Radioactive contamination in the Tokyo metropolitan area in the early stage of the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident and its fluctuation over five years

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Abstract

Radioactive contamination in the Tokyo metropolitan area in the immediate aftermath of the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident was analyzed via surface soil sampled during a two-month period after the accident. $^{131}$I, $^{134}$Cs, and $^{137}$Cs were detected in these soil samples. The activity and inventory of radioactive material in the eastern part of Tokyo tended to be high. The $^{134}$Cs/$^{137}$Cs activity ratio in soil was 0.978 ± 0.053. The $^{131}$I/$^{137}$Cs ratio fluctuated widely, and was 19.7 ± 9.0 (weighted average 18.71 ± 0.13, n = 14) in the Tokyo metropolitan area. The radioactive plume with high $^{131}$I activity spread into the Tokyo metropolitan area and was higher than the weighted average of 6.07 ± 0.04 (n = 26) in other areas. The radiocesium activity and inventory surveyed in soil from a garden in Chiyoda Ward in the center of Tokyo, fell approximately 85% in the four months after the accident, and subsequently tended to rise slightly while fluctuating widely. It is possible that migration and redistribution of radiocesium occurred. The behavior of radiocesium in Tokyo was analyzed via monitoring of radiocesium in sludge incineration ash. The radiocesium activity in the incineration ash was high at wastewater treatment centers that had catchment areas in eastern Tokyo and low at those with catchment areas in western Tokyo. Similar to the case of the garden soil, even in incineration ash, the radiocesium activity dropped rapidly immediately after the accident. The radiocesium activity in the incineration ash fell steadily from the tenth month after the accident until December 2016, and its half-life was about 500 days. According to frequency analysis, in central Tokyo, the cycles of fluctuation of radiocesium activity in incineration ash and rainfall conformed, clearly showing that radiocesium deposited in urban areas was resuspended and transported by rainfall run-off.
Introduction
The tsunami triggered by the East Japan Great Earthquake Disaster of March 11, 2011 cut off all electric power to Fukushima Daiichi Nuclear Power Plant (FDNPP). No. 1, No. 2, and No. 3 reactors lost their cooling capability, their nuclear fuel went into melt down and their reactor vessels were destroyed between March 12 and 15 [1–6]. As a result, part of the radioactive material discharged into the atmosphere was deposited across eastern Japan [7–11]. Based on monitoring data from each location, it is assumed that radioactive material in the plume that spread into the Tokyo metropolitan area on March 16 and 22 was deposited on the ground via rainfall. The timing and route of the spread of the radioactive plume have been variously hypothesized, but have not been clarified in detail as of yet [12–17]. When we started our measurements, almost no public information about the radioactive contamination in the Tokyo metropolitan area and Kanto district had been shared.

The world has experienced nuclear power plant accidents before, including nuclear fuel melt down at Three Mile Island (1979) [18] and Chernobyl (1986) [19,20]. Six years after the FDNPP accident in 2011, much about the behavior of radioactive material in the environment has been clarified [1,21]. The FDNPP accident caused the nuclear fuel meltdown simultaneously in three reactors, discharging a large quantity of radioactive material into the environment [6–11]. One characteristic of the FDNPP accident is that a large quantity of radioactive material was discharged into the topographically and meteorologically complex natural environment of Japan. Because many of the urban areas are formed in the plains at the foot of steep mountainous areas, they are frequently affected by heavy rains such as typhoons.

Another major characteristic is that the plume containing a large amount of radioactive material drifted into the Tokyo metropolitan area [16,21–24]. The capital, Tokyo, where over 30 million residents live, is located south of Kanto Plain, about 230 km southwest of FDNPP. Conditions and radioactive contamination shortly after the FDNPP accident have been reported in a variety of publications [2–5]. However, these include almost no discussion of the fact that radioactive materials were carried into the Tokyo metropolitan area, which is a densely populated urban area and the socio-economic and cultural core of Japan, nor of any evaluation of the fluctuations in the radioactive material in Tokyo’s urban districts.

This study surveyed the distribution of radioactive materials in soil to evaluate the fluctuation of radioactive contamination in the Tokyo metropolitan area and the Kanto district in the immediate aftermath of FDNPP accident. Changes in radionuclide activities and inventories in soil between the time of the accident and the present time in the central part of Tokyo were monitored to clarify the behavior of the radioactive materials in the urban districts. Moreover, the behavior of radionuclides in the urban environment, which is covered by concrete and asphalt, was analyzed based on changes in the radiocesium activity in incineration sludge ash discharged from Tokyo’s wastewater treatment centers [25].

Sampling and methods
Soil sampling
Most soil samples were taken from the private site. These collections were approved by the owner of the land. In the case of collection at public places, sampling was carried out with the permission of the administrator when necessary.

For this study, soil samples were collected in the Tokyo metropolitan area and the Kanto district during the two months following the FDNPP accident. In general, the samples were obtained from the ground surface along roads where they were not subject to physical turba-
in many cases were hardly affected by the flow of rainwater and were not covered with plants. Soil in Site 'i' was sampled in an area $0.5 \times 1$ m, and the core samples were obtained to a depth of 20 cm, varying the position slightly for each sampling in the area. After sampling, the holes were refilled. There was grass, vegetation, etc. at the sampling locations, and the soil was exposed at the ground surface. The ground was slightly sloped, so it is assumed that when rain falls, rainwater flows gently over the ground surface. Pedestrians could walk on the sampling locations. Surface soil up to 1 cm deep below the surface was measured using a measure and removed from a $10 \times 10$ cm area. A core sample was collected to a depth of 20 cm using a core sampler with an inner diameter of 5 cm, and sliced to an appropriate thickness to prepare an analytical sample. The soil samples were air-dried and impurities, such as vegetation detritus and rocks with diameters of 1 mm or larger, were removed before their radioactivity was measured. The sampling sites were selected in the Tokyo metropolitan area and Kanto district approximately 200 km from FDNPP, and are shown in Fig 1. Sampling of soil should normally be done by the established IAEA method [26]. However, this method could not be applied to urban regions contaminated with activity, because buildings and roads are interspersed there. The inventory of radionuclides was calculated assuming soil density of 1,300 kg m$^{-3}$ in compliance with the method of the MEXT of Japan [27], because accurate evaluation of soil density is difficult and many already reported inventories of radiocesium are estimated using this value.

**Measuring radioactivity**

The activity concentrations of radionuclides in the soil were quantified by connecting a 4096 multichannel pulse height analyzer (Labo Equipment, MCA600) to a low energy HPGe.
detector (ORTEC LO-AX/30P) sheathed in 10 cm thick lead. The specimen was sealed inside a plastic container with a diameter of 5.5 cm and depth of 2.0 cm, in preparation measurement by γ-ray spectrometry. The Ge detector calculated the geometric efficiency relative to the sample volume using the American NIST Environmental Radioactivity Standards, SRM 4350B (River Sediment) and SRM 4354 (Freshwater Lake Sediment), and the numerical efficiency was corrected within the range of 2 to 30 g of sample weight [28]. The measurement time was set so that the counting error would be less than 5%, depending on the radioactive intensity of the samples. In this study, $^{131}$I (364 keV), $^{134}$Cs (605 keV), and $^{137}$Cs (662 keV) were quantified. A $^{134}$Cs solvent with known density was used to correct the sum peak effect for $^{134}$Cs.

The radioactivity was calculated based on a value for the day the samples were obtained. Late at night on March 15, 2011, the No. 2 reactor went into melt down, causing the discharge of the largest quantity of radionuclides during the FDNPP accident [2,3,6,23], and the radioactivity measured in this study was evaluated based on a value obtained by decay correction at noon on March 16, 2011. Because its half-life is short (8.04 days), $^{131}$I was not detected from samples collected after late June 2011. The samples that were not analyzed immediately after sampling showed lower measurement precision for $^{131}$I than for $^{134}$Cs or $^{137}$Cs.

Radioactivity of sludge incineration ash
The radioactivity in incineration sludge ash discharged by Tokyo’s wastewater treatment centers was monitored by Tokyo Metropolitan Government beginning in May 2011, and the results have been provided on the Metropolitan Tokyo website [25]. In most areas of Tokyo, domestic wastewater is mixed with rainwater in the wastewater treatment system and this mixture is fed to the wastewater treatment centers for purification. Radiocesium is very efficiently captured in the sludge that is recovered, which is incinerated and disposed of as incineration ash. Therefore, it can be assumed that the radioactivity of incineration ash discharged from Tokyo’s wastewater treatment centers reflects the quantity of radioactive material deposited in their respective catchment areas [29]. Thus, changes in the radiocesium activity of the sludge incineration ash from the wastewater treatment centers was analyzed to evaluate the distribution and behavior of radiocesium in Tokyo for a five year period after the FDNPP accident.

Frequency analysis between radioactivity in sludge incineration ash and monthly rainfall
The relationship between the radiocesium activity in the sludge incineration ash and the monthly rainfall in the Tokyo metropolitan area was frequency analyzed using the MATLAB software.

Results and discussion
Distribution of radionuclides in soil of the Tokyo metropolitan area in the immediate aftermath of the accident
In this study, surface soil was sampled from inside Tokyo and in the prefectures of Fukushima, Tochigi, Gunma, Ibaraki, Chiba, and Saitama, as shown in Fig 1, between late March, immediately after the accident, and early June. The measurement results are shown in Table 1, as the activity and inventory normalized for the time each was sampled and for March 16, 2011. Regarding the radioactive contamination of soil caused by the FDNPP accident, the geographical heterogeneity of the radioactive materials deposited on the ground surface was uneven and narrow ranging. This based on the analytical results of five core samples taken from the four corners and center of a 10 × 10 m uncultivated paddy field at Site 'B' in Tochigi,
Table 1. Analytical results of the activity of radionuclides in the soil samples collected in the Tokyo metropolitan area and the Kanto district area.

| Prefecture | Site Description | Collected Date | Taken cm | Decay Corrected Activity | Ratio for $^{137}$Cs | Inventory kBq m$^{-2}$ |
|------------|------------------|----------------|----------|--------------------------|----------------------|----------------------|
|            |                  |                |          | $^{131}$I | $^{134}$Cs | $^{137}$Cs | $^{134+137}$Cs | $^{131}$I | $^{134}$Cs | $^{137}$Cs | $^{134+137}$Cs |
| Fukushima  | A Schoolyard (1) | 2011/3/19      | 0–1      | 122000 | 13900 | 14100 | 28000 | 8.652 | 0.986 | 1590 | 363 |
|            | A Schoolyard (2) | 2011/4/27      | 0–1      | 142000 | 21400 | 21900 | 43300 | 6.484 | 0.977 | 1850 | 563 |
| Tochigi    | B Pasture        | 2011/6/4       | 0–1      | 43600  | 11100 | 11300 | 22400 | 3.858 | 0.982 | 566  | 290 |
|            | Garden (1)       | 2011/6/13      | 0–1      | 24500  | 3020  | 3150  | 6170  | 7.778 | 0.959 | 318  | 80.2 |
|            | Garden (2)       | 2011/6/13      | 0–1      | 91800  | 6730  | 6810  | 13500 | 13.48 | 0.988 | 1190 | 176 |
| Nagano     | G Plateau Forest | 2011/4/28      | 0–1      | 513    | 550   | 557   | 1110  | 0.920 | 0.986 | 6.7  | 14.4 |
|            | Hotel Garden     | 2011/4/28      | 0–1      | 3350   | 726   | 755   | 1480  | 4.439 | 0.962 | 43.6 | 19.3 |
| Gunma      | C Paddy Field    | 2011/4/30      | 0–1      | 5640   | 1830  | 1850  | 3680  | 3.049 | 0.989 | 73.3 | 47.9 |
|            | D Forest         | 2011/3/26      | 0–1      | 582    | 67.4  | 70.7  | 138   | 8.232 | 0.953 | 7.6  | 1.8  |
|            | Farmland (1)     | 2011/4/29      | 0–1      | 1630   | 183   | 184   | 367   | 8.854 | 0.996 | 21.2 | 4.8  |
|            | Farmland (2)     | 2011/4/29      | 0–1      | 3860   | 740   | 745   | 1490  | 5.178 | 0.993 | 56.7 | 45.1 |
| Saitama    | I Rode Side      | 2011/4/28      | 0–1      | 4360   | 1760  | 1720  | 3480  | 2.535 | 1.023 | 56.7 | 45.1 |
|            | J Road Side      | 2011/4/28      | 0–1      | 3130   | 204   | 207   | 412   | 15.10 | 0.986 | 40.7 | 5.4  |
|            | K River Bank     | 2011/4/10      | 0–1      | 6130   | 248   | 252   | 500   | 24.29 | 0.981 | 79.6 | 6.5  |
|            | e Garden         | 2011/5/19      | 0–1      | 25200  | 1060  | 1080  | 2140  | 23.33 | 0.981 | 50.1 | 19.3 |
| Ibaraki    | L Garden         | 2011/4/20      | 0–1      | 4850   | 225   | 237   | 462   | 15.92 | 0.983 | 360  | 44.8 |
|            | M Garden         | 2011/4/20      | 0–1      | 8420   | 105   | 108   | 213   | 78.11 | 0.977 | 109  | 2.8  |
| Chiba      | N Garden         | 2011/4/10      | 0–1      | 582    | 59.7  | 69.2  | 129   | 8.41  | 0.863 | 7.6  | 1.7  |
|            | a Road Side      | 2011/4/11      | 0–1      | 7020   | 672   | 672   | 1340  | 10.45 | 1.000 | 91.3 | 17.5 |
|            | b Road           | 2011/4/11      | 0–1      | 12100  | 2460  | 2510  | 4970  | 4.821 | 0.980 | 157  | 64.6 |
|            | c Garden         | 2011/5/19      | 0–1      | 40700  | 5300  | 5330  | 10600 | 7.636 | 0.994 | 529  | 138  |
|            | Road Sludge      | 2011/6/11      | 0–1      | 114000 | 33900 | 33900 | 67800 | 3.363 | 1.000 | 1480 | 881  |
|            | Road Side (1)    | 2011/6/11      | 0–1      | 28500  | 17700 | 17700 | 35400 | 1.610 | -     | -    | -    |
|            | Road Side (2)    | 2011/6/11      | 0–1      | 26100  | 3210  | 3190  | 6400  | 8.182 | -     | -    | -    |
|            | Road Side (3)    | 2011/6/2       | 0–1      | 11600  | 514   | 536   | 1050  | 21.66 | 0.959 | 151  | 13.6 |
|            | m Garden         | 2011/5/19      | 0–1      | 6740   | 432   | 442   | 874   | 15.24 | 0.976 | 87.6 | 11.4 |

Decay corrected activity as corrected on the March 16, 2011. Inventory was calculated assuming 1,300 kg m$^{-3}$ for the soil density. At Site ‘i’, measurements continued from April 10, 2011 until August 16, 2016. The results are shown in Fig 3. nd: Not Detected ($^{131}$I; < 2 Bq kg$^{-1}$, $^{134}$Cs and $^{137}$Cs; < 0.6 Bq kg$^{-1}$).

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shown in Fig 1. More than 95% of radiocesium deposited on the soil surface at these points was retained in the upper 5 cm depth from the surface of the 20 cm deep core, while the standard deviation of the inventory was ± 28.9% (125–280 kBq m⁻², n = 5). The uneven distribution of radioactive materials deposited on the surface in this way was confirmed at many points. This suggests the possibility that radioactive materials in the plume were not uniformly mixed physically while they were transported in the atmosphere.

Geographical distribution of radionuclides

The geographical distribution of radioactive materials in the soil in the Tokyo metropolitan area and Kanto district, as shown in Fig 1, was interpolated from the results of aerial monitoring by the MEXT that were announced at December 16, 2011 [30]. The radionuclide inventory in the Kanto district was high in Tochigi, northwest Gunma, eastern Nagano, and northern Chiba, but lower in Saitama and southern Chiba. In the Tokyo metropolitan area, it was high in part of eastern Tokyo. The contamination level was extremely high in a roadside ditch sludge in Kashiwa City (Site ‘c’) of northern Chiba, because radionuclides were concentrated in the drainage. Excluding this area, places showing the highest inventories in the Tokyo metropolitan area were 1010 kBq m⁻² in Koto Ward (Site ‘g’) for ¹³¹I and 460 kBq m⁻² in Kashiwa City (Site ‘c’) for ¹³⁴+¹³⁷Cs (total of ¹³⁴Cs and ¹³⁷Cs). In central Tokyo (Sites ‘f’ to ‘m’, n = 12), for ¹³¹I, it was 695 ± 291 kBq m⁻², and for ¹³⁴+¹³⁷Cs, it was 74.9 ± 27.3 kBq m⁻², which are about 40% and 20% of the ¹³¹I and ¹³⁴+¹³⁷Cs levels in Fukushima City (Site ‘A’), respectively. This result also suggests that the plume, which includes radiiodine activities much higher than those of radiocesium during the immediate aftermath of the accident, spread into the Tokyo metropolitan area [16,22–24].

¹³¹I /¹³⁷Cs and ¹³⁴Cs /¹³⁷Cs activity ratios

The isotope ratios are discussed in relation to the source of the radionuclides [23,31–33]. As shown in Table 1 and Fig 2, the ¹³⁴Cs/¹³⁷Cs activity ratio was determined to be 0.989 ± 0.017

![Fig 2. Activity ratios of ¹³¹I/¹³⁷Cs and ¹³⁴Cs/¹³⁷Cs in the soil collected from eastern Japan.](https://doi.org/10.1371/journal.pone.0187687.g002)
(weighted average $0.992 \pm 0.002, n = 40$). This value conformed closely to the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio 1.0 of radiocesium discharged due to the FDNPP accident within the limits of counting errors [32,33]. The $^{131}\text{I}/^{137}\text{Cs}$ activity ratio caused by the accident has been reported to be about 10 [6,7], but in Fukushima and Tochigi, which are near the FDNPP, it was $8.05 \pm 3.53$ (weighted average $7.64 \pm 0.07, n = 5$), indicating an activity ratio that approaches this. But in Nagano, Gunma and southern Chiba (Site ‘N’) which are far from the FDNPP, it was $4.91 \pm 2.95$ (weighted average $3.48 \pm 0.09, n = 9$), and in the Saitama, northern Chiba, Ibaraki and Tokyo metropolitan area, it was $18.94 \pm 15.38$ (weighted average $9.67 \pm 0.07, n = 25$), indicating significant regional differences. As shown in Fig 2, the $^{131}\text{I}/^{137}\text{Cs}$ ratio was high especially in the central part of Tokyo. It is highly likely that such differences in $^{131}\text{I}/^{137}\text{Cs}$ ratios reflect differences in the physical and chemical behavior of iodine and cesium in the transport and deposition processes of the radioactive plume. From the vertical distribution of radioiodine and radiocesium at Sites ‘D’ and ‘I’ (Table 1), there is a small possibility that differences in their behavior in the soil after deposition on the ground surface had an effect on the measured $^{131}\text{I}/^{137}\text{Cs}$ ratios. In other words, it is assumed that the $^{131}\text{I}/^{137}\text{Cs}$ ratio was extremely high in the Tokyo metropolitan area, because a plume retaining a relatively high $^{131}\text{I}$ activity spread into Tokyo. This indicates that the radiocesium deposited preferentially compared to $^{131}\text{I}$ in the process of a plume moving from FDNPP [16,22–24].

Change over time of the radionuclide activity and inventory in garden soil in central Tokyo

At Site ‘I’ in a garden in Chiyoda Ward, located in the center of Tokyo, the radioactivity and inventory of radionuclides in the soil were surveyed continuously. The first sampling was done on April 10, 2011, and the $^{131}\text{I}$ (2,140 Bq kg$^{-1}$) and $^{134+137}\text{Cs}$ (1,900 Bq kg$^{-1}$) were detected at high activities. Both nuclides were concentrated mostly on the surface, as other locations [34, 35]. Fig 3 shows the temporal changes in radioactivity in the layer 1 cm from the surface and

![Radioactive Decay Curve of $^{134+137}\text{Cs}$](https://doi.org/10.1371/journal.pone.0187687.g003)
of inventory in all layers (depth from 0–15 cm) of $^{134+137}$Cs from the accident. The decay curve of radiocesium at the time when it was hypothesized that the activities of $^{134}$Cs and $^{137}$Cs deposited in the immediate aftermath of the accident were the same is also shown in Fig 3. From April 10 to 28, 2011, in the immediate aftermath of the accident, the activities and inventories of $^{131}$I and $^{134+137}$Cs increased abruptly (Table 1). If this increase is not a result of uneven distribution of radionuclides in the samples, the spread of the radionuclides into the Tokyo metropolitan area from the FDNPP probably continued at this time [24]. However, the $^{134+137}$Cs activity fell to about 10% of its initial value by August 13, while $^{131}$I was not detected at the time, so we assumed that the spread of new radionuclides from the nuclear reactor had stopped.

The adsorption behavior of radionuclides to clay minerals has been studied for many years and it is well known that radiocesium penetrates layers of clay minerals where it undergoes strong ionic adsorption and is bound by the soil particles [36–45]. The torrential rainfall from Typhoon 2 on May 29 and 30, 2011 washed out soil particles that had adsorbed radiocesium, such that it is undeniable that this may have reduced the radiocesium activity. However, because heavy rainfall has fallen in the Tokyo metropolitan area since that time, soil run-off due to rainfall alone cannot account for the rapid decline in the radioactivity in the immediate aftermath of the accident. This fact suggests that radiocesium that was deposited immediately after the FDNPP accident could have contained a large quantity of a highly mobile form radiocesium. In other words, it can be assumed that this radiocesium existed as a chemical species that is not easily held in the soil. The radiocesium activity and inventory in the garden soil of Site ‘i’ appear to have fluctuated widely, as they steadily increased over time beginning in the second year after the accident. The radioactivity level predicted based on radioactive decay beginning in the third year after the accident is shown in Fig 3, and indicates that it is possible that the movement and recycling of radiocesium at this location were balanced. The changes over time in radioactivity of the soil in the Tokyo metropolitan area could possibly reflect the effects of uneven distribution due to the sampling locations, so fluctuation in radioactivity over time was studied using sludge incineration ash.

**Behavior of radionuclides in the Tokyo metropolitan area shown by sludge incineration ash from wastewater treatment centers**

Regarding the process of movement of radioactive materials that have pervaded the environment from their catchment areas via rainfall and rivers, various measurement data and the results of simulations based on these have been reported [46–53]. A detailed comparison of model predictions and actual observed results in Sweden after the Chernobyl accident was also conducted on the $^{137}$Cs migration process by sewage treatment system in the urban environment [54]. However, the fluctuation of radionuclides in urban areas remains almost completely unknown. Radiocesium in sludge incineration ash discharged from wastewater treatment centers in Tokyo was used as a tracer in this study to evaluate the fluctuation of radiocesium in the urban environment. Wastewater treatment centers at 12 locations in Tokyo, shown in Fig 4, have published changes over time in the radiocesium activities in combined sludge incineration ash from May 2011 up to the present time [25]. Of the wastewater treatment centers listed on the website, the Nanbu wastewater treatment center (NA) also processes sludge it receives from other treatment plants [29].

**Geographical distribution and change over time of radiocesium activity in combined sludge incineration ash**

Fig 5 shows the changes over time in radiocesium activity in sludge incineration ash discharged from each wastewater treatment center. In May 2011, when the measurement values
Fig 4. Sites of Tokyo’s wastewater treatment centers and their catchment areas. Base map is from website USGS National Map Viewer. (http://viewer.nationalmap.gov/viewer/).

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Fig 5. Temporal changes of the $^{134+137}$Cs radioactivity in the incineration ash from Tokyo’s wastewater treatment centers.

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were published, the Kasai center (KA), whose catchment area is in eastern Tokyo, showed the highest activity, at about 70,000 Bq kg\(^{-1}\), whereas the Minami Tama center (MT) in the southwest showed the lowest activity of about 3,000 Bq kg\(^{-1}\). The radiocesium activity in the sludge incineration ash decreased from the east to the west. This finding conforms to aerial monitoring and soil measurement values, showing that the radiocesium activity in sludge incineration ash reflects the radioactivity level of the catchment area of each wastewater treatment center.

The radiocesium activity in sludge incineration ash decayed almost exponentially with the passage of time. Its decay velocity was higher than the radioactive decay of radiocesium discharged by the FDNPP accident, showing that radiocesium in the environment was effectively removed by the wastewater treatment systems. The half-life of the eliminated radiocesium was estimated at between 460 and 580 days. Not all the radiocesium deposited in the Tokyo metropolitan area was discharged through the wastewater treatment systems, but in the center of Tokyo, from 0.12–0.15% of radiocesium deposited on the surface was removed each day.

Table 2 shows the annual average activity (arithmetic mean value) of radiocesium in sludge incineration ash discharged from the wastewater treatment centers. Furthermore, Table 2 also shows the annual discharge quantity of incineration sludge ash and the annual discharge quantity of radiocesium calculated based on the quantity and radioactivity discharged. The radiocesium activity and quantity discharged varied greatly according to the catchment area of the wastewater treatment centers. The quantity of radiocesium discharged as sludge incineration ash from immediately after the accident until December 2015 reached 0.60 TBq (×10\(^{12}\) Bq; not decay corrected). This value includes the value of the NA plant that also accepted sludge from other plants in central Tokyo. The radioactivity level in the Tokyo metropolitan area was categorized into regions X, Y, and Z, shown in Fig 4, based on the levels of radiocesium activity in the sludge incineration ash.

### Estimating radiocesium balance in the Tokyo metropolitan area by the sludge incineration ash wastewater treatment centers

The percentage of radiocesium deposited in the immediate aftermath of the FDNPP accident that was discharged through wastewater treatment centers was estimated. Table 3 shows the
total quantity of radiocesium discharged by each wastewater treatment center up to the end of
December 2015, decay corrected to March 16, 2011 in the immediate aftermath of the acci-
dent. The quantity of $^{134+137}$Cs deposited in the region studied was estimated from the results
of aerial monitoring in October 2011 as 2.46 TBq [30], and from values measured in the soil, it
was estimated as 5.35 TBq, despite the inadequate number of samples. The quantity discharged
by sludge incineration ash was 0.55 TBq, so the percentage of radiocesium that was washed-
out as run-off by wastewater treatment systems until the end of 2015 was calculated to be
approximately 22% from aerial monitoring and approximately 10% from soil measurements.

Either this means that more than approximately 80% of the radiocesium deposited at the time
of the accident is still held in the soil. The remaining approximately 20% has been washed-out
of the soil as run-off through a route other than wastewater treatment centers.

Run-off process of radiocesium via sludge incineration ash

As already stated, in the garden soil of Site ‘i’, the radiocesium activity in the soil fell abruptly
in the immediate aftermath of the FDNPP accident. This phenomenon was also found in the
school ground soils of Koto Ward in eastern Tokyo [55]. Even in the case of sludge incineration
ash, as shown in Fig 6, with the monthly rainfall in the Tokyo metropolitan area, the rapid
decline of the radiocesium activity in the immediate aftermath of the accident has been con-


### Table 3. Estimating the radiocesium balance in the Tokyo metropolitan area from the wastewater treatment systems.

| Wastewater Treatment Centers | Catchment Area km² (s) | Radiocesium Discharged Amount as Ash | Estimated Inventory from the MEXT Aircraft Monitoring | Estimated Inventory from the Soil Radioactivity | Estimated Outflow of the Past Five Years (%) |
|-----------------------------|------------------------|-------------------------------------|-------------------------------------------------|-----------------------------------------------|---------------------------------------------|
|                            |                        | TBq (t) | kBq m⁻² | TBq (u) | kBq m⁻² | TBq (v) | (t/u) | (t/v) |
| X KA Kasai                 | 48.93                  | 0.280   | 25      | 1.223  | 50      | 2.447   | 26.7   | 13.3  |
| TB Tobu                   | 35.12                  | 0.157   | 7       | 0.246  | 40      | 1.405   | 72.3   | 12.7  |
| MY Miyagi                 | 16.87                  | 0.027   | 6       | 0.101  | 20      | 0.101   | 30.6   | 9.2   |
| Y TK2 Kitatama 2          | 27.44                  | 0.005   | 5       | 0.137  | -       | -       | -      | -     |
| SN Shingashi              | 28.35                  | 0.054   | 5       | 0.142  | 7       | 0.199   | 43.6   | 31.2  |
| TK1 Kitatama 1            | 51.23                  | 0.015   | 5       | 0.256  | 11      | 0.564   | 8.5    | 3.0   |
| NA Nanbu                  | 4.37                   | 0.143   | 5       | 0.022  | -       | -       | -      | 653.1 |
| Z HC Hachioji             | 85.33                  | 0.002   | 1       | 0.085  | -       | -       | 3.0    | -     |
| TAR Tamagawa              | 93.49                  | 0.004   | 1       | 0.095  | -       | -       | 5.3    | -     |
| KY Kiyose                 | 80.42                  | 0.006   | 1       | 0.080  | 5       | 0.402   | 7.8    | 1.6   |
| AS Asagawa                | 39.02                  | 0.001   | 1       | 0.037  | -       | -       | 2.8    | -     |
| MT Minamitama             | 59.00                  | 0.001   | 1       | 0.059  | -       | -       | 2.5    | -     |
| Total or Average          | 542                    | 0.552   | 4.5 (u/s) | 2.46 | 20.5 (v/s) | 5.35 | 22.4 | 10.1 |

Radiocesium discharged amount as ash was obtained by decay correcting an annual discharged amount on March 16, 2011. Radioactivities of $^{134}$Cs and $^{137}$Cs immediately after the accident were assumed to be equal, as shown in Table 1. The values of the Nanbu Plant (NA) were excluded because there is sewage processing from other facilities. Since there is no data that can be referenced, the Nanbu, Hachioji, Tamagawa, Asagawa and Minamitamagawa were excluded from the calculation of the total and average. Precipitations of radiocesium in area Z are estimated values.

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deposited immediately after the accident flowed into and were discharged from wastewater treatment systems. We could not find these radiocesium species directly, but cesium-bearing microparticles alluded to by many researchers [57–62] are considered to be the highly mobile form of cesium that we estimated. As shown in Table 4, from 82–85% of the radiocesium discharged from wastewater treatment systems was discharged during the 10 months when the radioactivity fell rapidly in the immediate aftermath of the accident. Regarding the steady state period from October 2011 to the present, the periodicity was examined between rainfall and radiocesium activity in the sludge incineration ash using MATLAB. The results are presented in Fig 7, and show that in the urbanized X and Y districts, the rainfall and radiocesium activity conform in 4, 8, and 12 month cycles. This suggests that in these districts, radiocesium flowed into the wastewater treatment systems, carried by rain water. However, in district Z, which has a different urban structure, such periodicity was not found. This result suggests that the abrupt fall in the radiocesium activity observed in the immediate aftermath of the accident in garden soil in the center of Tokyo and in sludge incineration ash was not dependent on rainfall.

Table 4. Proportion of the migratory radiocesium discharged soon after the accident.

| Area | Amount of Migratory $^{134+137}$Cs Bq kg$^{-1}$ | $^{134+137}$Cs in the Sharp Decline Period |
|------|---------------------------------|---------------------------------|
|      | Initial Amount Bq kg$^{-1}$ | Discharged Amount Bq kg$^{-1}$ | Discharged % |
| X    | 50000                          | 30000                           | 42000         | 84  |
| Y    | 20000                          | 13400                           | 17000         | 85  |
| Z    | 6000                           | 3770                            | 4900          | 82  |

Amount of migratory $^{134+137}$Cs was estimated from Fig 6 on March 16, 2011. Period of Sharp decline for $^{134+137}$Cs from immediately after the deposition to the December 2011. Initial amount was the measured radioactivity of the ash at May 2011. *Discharged %* indicates that the percentage of $^{134+137}$Cs discharged in the initial ten months from the total amount of $^{134+137}$Cs discharged by the wastewater treatment system from October 2011 to December 2016.

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The adsorption behavior of cesium to suspended particles has been mentioned already [63]. Recently, extremely fine particles containing high activity of radiocesium discharged by the FDNPP accident have been detected in different regions [57–62]. It is assumed that radiocesium on such extremely fine particles does not exist as an ionic species, and hence it is unlikely that it can be maintained adsorbed in clay layers. If it is assumed that extremely fine particles containing radiocesium were deposited on the ground surface in the immediate aftermath of the accident, it is highly likely that it was more easily transported by falling rain or blowing wind than occur as ionic cesium that is easily held in soil. Furthermore, it is considered that radiocesium contained in such extremely fine particles is insoluble in water, so it is presumed that when radiocesium contained in these particles deposited on concrete or asphalt, it is selectively washed-out by rainfall. Quantitative data on the coverage of buildings, concrete and asphalt to the ground in areas X, Y, and Z could not be obtained. However, the period of decrease in radiocesium in the sludge incineration ash in areas X and Y is consistent with the fluctuating cycle in rainfall amounts. It can be thought that radiocesium deposited in these areas located in the central area of Tokyo is effectively decontaminated by rainfall. On the other hand, the radiocesium activity in the garden soil of Site ‘i’ located in area X, has not fluctuated significantly due to rainfall after the accident. Also, in the case of area Z, periodicity was not observed between rainfall and its radiocesium out-flow. This result indicates that not only the physical and chemical states of radionuclides but also the urban structure may have a significant influence on the behavior of radionuclides when urban areas, such as the Tokyo metropolitan area, receive radioactive contamination.

**Conclusions**

Changes during the past five years in the distribution of environmental radioactive contamination in the Tokyo metropolitan area from the FDNPP accident were discussed. In the
immediate aftermath of the accident, $^{131}$I, $^{134}$Cs, and $^{137}$Cs were detected in the soil of the Tokyo metropolitan area. High activities and inventories of the radionuclides were found in eastern Tokyo and northern Chiba. Inventories of decay corrected $^{131}$I and $^{134+137}$Cs on March 16, 2011 were 88–1010 kBq·m$^{-2}$ and 11–93 kBq·m$^{-2}$, respectively, which are about 40% and 20%, respectively, of those in Fukushima City. The contamination was even higher in the adjoining northern part of Chiba located east of Tokyo.

A rapid decline in the radiocesium activity in the immediate aftermath of the accident was observed in surface soil, suggesting the possibility that the radiocesium deposited on the ground surface occurred as highly mobile chemical species. By the end of 2016, approximately 85% of the spilled radiocesium in the wastewater treatment system flowed out during the 10 months after the accident. The run-off rate was estimated between 0.12–0.15%·day$^{-1}$, signifying that the run-off half-life of radiocesium by wastewater treatment systems ranges from 460 to 580 days. From the radiocesium activity in the soil and the emissions as sludge incineration ash, it was estimated that 78–90% of the radiocesium deposited in the Tokyo metropolitan area still remains in the surface soil.

The radiocesium activity in the sludge incineration ash was higher in the urbanized eastern part of Tokyo, and there was correlation with changes in rainfall in that area. This suggests that radiocesium deposited on concrete and asphalt is more likely to be discharged than that on soil. The results reveal that the Tokyo metropolitan area even now continues to be affected by radioactive contamination caused by the FDNPP accident. We hope this study will contribute to the improved understanding of the radioactive contamination issue in Japan.

Supporting information

S1 Fig. Photos of site ‘i’. (PPTX)

S1 Table. Analytical results of the activity of radionuclides in the soil samples collected in the Tokyo metropolitan area and the Kanto district area. Decay corrected activity was corrected on the March 16, 2011. Inventory was calculated assuming 1,300 kg·m$^{-3}$ for the soil density. At Site ‘i’, measurements were continued from April 10, 2011 until August 16, 2016. The results are shown in Fig 3. nd: Not Detected ($^{131}$I; < 2 Bq·kg$^{-1}$, $^{134}$Cs and $^{137}$Cs; < 0.6 Bq·kg$^{-1}$).
(XLSX)

S2 Table. Analytical results of the activity of radionuclides in the core samples collected in the site ‘i’. Decay corrected activity was corrected on the March 16, 2011. Inventory was calculated assuming 1,300 kg·m$^{-3}$ for the soil density. nd: Not Detected ($^{131}$I; < 2 Bq·kg$^{-1}$, $^{134}$Cs and $^{137}$Cs; < 0.6 Bq·kg$^{-1}$).
(XLSX)

S3 Table. Temporal changes of analytical results of the activity of radionuclides in the incineration sludge ash in each wastewater treatment plant. nd: Not Detected. The data was referred to website of Tokyo Metropolitan Government. (http://www.gesui.metro.tokyo.jp/english/oshi/).
(XLSX)

S4 Table. Annual amount of the incineration sludge ash collected from each wastewater treatment plant. The data was referred to website of Tokyo Metropolitan Government. (http://www.gesui.metro.tokyo.jp/business/pdf/2016tokyo.pdf (in Japanese)).
(XLSX)
S5 Table. Monthly rainfall precipitation in the Tokyo metropolitan area. The data was referred to website of Japan Meteorological Agency. (http://www.data.jma.go.jp/obd/stats/etrn/index.php?sess=6e5f25a9cdef28e6a634ce58ca736e68) (in Japanese).

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