stagnant water bodies (e.g., ponds and lakes). The present study has demonstrated the utility analyzing large amounts of publicly available data for evaluating the environmental behavior of Cs-137.

Key words: long-term fate, monitoring data, radioactive cesium, terrestrial area

1. Introduction

More than five years ago, a massive amount of radionuclides was released into the atmosphere from the Fukushima Daiichi Nuclear Power Plant (FDNPP), transported over eastern Japan through advection and diffusion, and deposited onto terrestrial and marine areas. Among the released radionuclides, Cs-137 is one of the most important radionuclides in terms of exposure to humans and the environment, because of the large amounts released and long half-life of 30.1 years (Yasunari *et al*., 2011). While decontamination work has been carried out to reduce air dose rates, understanding the long-term fate of Cs-137 is important for predicting future contamination levels in living environments. This must be done on a large spatial scale, because the region affected by the radioactive cesium from the FDNPP extends beyond Fukushima Prefecture, affecting a large surrounding region in eastern Japan. While a few studies suggest the utility of modeling approaches (e.g., Kitamura *et al*., 2014; Evrard *et al*., 2015), analyzing measurement data obtained since the accident is the first step toward evaluating the large-scale, long-term behavior of Cs-137 in the environment.

Since the Fukushima accident, the Japanese government and several institutes have made huge efforts to monitor contamination levels and study the behavior of radioactive Cs in the environment through various surveys, such as airborne monitoring (Ministry of Education, Culture, Sports, Science and Technology (MEXT), 2011), car-borne monitoring (e.g., Andoh *et al*., 2015), *in situ* measurement (MEXT and the Secretariat of the Nuclear Regulation Authority (NRA), 2014), and public water monitoring (Ministry of the Environment (MOE), 2011). The results of each of those surveys have been useful in elucidating the contamination status since the accident. There have been very few studies, however, that comprehensively utilize such vast amounts of data to understand the spatial and temporal patterns of the environmental behavior of Cs. Gonze *et al*. (2016) compared these survey results and revealed that the temporal change in mean (spatial

average) air dose rates revealed by *in situ* measurement was slower than that revealed by other survey methods. They also compared car-borne monitoring data, classified by Kinase *et al.* (2014), according to land-use categories of urban areas, agricultural environments, deciduous forests and evergreen forests. The rate of decrease of air dose rates in urban areas was the fastest among these in the four land use categories. Andoh *et al.* (2015) showed through car-borne surveys that the rate of decrease of air dose rates at built-up sites was faster than in forested areas and farmland, probably due to the effect of weathering. These results indicate that *in situ* measurement, which is measured at 1 m above the ground surface, is most suitable for analyzing the characteristics of Cs-137 behavior based on air dose rates for each land use type. There have been no studies, however, analyzing the behavior of Cs-137 with respect to air dose rates and land use types, on the basis of large-scale air dose rate data obtained by *in situ* measurements. Only in one study in 2015 were air dose rates obtained by *in situ* measurements analyzed for temporal changes, which did not differ by land use (Mikami & Saito, 2013). In this study, however, the air dose rate data were analyzed as a sum of all survey locations. Therefore, the temporal changes at locations with high air dose rates would naturally dominate temporal changes overall, which may have masked temporal changes at locations with lesser air dose rates.

Most of the Cs-137 deposited in the surface soil layer is observed to be present in a particulate phase (International Atomic Energy Agency (IAEA), 2010). Some of the Cs-137 deposited on the ground surface is discharged by various processes such as weathering and human activities, and most of the discharged Cs-137 finds its way into rivers and lakes. In those surface waters, a fraction of the Cs-137 is present in a dissolved phase (Matsunaga *et al.*, 1991; Eyrolle & Charmasson, 2001), which includes a pseudo-dissolved one (i.e., present on microscopic particles that could pass through a filter used to remove particles). Field studies have shown that the contribution of each phase to Cs-137 flux from surface soil to rivers varies, but in general, more Cs is discharged in particulate phases. Yamashiki *et al.* (2014) estimated that the contribution of particulate Cs-137 to the total flux into the Pacific Ocean was higher than that of dissolved Cs-137. They estimated that particulate fractions of radioactive Cs accounted for 84–92% of total radioactive Cs transported in the Abukuma River from August 10, 2011 to May 11, 2012. Nagao *et al.* (2013) estimated that particulate fractions of radioactive Cs accounted for 21–56% of total radioactive Cs, but approached 100% after Typhoon Roke during September 21–22, 2011 in the Natsui and Same rivers during July to December 2011. They also estimated the flux of radioactive Cs attributable to the typhoon to have accounted for 30–50% of the annual flux to the Pacific Ocean. As particles eventually settle in surface waters, the sediments of lakes and ponds naturally accumulate radioactive Cs (Hayashi, 2017).

Detailed evaluations of the behavior of radioactive Cs in the sediments of lakes and ponds, however, are very rare, except for a few studies on the radioactive Cs profile with regard to particle size in the sediments of surface waters (e.g., Aoi *et al.*, 2014; Yoshimura *et al.*, 2015). No evaluations of temporal changes in sedimentary Cs-137 have been conducted on a large spatial scale.

In this paper, we have attempted to provide insight into the environmental behavior of Cs-137 through analysis of large amounts of publicly available monitoring data obtained from various governmental and institutional surveys. Our objectives were to evaluate the temporal changes in i) terrestrial Cs-137 amounts with respect to air dose rate levels and land use, and ii) Cs-137 concentration in river and lake sediments on a large scale in eastern Japan. Our novel approach was to evaluate the temporal changes at each of the monitoring locations, so that differences according to air dose levels and land use types could be evaluated in detail.

2. Materials and Methods

2.1 Analysis of Terrestrial Cs-137 Behavior from Air Dose Rate Data

In order to investigate terrestrial Cs-137 behavior on a large scale, we used air dose rate data obtained by *in situ* measurement, which is the air dose rate measured at 1 m above the ground surface (MEXT & NRA, 2014). The measurements were generally conducted in open, flat areas with little disturbance from human activities. Because the measured air dose rate reflects radiation from both Cs-137 and Cs-134 existing in various topographies and at various depths, and because the data had different measurement dates, we used the following equation and assumptions to correct for the radioactive decay and estimate the *virtual* Cs-137 deposition amount at the soil surface, as explained below.

$$\widehat{DR} = CF_{137}Dp_{v137}exp\left(-\frac{ln2}{\tau_{137}}t\right)$$
$$+ CF_{134}Dp_{v134}exp\left(-\frac{ln2}{\tau_{134}}t\right) + DR_{base} \qquad (Eq. 1)$$

where $\widehat{DR}$ is the measured air dose rate at time t, t is the time elapsed from March 11, 2011 until the date of measurement, CF is the conversion factor from the amount of radioactive cesium deposited to the air dose rate, Dp_v is the *virtual* amount of radioactive cesium deposited onto the soil surface at time zero, assuming that all of the radioactive cesium exists on the soil surface, τ is the half-life, and DR_{base} is the background radiation air dose rate. The subscript numbers in the variables indicate the radioactive cesium mass number. For data analysis, we assumed that Dp_{137} was equal to Dp_{134} (Saito *et al.*, 2015), DR_{base} was equal to zero, and CF_{137} and CF_{137} were set at 2.1×10^{-6} and 5.4×10^{-6} (Sv h^{-1} Bq^{-1} m^2), respectively, which are the values

assuming that all of the radioactive cesium exists on an infinitely flat surface (IAEA, 2000). We also assumed that the radioactive cesium had been deposited all at once at time zero, which meant that the time difference between time zero and the actual deposited dates (the actual depositions occurred on multiple dates) could be ignored. Hence, if the *virtual* Cs-137 deposition amount changed with different measurement campaigns at the same locations, it would be because there were processes which altered the conversion factor between the amount deposited and the air dose rate, such as vertical migration of Cs-137, or it would be because the actual amount of Cs-137 deposited changed between the measurement campaigns (e.g., by horizontal migration of Cs-137).

For the analysis, we used the 3rd, 4th, 5th and 6th *in situ* measurements, which were performed in August 2012, November 2012, June 2013 and November 2013, respectively (MEXT & NRA, 2014). Although the multiple measurements were repeatedly conducted at the same locations, the data available lacked a location identifier, having geocordinates only. Therefore, the data seemingly conducted at the same monitoring points were judged by latitude and longitude data with ArcGIS® 10.2. First, the monitoring points in the 6th survey were set as the standard, and any monitoring points in the other surveys that were within 10 m of one of the points in the 6th survey were judged as the same location. Second, we divided these surveys into a first group (the 3rd and 4th surveys) and a second group (the 5th and 6th surveys), and the monitoring points that had survey data in both the first and second group were selected for further analysis, in order to choose monitoring points that had a long enough time interval between the measurements. Third, we calculated the "change rate (year^{-1})" in the *virtual* Cs-137 depositions at each selected monitoring point, which is a first-order rate coefficient obtained from the regression line of decay-corrected, log 10-normalized concentrations; these data were then categorized according to land use type as of 2009 (Ministry of Land, Infrastructure, Transport and Tourism of Japan, 2009). Note that inside the evacuation zone near the FDNPP, the land use type may have changed since the FDNPP accident. We focused on four major land use types for comparison of Cs-137 change rates: built-up areas, forests, paddy fields, and agricultural fields excluding paddy fields. Finally, we obtained 4,043 monitoring points that met our conditions. Their locations are shown in Fig. 1. The Steel-Dwass method was used to evaluate the differences in the average change rates among land uses.
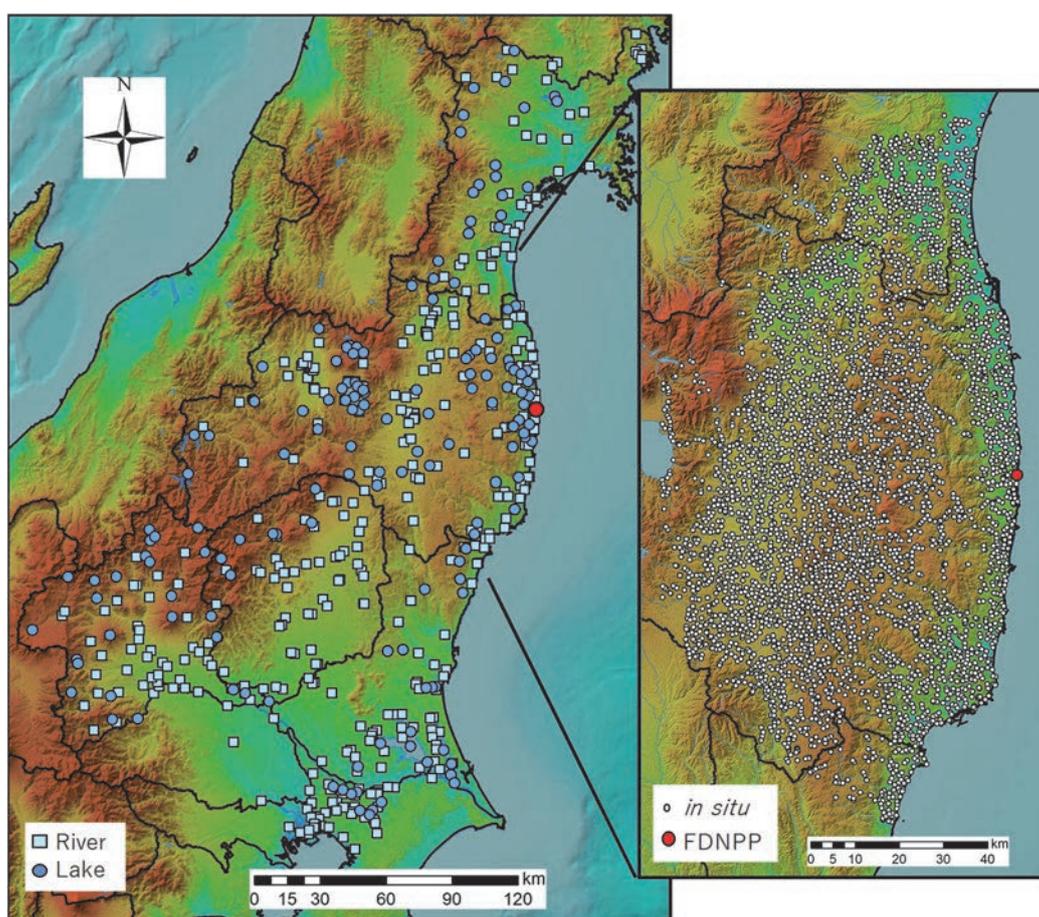
Fig. 1 The locations of *in situ* measurement and surface water sediment surveys. The altitude map was adapted from the Geospatial Information Authority of Japan (2017).

2.2 Analysis of Changes in Radioactive Cs Concentration in Surface Water Sediments

The MOE has conducted monitoring surveys of radioactive Cs in environmental waters since 2011 (MOE, 2011). We collected monitoring data on Cs-137 concentrations in river and lake sediments obtained from August 2011 to June 2014 in Miyagi, Fukushima, Ibaraki, Tochigi, Gunma, Saitama and Chiba prefectures and the Tokyo Metropolis. The numbers of sampling points were 512 for rivers and 167 for lakes; the respective total numbers of sampling data were 5,373 and 1,888.

He and Walling (1996) reported effects of soil particle size on the amount of Cs-137 adsorbed. Therefore, we corrected the size effect of Cs-137 for each sediment sample following He and Walling's method, which suggests that the concentration of Cs-137 is proportional to a power function of the mean specific surface areas, and that the exponent of the power function is 0.65. The concentration of Cs-137 was standardized to that of a standard specific surface area of 90 $(cm^2\ g^{-1})$ using the mass ratios of soil size fractions (clay, silt, fine sand, medium sand, coarse sand, granules, pebbles and coarse gravel) and the soil particle density of each sample.

We selected sampling points where there were more than three measurements on different dates in which the Cs-137 concentration was detected and the particle-size effect correction was implemented as above. The numbers of sampling points that met the above-mentioned conditions were 374 and 165 for rivers and lakes, respectively (the locations are shown in Fig. 1). Then, similarly to the previous section, we calculated the first-order rate coefficient as the change rate (year^{-1}) of the sedimentary Cs-137 concentrations at each sampling point, based on the regression line of decay-corrected log 10-normalized concentrations.

3. Results and Discussion

3.1 Variance in Temporal Changes in Cs-137 Depending on Air Dose Levels and Land Use

Temporal changes in the *virtual* amount of Cs-137 deposited had a distinct pattern depending on the air dose levels and land use (Fig. 2). In Fig. 2, the measurement points were divided into eight groups of equal size, according to the magnitude of air dose rates averaged arithmetically for each point. Each resulting group was further categorized into four land use types. For the three groups with low air dose rates (Lv0–2: 0–0.30 μSvh^{-1}), no decrease in *virtual* amounts deposited could be clearly observed (median change rate ~0 year^{-1}). The distribution of the change rate was almost the same among all land use types (change rate: −0.1–0.1 year^{-1}). In the remaining groups with higher air dose rates, the *virtual* amount deposited showed decreases at more than half of the measured locations for Lv3–4 (0.30–0.53 μSv h^{-1}) and in more than three quarters for Lv5–7 (>0.53 μSv h^{-1}). Interestingly, the *virtual* amount deposited decreased faster with increasing air dose levels toward Lv5, but then decreased more slowly toward Lv7 in all four land-use categories.
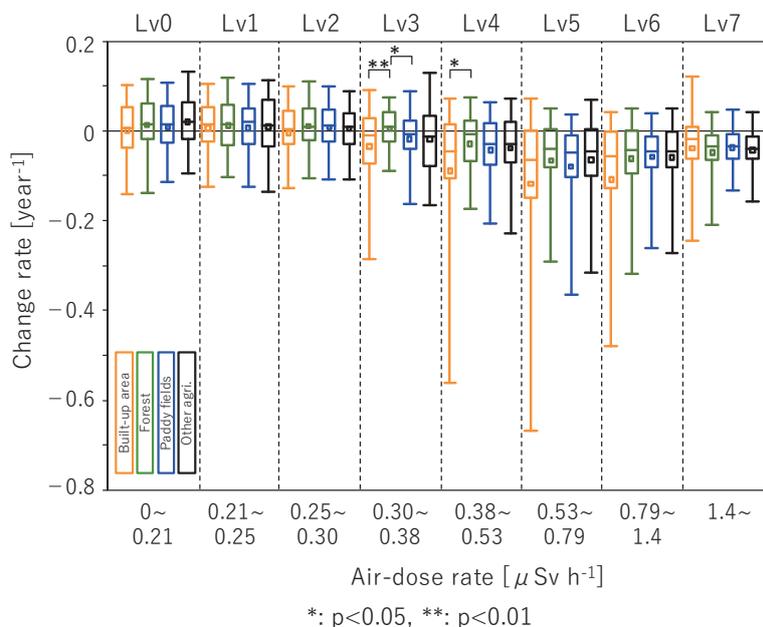
Fig. 2 Comparison of change rates (negative values indicate decreases) of the *virtual* amounts of Cs-137 deposited, not involving radioactive decay, among land-use categories and air dose rate levels. The land use categories are built-up areas, forest, paddy fields and other agricultural fields. The air-dose rate categories are Lv0: 0 – 0.21, Lv1: 0.21 – 0.25, Lv2: 0.25 – 0.30, Lv3: 0.30 – 0.38, Lv4: 0.38 – 0.53, Lv5: 0.53 – 0.79, Lv6: 0.79 – 1.4, Lv7: >1.4 (μSv h^{-1}, decay corrected, measured at 1 m above ground level). The box plots indicate the 5th, 25th, 50th, 75th, 95th percentiles; the squares indicate arithmetic mean values. Significant differences are indicated in arithmetic mean values among land uses, evaluated by the Steel-Dwass method.

The median value of the change rate for all the measurement points was approximately −0.015 year^{-1}, which was comparable to the radioactive decay rate of Cs-137 (−0.023 year^{-1}). In comparison, the median values for Lv5 and Lv6 were −0.047 year^{-1}, which was twice the radioactive decay rate. The rate of decrease at those high air dose rate locations tended to be largest in the built-up areas. The built-up areas had the largest number of locations with extremely fast decreases (e.g., change rates of −0.1 year^{-1} or smaller). The built-up areas in Lv5 showed the fastest decrease, with a median change rate of −0.065 year^{-1}. Moreover, in Lv3 and Lv4, the built-up areas showed significantly larger rates of decrease than the forested areas (Fig.2). Notably, however, in areas with high air dose rates (i.e., Lv 6 and Lv7: >0.79 μSv h^{-1}), the difference in change rates between built-up areas and the other land uses seemed much smaller or nonexistent.

The reasons for these tendencies were probably one or a combination of the following effects. First, background radiation, which was assumed to be nonexistent in the analysis, could have masked temporal changes in measured air dose rates in areas with low air dose rates (i.e., Lv0–2). Second, human activities could accelerate vertical and/ or horizontal migration and weathering of Cs-137. Such human activities would include decontamination activities and the movement of vehicles on roads. This mechanism would explain the highest decrease rates in the *virtual* amount deposited in the built-up areas among all the land uses. It is not possible at this point, however, to distinguish each effect of human activities. For this, a more detailed evaluation would be needed with detailed information on decontamination activities. In comparison, the highest air dose rate areas (i.e., Lv6 and Lv7) showed somewhat similar rates of decrease among land uses, probably because a part of those monitoring points were located in the evacuation zones, where such human activities

would have been limited. Seventy-one percent of the locations surveyed for Lv7 and 7 percent of the locations surveyed for Lv6 were within the "special decontamination area" designated by the MOE, where decontamination activities would have been limited as of the time of the *in situ* measurements (MOE, 2017). Therefore, when human activity is excluded, the horizontal and/ or vertical migration and weathering effects of radioactive cesium are considered to occur similarly among all land uses, with a rate of decrease (e.g., Lv7: −0.035 year^{-1}) comparable to the radioactive decay rate. These results are consistent with those of Mikami and Saito (2013). The fast decrease in the *virtual* amount deposited in the built-up areas is consistent with the decrease in air dose rates observed in several other studies (Kinase *et al*., 2014; Andoh *et al*., 2015). For the first time, however, we showed that the rates of decrease in built-up areas vary considerably depending on the air dose rate levels.

3.2 Change Rates in Cs-137 Concentration More Variable in Lake Sediments than River Sediments

Figure 3 shows a scattering plot between the change rates of Cs-137 concentrations and residual errors in the regression for each sampling point. The median change rates were one order of magnitude larger than the radioactive decay rate (−0.023 year^{-1}), at −0.29 year^{-1} (5th percentile −0.59, 95th percentile −0.06) for river sediments and −0.15 year^{-1} (−0.73, 0.35) for lake sediments. The river sediments had larger rates of decrease, but had smaller variation in change rates and residual errors than lake sediments. This lower variation in river sediments indicates that Cs-137 in river sediments could behave more similarly regardless of location. On the other hand, the behavior of Cs-137 in lake sediments could vary more among the locations, probably depending on the characteristics of the catchments and lakes, such as land use, radioactive Cs
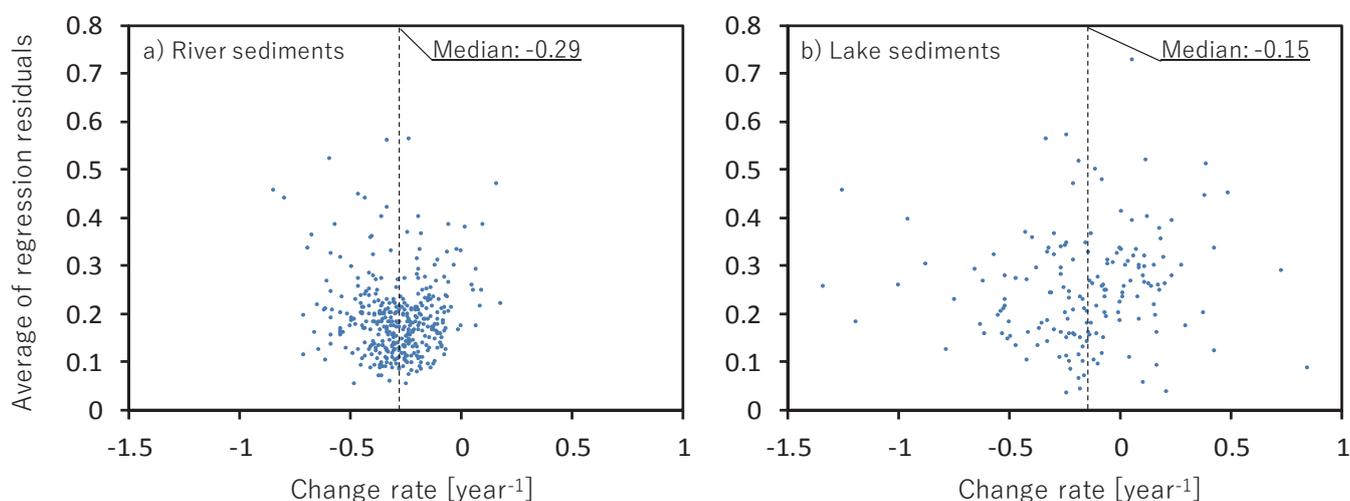
Fig. 3 Distribution of the change rates of Cs-137 concentrations, for which radioactive decay and particle-size effects were corrected, in log10 scale (negative values indicate decreases) and residual errors in the regression for each sampling point. The median values of the change rates are also indicated. a) River sediments, b) Lake sediments.

inventory, sediment properties, flow rates and amount of sediment transported. Sediments of lakes and ponds are known to accumulate radioactive Cs (Hayashi, 2016), but there are very few detailed evaluations of the behavior of radioactive Cs in the sediments of lakes and ponds (e.g., Aoi *et al.*, 2014; Yoshimura *et al.*, 2015). Future studies should therefore explore the behavior of radioactive Cs further in such stagnant water bodies and the factors influencing it.

3.3 Comparison between Temporal Change Rates of *Virtual* Amounts Deposited and Those of Sedimentary Cs-137 Concentrations

The change rates of *virtual* amounts deposited and sedimentary Cs-137 concentrations could be roughly categorized into three groups: fast (about twice or more faster than the radioactive decay of Cs-137), medium (the same as radioactive decay), and slow (about one or more orders slower than radioactive decay). The change rate of *virtual* Cs-137 depositions on the ground surface was medium, although some areas with fairly high air dose rates (0.53–1.4 μSv h^{-1}) showed fast decreases in air dose rates. The decreases in Cs-137 concentrations in sediments were fast at many sampling points, especially in river sediments.

The observed decrease in Cs-137 levels in surface water sediments was far faster than the decrease of *virtual* amounts of Cs-137 deposited on the ground surface. This would be partly due to the effect of direct deposition of Cs-137 into surface water bodies and mixing of the sediment. Because of the fast decrease of Cs-137 levels in surface water sediments, the concentrations of Cs-137 in surface water sediments just after the Fukushima accident would have been much higher than the measured concentrations of Cs-137 in the sediments. The high Cs-137 concentrations could have been due to the direct deposition of Cs-137 into the surface water bodies. While part of the surface water sediments flow out into downstream catchments, soil and sediments from upstream catchments would flow into the surface water bodies. The soil and sediments from upstream catchments, however, would originate largely from terrestrial soils, whose Cs-137 concentration would be much lower than surface water sediments affected by direct deposition. Thus, the Cs-137 concentration in surface water sediments would decrease fast, by mixing with upstream soil and sediments. Another factor in the fast decrease in Cs-137 in surface water sediments could be the effect of fast flushing, which was inferred by Smith *et al.* (2004) just after fallout following global atmospheric nuclear weapons testing during 1954–1963 and the 1986 nuclear accident at Chernobyl. Therefore, for accurate evaluation of the long-term, large-scale fate of Cs-137 in terrestrial environments, it is thought essential to consider vertical and/or horizontal migration of Cs-137 within the terrestrial area, especially just after the Fukushima accident, and direct deposition of Cs-137 into surface water bodies.

4. Conclusions and Suggestions

We analyzed temporal changes in *virtual* Cs-137 depositions and Cs-137 concentrations measured in river and lake sediments. Decreases in the *virtual* Cs-137 depositions were in general comparable to the decay rate of radioactive cesium, but larger where the air dose rate was relatively high and human activities occurred. Cs-137 concentrations in surface water sediments showed a fast decrease, especially in river sediments. The decrease rate coefficient was one order of magnitude larger than the radioactive decay rate.

In comparison, in a model simulation of the fate of terrestrial Cs-137 in rivers, the overall outflow rate from river catchments was estimated to be much slower (change rate: ~-0.005 year^{-1}; Kitamura *et al.*, 2014). The discrepancy between the observed decrease rates of *virtual* amounts deposited based on air dose rates and the modeled Cs-137 fate could be explained by the contribution of vertical and/or horizontal migrations of Cs-137 within the terrestrial area, including decontamination activities, which would weaken the observed air dose rates at 1 m above the ground surface. Therefore, for an accurate evaluation of air dose rates and Cs-137 behavior over the long term on a large spatial scale, future studies should consider details of the effects of vertical and/or horizontal migrations of Cs-137 within the terrestrial area and direct deposition of Cs-137 into surface water. The behavior of Cs-137 in the built-up areas and stagnant water bodies and the factors influencing it are also identified as important for future consideration. The present study also demonstrated the utility of analyzing the large amounts of publicly available data for evaluating the environmental behavior of Cs-137.

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