Chapter 3
Spatiotemporal Distribution of Radionuclides in Soil in the Tokyo Metropolitan Area

Abstract In the first stage after the FDNPP accident, radioactive contamination on the ground surface in the Tokyo metropolitan area was controlled by weather conditions such as wind direction and rainfall. Many radionuclides in the radioactive plume released from the FDNPP reactors were advected in the atmosphere without falling to the surface. However, some of them precipitated to the ground surface, causing environmental radiation contamination in Tokyo. These radionuclides diffused, which is believed to have caused temporary but significant external and internal exposure to the Japanese people. However, given the destruction of infrastructure due to the massive earthquake, there are almost no measurement data regarding environmental radioactive contamination immediately after the FDNPP nuclear accident. The collapse of infrastructure occurred not only in Fukushima Prefecture where the FDNPP operated but also in the capital Tokyo more than 200 km from the FDNPP. However, the status of environmental radioactive contamination immediately after the accident can be estimated by measuring the contamination by radioactive material recorded in the geosphere or hydrosphere. In this chapter, the geographic distribution of radioactive contamination of soil in the Tokyo metropolitan area during the initial phase of the FDNPP accident is discussed. The results of field sample measurements show that severe environmental radioactive contamination continued for approximately 5 years after the accident. However, after more than 9 years had passed, the radioactive contamination seemed to have changed as radionuclides migrated and were redistributed from the initial contamination situation.

Keywords Tokyo metropolitan area · Radioactive contamination · Soil · Inventory · Spatiotemporal distribution

3.1 Introduction

Immediately after the FDNPP accident, radioactive contamination in the metropolitan area was presumed to have occurred according to the following process. In the first stage, as described in Chap. 2, radioactive contamination on the ground surface

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was controlled by weather conditions such as wind direction and rainfall. Many radionuclides in the radioactive plume released from the FDNPP reactors were advected in the atmosphere without falling to the surface. However, some of them precipitated to the ground surface, causing environmental radiation contamination in Japan. These radionuclides diffused and caused global radioactive contamination, which is believed to have caused temporary but significant external and internal exposure to the Japanese people. However, given the destruction of infrastructure due to the massive earthquake, there are almost no measurement data regarding environmental radioactive contamination immediately after the FDNPP nuclear accident. The collapse of infrastructure occurred not only in Fukushima Prefecture where the FDNPP operated but also in the capital Tokyo more than 200 km from the FDNPP. However, the status of environmental radioactive contamination immediately after the accident can be estimated by measuring the contamination by radioactive material recorded in the geosphere or hydrosphere.

The first phase of environmental radioactive contamination from the Fukushima nuclear accident is considered as follows (References 25–27 in Chap. 2). Initially, hydrogen gas produced by the reaction between the nuclear fuel rod zirconium and the cooling water leaked from a crack in the reactor pressure vessel and accumulated in the reactor building; later, the hydrogen gas exploded, and the reactor building was destroyed. The fission products leaked from the Unit 1 and 3 reactors were released to the environment by the hydrogen explosion in each building. The Unit 4 reactor was not in operation, but the building was destroyed by the hydrogen gas explosion from the Unit 3 reactor. The spent nuclear fuel storage pool was also destroyed, but the nuclear fuel rods were mostly unbroken.

On the other hand, in the Unit 2 reactor, dry venting to prevent the destruction of the reactor pressure vessel was successful, but at this time, massive amounts of fission nuclides that accumulated in this meltdown reactor were released into the atmosphere. Most of these radionuclides are believed to have been released from the exhaust stack of the Unit 2 reactor. Next, these nuclides released from the FDNPP formed radioactive plumes that migrated and diffused into the atmosphere over Japan. At this time, estimates indicate that the people living in the places through which the plume passed received high internal and external exposures to the significant quantity of volatile radionuclides such as radioactive iodine and radioactive cesium, but the actual situation has scarcely been elucidated yet. Third, rainfall caused radionuclides in the plume to precipitate onto the ground, and most radionuclides were adsorbed and fixed to clay minerals in the soil. Alkali earth elements such as radioactive cesium are naturally dissolved in water as soluble ionic species, and radioactive cesium precipitated on concrete or asphalt in urban areas is considered to have been transferred to the ecosystem through rivers as ionic chemical species or to sludge in sewage treatment systems (Chap. 5). In the case of the forests, most radioactive nuclides were retained in litter and plants (Chap. 7). Radionuclides deposited in the hydrosphere were introduced into ecosystems via aquatic organisms and fish (Chap. 8). Radioactive materials released in the FDNPP accident that polluted Japan's environment moved into the hydrosphere such as lakes and oceans, due to rainfall and groundwater. In the Tokyo metropolitan area, Tokyo Bay played an essential role as the final sink for radioactive materials (Chap. 6).

In this chapter, the geographic distribution of environmental radioactive contamination in the Tokyo metropolitan area during the initial phase of the FDNPP accident is discussed. The results of field sample measurements show that severe environmental radioactive contamination continued for approximately 5 years after the accident. However, after more than 9 years had passed, the radioactive contamination seemed to have changed as radionuclides migrated and were redistributed from the initial contamination situation.

3.2 Sample and Radioactivity Measurements

3.2.1 Radioactivity Measurements

The radioactivity of gamma-emitting nuclides in soil, water, aquatic sediments, atmospheric dust, animals, plants, fish, and shellfish described in this book was quantified using the Ge semiconductor gamma-ray measurement system described below.

The activity intensities of radionuclides in the samples 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 a depth of 2.0 cm in preparation for measurement by gamma-ray spectrometry. The relative geometric efficiency of the Ge detector with respect to the sample volume was calculated using the American NIST Environmental Radioactivity Standards, SRM 4350B (river sediment) and SRM 4354 (freshwater lake sediment). Moreover, the counting efficiency was corrected within the range of sample weight from 2 to 30 g [1]. For samples with a measured weight of less than 2 g, such as air dust samples on the filter, the detection efficiency curve obtained with the NIST SRM standards was extrapolated to correct the counting efficiency. The measurement time was set such that the counting error would be 5% or less, depending on the radioactive intensity of the sample.

In this study, the following nuclides were quantified: ^{95}Zr (γ-ray energy: 757 keV, half-life: 64.0 days), ^{95}Nb (766 keV, 35.0 days), ^{110m}Ag (885 keV, 250 days), ^{131}I (364 keV, 8.04 days), ^{134}Cs (605 keV, 2.06 years), ^{137}Cs (662 keV, 30.1 years), ^{210}Pb (46.5 keV, 22.3 years), and ^{235}U (186 keV, 7×10^8 years). A standard ^{134}Cs solution with known concentration was used to correct the sum peak effect for the ^{134}Cs determination. The detection limits for radionuclides under appropriate conditions were 2.0 Bq/kg for ^{131}I and 0.6 Bq/kg for ^{134}Cs and ^{137}Cs in dry soil samples. The detection limits of these nuclides were almost the same in wet samples, such as fish muscle. When the samples could not be analyzed immediately after sampling, they showed lower measurement precision for ^{131}I than for ^{134}Cs or ^{137}Cs because the activity of ^{131}I decreased by radioactive decay.

The radioactivity was calculated based on a value for the day the sample was obtained. As described in Chap. 1, the Unit 2 reactor melted down late at night on

March 15, 2011, causing the discharge of the most significant quantity of radionuclides during the FDNPP accident [2–6]. Therefore, the radioactivity measured in this study was evaluated based on a value obtained by the radioactive decay corrected to March 16, 2011. Because its half-life is short (8.04 days), ^{131}I was not detected from samples collected after late June 2011.

To evaluate the reliability of the author's radioactivity measurements, they were compared with the quantitative values of the same samples measured at other institutions. Here, in addition to the soil samples, the results of cross-checking are shown for the fish sample discussed in Chap. 8. The soil samples were thoroughly air-dried in an oven at 60 °C after sample collection. To avoid secondary contamination, sieving was not performed, and plant pieces and sand grains with diameters greater than 1 mm were removed mainly with tweezers. To homogenize the sample used for the cross-check, the quartering method was repeated five times and divided into two parts; one part was measured by another institution and the other part by the author's measuring instrument. For cross-checking, the fish samples were frozen after collection. The muscle samples were half-thawed in the laboratory, and muscles were collected on both sides of the spine. One part was measured at another institution, and the other part was measured in the author's laboratory. The fish skin was removed from the muscle to avoid external contamination. Table 3.1 shows the results of cross-checking for these samples. In the sample with low radioactive contamination, the quantification

Table 3.1 Reliability of the radioactivity measurements by gamma-ray spectrometry

Sample	Facility	Decay corrected activity[a], Bq/kg			$^{134}Cs/^{137}Cs$
		^{134}Cs	^{137}Cs	$^{134+137}Cs$	
Soil 1[b]	Author (A)	13,900 ± 82.2	14,000 ± 84.6	13,950 ± 118	0.993 ± 0.008
	Other (O)	13,500 ± 106	13,400 ± 110	13,450 ± 153	1.007 ± 0.011
	A/O ratio	1.030 ± 0.010	1.045 ± 0.010	1.038 ± 0.014	1.000 ± 0.014
Soil 2[b]	Author (A)	21,500 ± 161	21,800 ± 174	21,650 ± 237	0.986 ± 0.011
	Other (O)	22,400 ± 209	22,200 ± 216	22,300 ± 301	1.009 ± 0.013
	A/O ratio	0.960 ± 0.012	0.982 ± 0.013	0.971 ± 0.018	0.998 ± 0.017
Soil 3[c]	Author (A)	900 ± 17.6	896 ± 18.8	898 ± 25.8	1.004 ± 0.029
	Other (O)	915 ± 20.3	940 ± 22.7	928 ± 30.5	0.973 ± 0.033
	A/O ratio	0.984 ± 0.030	0.953 ± 0.032	0.969 ± 0.044	0.989 ± 0.044
Fish 1[d]	Author (A)	1130 ± 23.1	1110 ± 22.5	1120 ± 32.2	1.018 ± 0.029
	Other (O)	1110 ± 41.2	1070 ± 40.7	1090 ± 57.9	1.037 ± 0.053
	A/O ratio	1.018 ± 0.042	1.037 ± 0.043	1.028 ± 0.060	1.028 ± 0.060
Fish 2[d]	Author (A)	465 ± 13.3	455 ± 11.4	460 ± 17.5	1.022 ± 0.038
	Other (O)	473 ± 18.2	456 ± 17.2	465 ± 25.0	1.037 ± 0.054
	A/O ratio	0.983 ± 0.048	0.998 ± 0.045	0.991 ± 0.066	1.030 ± 0.066

Radioactivities were measured by the method shown in Sect. 3.2. Soil samples bisected after being sufficiently homogenized by quartering method. Fish samples were cut muscles on both sides of the spine, and the frozen muscles sent one side to the cross-checking institutions

[a]Decay-corrected value to March 16, 2011

[b]Collected in Fukushima Prefecture in April 2011

[c]Collected in Tochigi Prefecture in June 2011

[d]Caught in offshore Fukushima Prefecture in December 2011

error is slightly larger, less than approximately ±5%. However, the author's values and other institutions' values are in good agreement within the range of counting error and sample heterogeneity, and the author concludes that the radioactivity values of the environmental samples discussed in this book are sufficiently reliable.

3.2.2 Soil Sampling

Most soil samples were collected from private sites. The owners approved these sample collections. 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 5 years following the FDNPP accident. Figure 3.1 shows the sampling sites. The sites were selected in the Tokyo metropolitan area and the Kanto district approximately 200 km from the FDNPP. Soil sampling was performed by the established IAEA method [7]. In the case of buildings and roads interspersed throughout urban regions, this method could not be applied to radioactive contamination urban areas. In general, the soil samples were obtained from the ground surface along roads where they were not subject to physical perturbation. The sampling sites were located on the flat ground surface by the roadsides. In many cases, they were hardly affected by the flow of rainwater and not covered with plants.

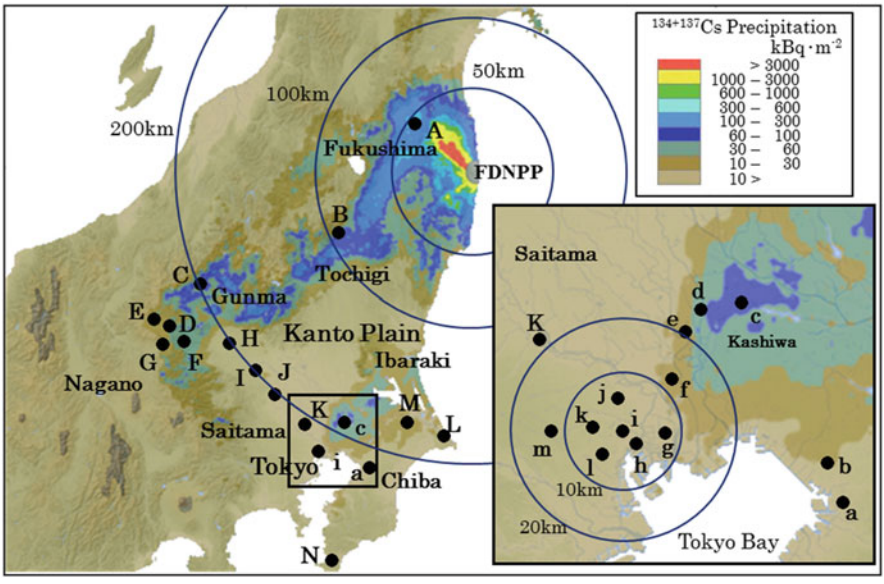

Fig. 3.1 Sampling sites of the soil sample in the Tokyo metropolitan area and Fukushima Prefecture. Geographic distribution of the $^{134+137}$Cs precipitation was cited from the MEXT airborne monitoring map published on November 11, 2011 ([14] a)

Photo 3.1 The sampling Site "i" in the Imperial Palace Outer Garden. A person is sunbathing on the lawn. Probably, he doesn't know that this place is radioactively contaminated (right side in the photograph above)

For example, the soil at Site "i" in Photo 3.1 was sampled in an area of 0.5 m × 1 m and obtained to a depth of 20 cm with the core sample, varying the sampling position slightly at each sampling period in this area. After sampling, the holes were refilled. There was grass, vegetation, etc., at this sampling location, but the soil was exposed at the ground surface. The ground was slightly sloped, so rainwater was assumed to flow gently over the ground surface. Pedestrians could walk on the sampling point. A surface soil up to 1 cm deep below the surface was measured using a ruler and taken from a 10 cm × 10 cm area. The core sediment 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 at 60 °C, and impurities such as vegetation detritus and rocks with diameters of 1 mm or more were removed before their radioactivity was measured.

The inventory of radionuclides in the ground surface was calculated, assuming a soil density of 1.3 g/cm^3 in compliance with the method of the MEXT of Japan [8] because the accurate evaluation of soil density is difficult. The inventories of the radioactively contaminated soil had been estimated in many previous reports using this value. The author measured the density of several soils, and the values were approximately 1.0–1.2 g/cm^3. This difference occurred because soil often contains organic substances such as humus and plant material. If the value of 1.3 g/cm^3 given by MEXT is used for inventory calculation, the soil density is slightly overestimated, but the use of this value is considered safe when radiation exposure from the soil to residents occurs.

3.3 Geographic Distribution of Radioactive Cesium and Radiation Dose in Central Japan by MEXT Airborne Monitoring and Typical Gamma-Ray Spectra of Contaminated Soil

To discuss the radioactive contamination of the Tokyo metropolitan area caused by the FDNPP accident, it is necessary to understand the radioactive contamination of the soil by the contaminated radioactive plume released from the FDNPP reactors.

The spread of radioactive plumes in Japan is described in Chap. 2. During the advection-diffusion process of contaminated radioactive plumes, radioactive nuclides precipitated from the plume to the ground surface, which caused radioactive contamination of the soil. Regarding soil contamination in Japan due to the FDNPP accident, monitoring surveys using aircraft were carried out by the MEXT (Ministry of Education, Culture, Sports, Science, and Technology) immediately after the accident. In these monitoring surveys, the air doses 1 m from the surface and the precipitation amounts of ^{134}Cs and ^{137}Cs were estimated. The earliest publicly available air dose measurements were obtained by airborne survey monitoring on March 25, 2011, at a distance of more than 30 km from the FDNPP [9]. Atmospheric ^{131}I and ^{137}Cs were measured by MOD (Ministry of Defense, Japan) aircraft on March 24, 2011, and the results were announced on March 25 [10].

In addition, the monitoring survey was conducted as a joint project between Japan and the United States [11, 12]. The US government's response to the FDNPP accident began with air dose measurements at the Yokota Base in Tokyo and the Embassy in Japan, as shown in Fig. 2.2. However, in Fukushima Prefecture around the FDNPP, before the start of joint airborne monitoring between Japan and the United States, the measurements of air filters (March 18, 58 km northwest of FDNPP, 4:15. March 19, 19 km north, 14:59) and soils (March 20, 66 km northwest of FDNPP. March 26, 19 km south) had begun [13]. The DOE (U.S. Department of Energy) participated in the airborne monitoring survey and data analysis from April 6 to July 2, 2011. Immediately after the accident, the monitored area was within 60 km of the FDNPP by DOE and 60–80 km by MEXT.

Over time, the areas of airborne monitoring surveys successively expanded. The radioactive contamination in Tokyo was monitored by an airborne monitoring survey from September 14–18, 2011. A radiation pollution map of the entire eastern part of Japan, including the Tokyo metropolitan area, obtained by this airborne monitoring was published by MEXT on November 11, 2011. Figure 3.2 shows a map of radioactive contamination in eastern Japan ([14] a). The air dose map at 1 m from the ground surface estimated simultaneously with the measurement of radioactive cesium deposition by the MEXT airborne monitoring is shown on the left in Fig. 3.3. Since the survey period varies from region to region, the estimated values of radioactive cesium deposition and air dose shown here have been corrected to the values for October 13, 2011 ([14] a). The right side of Fig. 3.3 is the result of a survey in 2018, which is the corrected value for November 15, 2018 ([14] b).

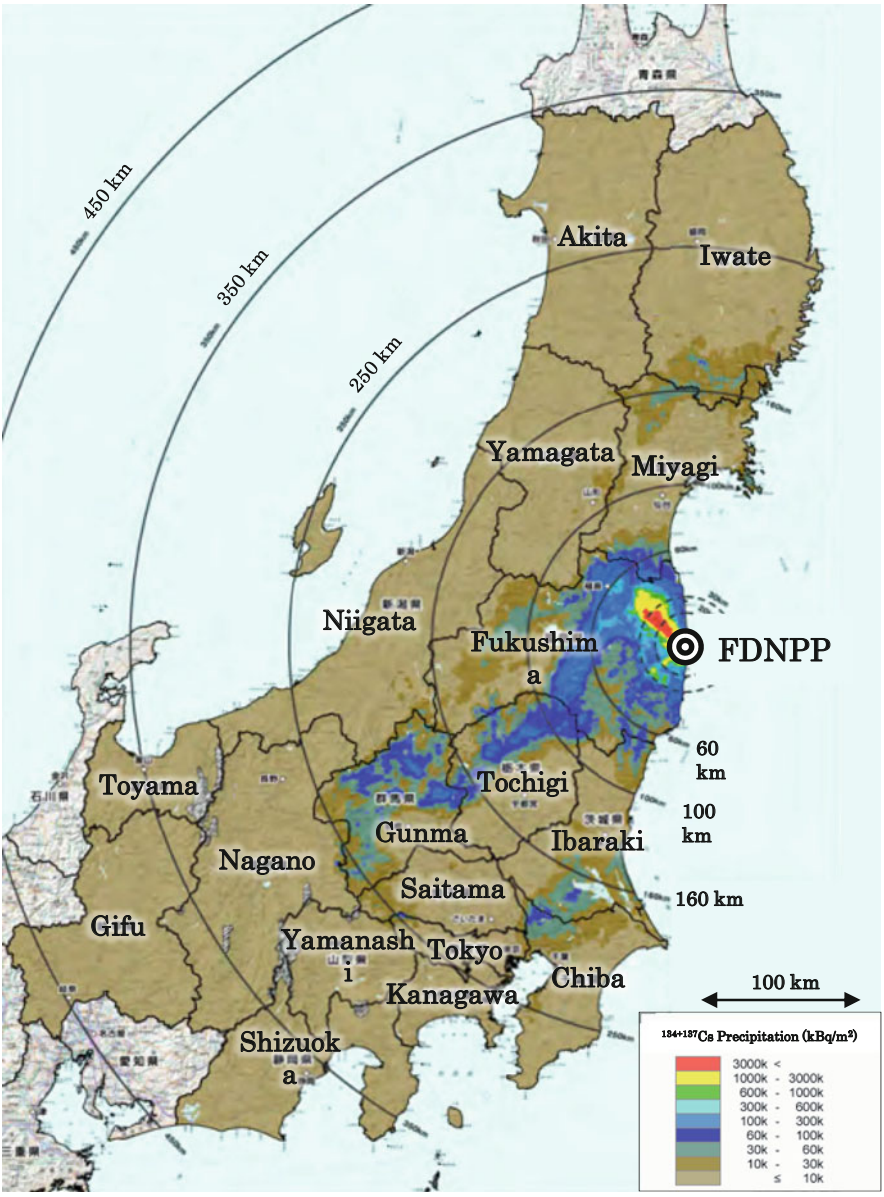

Fig. 3.2 Geographic distribution of the $^{134+137}$Cs precipitation in eastern Japan. This monitoring survey was carried out by the MEXT from April to October 2011, and the results were published in November 2011 ([14] a)

Since the FDNPP accident, an airborne monitoring survey of radioactive contamination on the surface of eastern Japan has been carried out by the NRC (Japanese Nuclear Regulatory Commission), which carried over from the work by

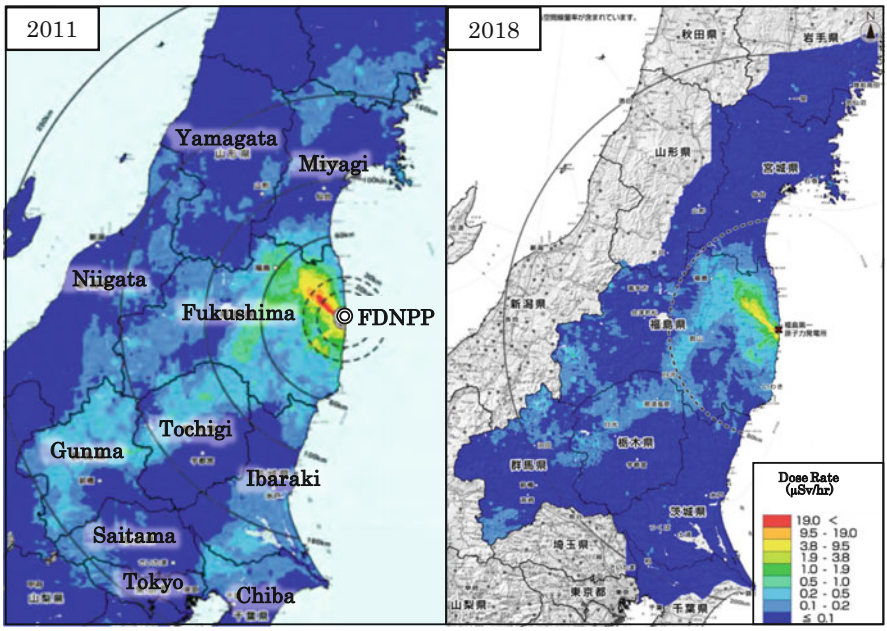

Fig. 3.3 Geographic distributions of the estimated air doses 1 m above the ground surface in eastern Japan. These figures show the results measured by the MEXT airborne monitoring in 2011 ([14] a) and 2018 ([14] b)

the MEXT. However, airborne monitoring surveys have not been conducted since 2012 in Tokyo, Saitama, Chiba, and Kanagawa prefectures, although the population is most concentrated and radioactive contamination is still high in these areas. The response of the Japanese government, which has not continued to monitor the actual state of radioactive contamination in the central area of Japan, is not appropriate from the viewpoint of assessing radiation exposure for residents.

As evident from the geographic distribution of radioactive cesium precipitation and air dose, the contaminated radioactive plumes released from the FDNPP reactors exposed almost all areas within 60 km of the reactor to severe radioactive contamination. In an area approximately 50 km long and 20 km wide from the periphery of the reactor, the precipitation amount of $^{134+137}$Cs was greater than 1000 kBq/m^2, and the air dose exceeded 9.5 μSv/h. The annual dose in the area shown in red on the left in Fig. 3.3 exceeded 166 mSv. The radioactive plume that caused radioactive contamination in the Tokyo metropolitan area was discharged from the reactor and then moved northwest, causing radioactive contamination of the ground surface. This plume changed direction near the border between Fukushima and Miyagi prefectures and moved southwest along the mountainous area that stretches to the central part of eastern Japan. As a result, large parts of Tochigi and Gunma prefectures were also radioactively contaminated. The radioactive contamination of Tokyo was caused by plumes migrating south from the FDNPP reactors. This

radioactive plume also caused radioactive contamination in the densely populated areas of southern Ibaraki Prefecture and northern Chiba Prefecture.

The right side of Fig. 3.3 is the newest airborne monitoring chart available. Although seven and a half years had passed since the accident, the figure illustrates that radioactive contamination continued even then in a wide range of eastern Japan when this map was made. Typical radionuclides released from the FDNPP reactors are ^{131}I, ^{134}Cs, and ^{137}Cs, but ^{131}I disappeared entirely due to radiative decay. The amount of ^{134}Cs decreased to 10% of the initial level, and ^{137}Cs decreased to approximately 85%. Although nuclides with short half-lives have disappeared, long-lived nuclides such as ^{137}Cs remain, so radioactive contamination in eastern Japan is thought to have continued for a while, as shown by this map.

Figure 3.4 presents the gamma-ray spectra of soils in Fukushima Prefecture and Tokyo contaminated by radioactive plumes released from the FDNPP reactors. The earliest soil sample obtained by the author was collected from a schoolyard in Fukushima City approximately 60 km from the FDNPP 8 days after the FDNPP accident and measured on March 22. The peaks of ^{132}Te with a half-life of 3.25 days and its daughter nuclide ^{132}I (half-life: 2.3 h) were still clearly detected. ^{136}Cs, with a half-life of 13.2 days, which is generated in small amounts by fission, was also detected; ^{136}Cs is produced by the (n, p) reaction of ^{138}Ba and the (n, α) reaction of ^{139}La. Although ^{140}Ba with a gamma-ray energy of 30.0 keV was detected, this nuclide is a fission product with a half-life of 12.8 days. At the same site in the schoolyard shown in Fig. 3.4a where decontamination work had not yet been performed, ^{132}Te and ^{132}I were not detected in the soil collected on April 27. Nevertheless, a small peak of ^{136}Cs is still visible in Fig. 3.4c.

Figure 3.4b is a gamma-ray spectrum of the washing sludge from a rescue helicopter collected 11 days after the accident. The flight history of the helicopter is unknown, but the radioactivity intensity is much higher than that of the soil in Fig. 3.4a. The helicopter may have flown over the FDNPP reactors or into a radioactive plume. In other words, this helicopter was highly radioactively contaminated, suggesting that radionuclides were still floating at extremely high concentrations around the DFNPP at this time. Furthermore, these measurements also indicate that radioactive plumes were continuously released from the collapsed FDNPP reactors.

The first soil sample in Tokyo was collected on April 10, 2011. The gamma-ray spectrum of the surface soil from the Imperial Palace Outer Garden in Tokyo is shown in Fig. 3.4d. Short-lived nuclides ^{132}Te and ^{136}Cs were detected even though approximately 1 month had passed since the accident. However, the significant radionuclides contaminating Tokyo soil were ^{131}I, ^{134}Cs, and ^{137}Cs. As shown in Fig. 2.3c, these nuclides carried from the FDNPP by the radioactive plumes that were transported on March 15–16 and March 21–22 were precipitated to the surface by the rain that fell at the time. Thus, the soil in Tokyo is estimated to have been radioactively contaminated because of the washing processes induced by rain. The temporal changes in radioactive cesium intensity in the soil at this site are shown in Fig. 3.6.

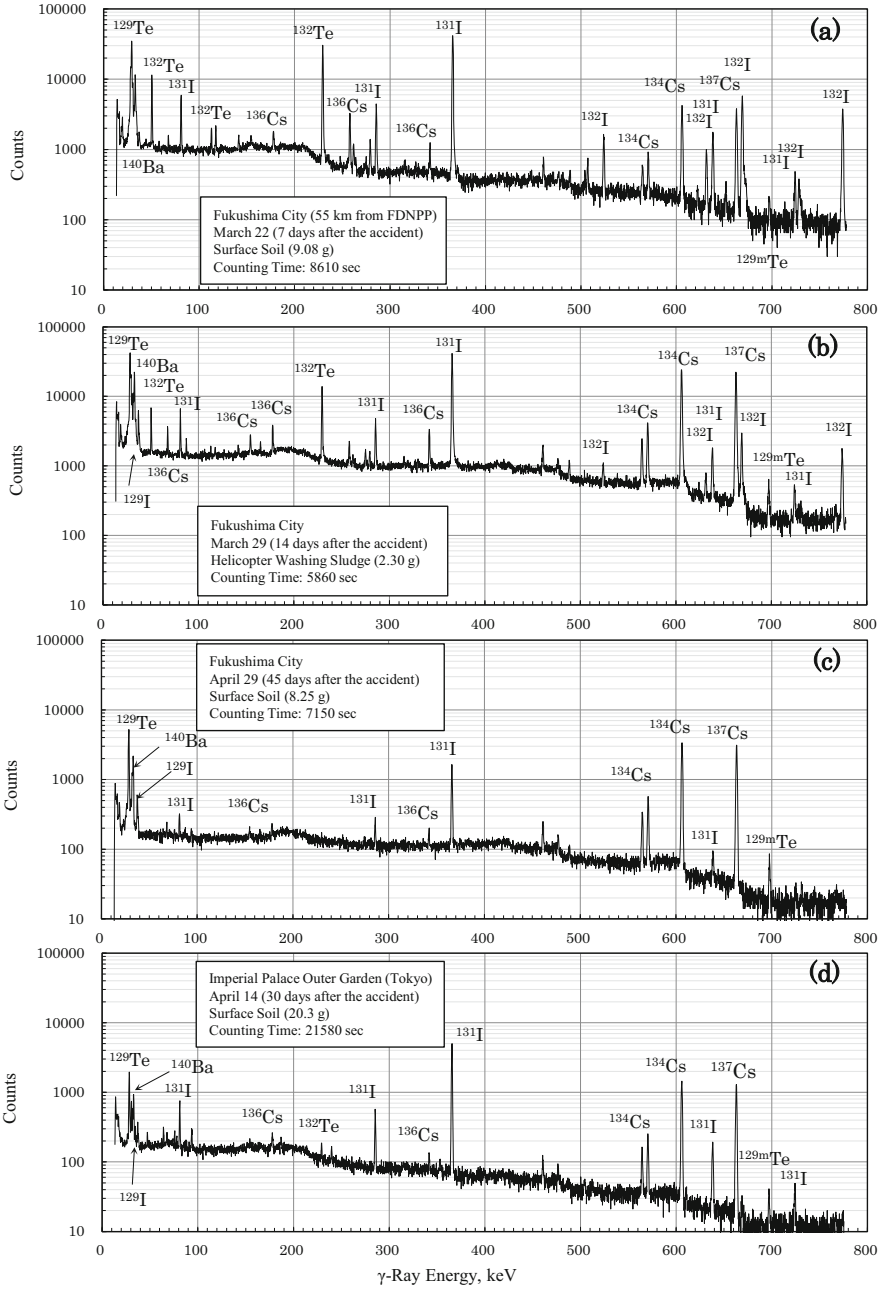

Fig. 3.4 Gamma-ray spectra of the surface soil in Fukushima City and central Tokyo immediately after the FDNPP accident. (**a**) Surface soil in the school garden collected 4 days after the accident. One week has passed before measurement, but the short half-lives fission product ^{132}Te ($T_{1/2}$ = 78 h) and its daughter ^{132}I ($T_{1/2}$ = 2.3 h) found. Neutron capture nuclide ^{136}Cs ($T_{1/2}$ = 13.2 days) with was also detected. (**b**) Collected from the rescue helicopter washing sludge. Probably the helicopter was flying over the FDNPP. The detection of short half-lives nuclides

3.4 Uniformity of Local Distribution of Radioactive Cesium Precipitated on the Ground Surface

In airborne monitoring, the aircraft flew with a trajectory width of approximately 3 km and an altitude of 150–300 m [14]. Therefore, the measured values in the radioactive contamination map of eastern Japan estimated by the MEXT and the NRA (Nuclear Regulation Authority, Japan) must be considered to indicate the average value in a region with a diameter of 300–600 m. In other words, this airborne monitoring has a large blind spot for the monitoring area. Such airborne monitoring is useful for analyzing the distribution and behavior of radioactive contamination in a broad area such as forests and cultivated land. However, cannot be considered suitable for analyzing complex terrains such as urban areas and residential areas. Immediately after the FDNPP accident, simple radiation surveys were performed by citizens in various parts of Japan. As a result, local highly contaminated radioactive areas called hot spots are found in various places throughout Japan. Such hot spots are challenging to detect by examining the local radioactive contamination distribution obtained by airborne monitoring. Therefore, the heterogeneity of radioactive cesium precipitated on the ground surface of Tochigi Prefecture through which the radioactive contamination plume passed is evaluated.

The monitoring site is an uncultivated paddy field at an elevation of 497 m in Nasu-Shiobara City, 110 km west-southwest of the FDNPP. On the north and east sides not far from the monitoring field are mountains. To the west and south are broad plains. Around the soil sampling area are paddy fields and pastures with houses scattered between them. Therefore, the wind is considered unlikely to have been perturbed by the buildings and trees as radioactive cesium precipitated from the plume to the ground surface. If radioactive cesium in the plume is deposited on the surface as soluble cations or their compounds, it is immediately absorbed by clay minerals in the soil. Therefore, if the soil contaminated with radioactive cesium is not physically mixed or moved, the distribution of radioactive cesium in the soil reflects the cesium distribution in the plume.

Figure 3.5 shows the sampling points at the monitoring site. Core samples with a depth of 5 cm were collected from five locations in the middle of an uncultivated paddy field of approximately 1000 m^2. According to the MEXT airborne monitoring, the inventory of $^{134+137}Cs$ at this site is estimated to be 100–300 kBq/m^2. Table 3.2 shows the analytical results for ^{134}Cs and ^{137}Cs in the soil collected from each point. The average radioactivity intensity at five points is 1480 ± 391 Bq/kg for ^{134}Cs and 1600 ± 457 Bq/kg for ^{137}Cs. The inventory of $^{134+137}Cs$ corrected for decay on October 13, 2011, calculated from this value, is was 189 ± 52 kBq/m^2, which agrees well with the value estimated by the airborne

Fig. 3.4 (continued) means that radionuclides continued to release from the reactors. (**c**) Collected at the same as site (**a**) approximately 6 weeks after the accident. (**d**) Collected at central Tokyo. High activities of ^{131}I were still detected. The date is the measurement date

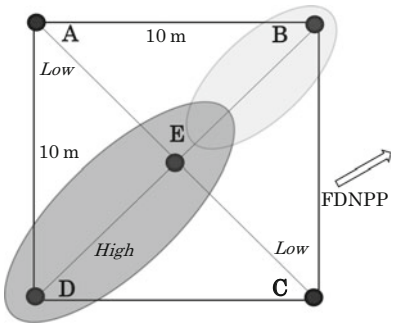

Fig. 3.5 Uniformity of the distribution of radioactive cesium precipitated from the plume. The observed site is the uncultivated paddy field in Nasushiobara City, Tochigi Prefecture. There is 110 km southwest of the FDNPP, and an elevation is 497 m

monitoring. However, the relative standard deviation of the $^{134+137}$Cs activity at these five sites is ±27.6%, which is much larger than the Ge detector counting error of 0.7–1.2% for this soil sample. The distribution of radioactive cesium at Points B, E, and D along the advection course of the radioactive plume is more significant than the average, and it is smaller at Points A and C. In particular, the radioactive cesium intensity at Point A is 40% smaller than the average value. Most likely, the radioactive plume released from FDNPP migrated in the direction from Point B to Point D. There is no clear correlation between the distribution of radioactive cesium and the soil particle size. Such a sizeable non-uniform distribution of radioactive cesium in a narrow area of not more than 10 m^2 suggests non-uniformity in the radioactive plume from which this radioactive cesium was transported. Nasushiobara City is more than 100 km from the FDNPP. However, it is highly likely that radioactive cesium in the plume advancing to this monitoring site retained the non-uniformity distribution released from the reactors.

The formation of hot spots in the radioactively contaminated zone must also take into account the process of radionuclide migration and enrichment after precipitation. Nevertheless, it is unlikely that the radionuclides adsorbed on the soil immediately after contamination can quickly move and be redistributed elsewhere. If the heterogeneity in the advection process of the radioactive plume is preserved in the distribution of radionuclides precipitated on the ground surface, it may be necessary to reconsider the survey method for assessing surface radioactive contamination. The secondary redistribution of radioactive contamination can be predicted by the physical and chemical properties of radionuclides and environmental conditions. Nevertheless, if a radionuclide precipitated from the plume and its distribution was non-uniform, then a sizeable contaminated area needs to be monitored with excellent point spacing. In the FDNPP accident, such a monitoring survey by the Japanese government could not be carried out.

Table 3.2 Uniformity of the distribution of radioactive cesium precipitated on the surface soil in Nasushiobara City, Tochigi Prefecture

Sampling date	Core depth, cm	Sampling point[a]	Average particle size, μm	^{134}Cs			^{137}Cs			$^{134+137}$Cs	
				Activity, Bq/kg	Deviation from average, %	RSD, %	Activity, Bq/kg	Deviation from average, %	RSD, %	Activity, Bq/kg	RSD, %
2011/6/4	0–5	B	9.6	1610	9.2	26.4	1710	6.9	28.6	3320	27.5
		E (Center)	12.2	1740	18.0		1950	21.9		3690	
		D	14.7	1830	24.1		2020	26.3		3850	
		C	8.0	1330	−9.8		1450	−9.4		2780	
		A	9.3	863	−41.5		891	−44.3		1750	
		Average	10.8 ± 2.7	1480 ± 391			1600 ± 457			3080 ± 847	
		Average inventory[b], kBq/m^2		96.2 ± 25.4			104 ± 29.7			200 ± 55.1	

[a]Sampling sites are shown in Fig. 3.5
[b]According to the MEXT airborne monitoring shown in Fig. 3.2 ([14] a), the $^{134+137}$Cs precipitation at this area 100–300 kBq/m^2

3.5 Vertical Distribution of Radionuclides Precipitated in the Soil

Many papers have been published on the adsorption of radionuclides on soil particles and clay minerals (References 19–30 in Chap. 1). The radionuclides are fixed in the soil and then move in the depositional layer because the clay particles adsorb the radionuclides released to the environment. However, despite many studies on the vertical mixing of aquatic sediments [15–30], research on the mixing of terrestrial soil has not progressed [31–35]. In the FDNPP accident, radioactive nuclides were precipitated onto the ground surface from the advecting radioactive plume in the atmosphere, causing radioactive contamination. To cope with such contamination, it is necessary to understand the adsorption behavior of radionuclides in soil and their migration and mixing processes. Information is also necessary regarding the decontamination of soil containing radionuclides. Furthermore, radioactive cesium deposited on the ground surface in a spike form may be an excellent tracer for elucidating the process of vertical mixing of terrestrial soil. Of course, it is also useful for evaluating the mixing parameters of aquatic sediments as shown in Figs. 2.4 and 6.9.

Chapter 6 discusses the transport and accumulation processes of radionuclides in Tokyo Bay and its inflowing rivers. In this case, it is also essential to understand the vertical mixing of aquatic sediments and the transport of contaminated soil particles by rivers. Table 3.3 shows the vertical distribution of radionuclides in the soil after the FDNPP accident at several points. Site D is a pre-cultivated field of Andosols rich in organic matter on a plateau at an elevation of approximately 1000 m in Gunma Prefecture. The amount of radioactive cesium precipitated is not very large. Since it is a cultivated field, the porosity of the soil is high, but most ^{131}I, ^{134}Cs, and ^{137}Cs was present in the 0–1 cm layer at the surface. For more than 40 days after the radionuclides were deposited here, their vertical distribution was not affected by rainfall infiltration or physical soil mixing. In the forest, approximately 50 m from the cultivated field, radionuclides were adsorbed by fallen leaves and litter that covered the soil surface, and no radioactive contamination of the underlying soil was observed. In this forest, radioactive iodine and radioactive cesium adsorbed on fallen leaves and litter did not easily migrate in the vertical direction. The cultivated land and forest are only 50 m apart, but the concentrations and isotope ratios of radionuclides differed significantly between these areas. This difference may suggest the heterogeneous deposition of these nuclides from the radioactive plume as described in Sect. 3.4. Most of the trees in this forest are deciduous trees, and no new leaves had yet sprouted in mid-March when radionuclides arrived. As discussed in Chap. 7, radionuclides from the radioactive plume that passed through the forest may have been effectively trapped in the bark. Since radioactive cesium adsorbed strongly to the bark, it was expected not to readily move down to the ground surface due to rainfall. However, rain was falling when this area was exposed to the radioactive plume. Some of the radionuclides precipitated on the bark may have been washed away by rainwater and accumulated in fallen leaves.

Table 3.3 Vertical distribution of radionuclides in the soil of the Tokyo metropolitan area

Site	Description	Sampling date	Depth, cm	Activity, Bq/kg						Inventory[a], kBq/m^2		
				^{131}I	^{134}Cs	^{137}Cs	^{131}I	^{134}Cs	^{137}Cs	^{131}I	^{134}Cs	^{137}Cs
				Detected			Decay corrected[b]			Decay corrected[b]		
D	Farmland	2011/4/29	0–1	36.7	176	184	1630	183	184	21.2	2.38	2.39
			1–3	nd	nd	4.5	–	–	4.5	–	–	0.12
			3–5	nd	nd	nd	–	–	–	–	–	–
			5–10	nd	nd	nd	–	–	–	–	–	–
	Forest	2011/4/29	0–5[c]	267	4380	4730	12,900	4570	4740	516[d]	183[d]	190[d]
			6–7[e]	nd	nd	nd	–	–	–	–	–	–
B	Garden	2011/6/13	0–1	nd	6210	6810	–	6920	6860	–	90.0	89.2
			1–5	nd	2800	3120	–	3120	3140	–	162	163
			5–10	nd	80	84.5	–	89.1	85.1	–	5.79	5.53
			10–15	nd	nd	nd	–	–	–	–	–	–
			15–20	nd	nd	nd	–	–	–	–	–	–
i	Park	2011/4/10	0–1	2150	944	961	18,500	966	962	241	12.6	12.5
			1–3	nd	nd	nd	–	–	–	–	–	–
		2011/4/28	0–1	1320	1130	1170	53,700	1180	1180	698	15.3	15.3
			1–3	33.3	33.2	41.9	1360	34.6	42.0	35.4	0.90	1.09
			3–5	nd	nd	14.0	–	–	1.4	–	–	0.04
			5–10	nd	nd	nd	–	–	–	–	–	–
		2011/8/13	0–1	nd	106	121	–	122	122	–	1.59	1.59
			1–3	nd	8.0	14.2	–	9.2	14.3	–	0.24	0.37
			3–5	nd	4.7	5.2	–	5.4	5.2	–	0.14	0.14
			5–10	nd	nd	nd	–	–	–	–	–	–
		2013/5/1	0–2	nd	165	343	–	342	361	–	8.89	9.39
			2–5	nd	33.2	64.1	–	68.7	67.4	–	1.79	1.75
			5–10	nd	nd	5.2	–	–	5.5	–	–	0.14
			10–15	nd	nd	nd	–	–	–	–	–	–

		2013/12/31	0–2	nd	23.8	57.8	–	61.8	61.7	–	1.61	1.60
			2–5	nd	4.0	10.8	–	10.4	11.5	–	0.41	0.45
			5–10	nd	nd	nd	–	–	–	–	–	–
		2014/5/3	0–2	nd	45.7	123	–	133	133	–	3.46	3.46
			2–5	nd	4.0	12.3	–	11.7	13.2	–	0.46	0.52
			5–10	nd	2.1	6.3	–	6.1	6.8	–	0.40	0.44
			10–15	nd	nd	nd	–	–	–	–	–	–
		2014/8/20	0–2	nd	47.4	150	–	152	162	–	3.95	4.21
			2–5	nd	13.2	34.2	–	42.3	37.0	–	1.65	1.44
			5–10	nd	6.8	13.2	–	21.8	14.3	–	1.42	0.93
			10–15	nd	nd	nd	–	nd	nd	–	–	–
		2015/1/3	0–1	nd	215	704	–	787	769	–	10.2	10.0
			1–3	nd	95.2	325	–	349	355	–	9.07	9.23
			3–5	nd	10.7	34.0	–	39.2	37.1	–	1.02	0.97
			5–10	nd	5.3	16.8	–	19.4	18.4	–	1.26	1.20
			10–15	nd	nd	7.2	–	–	7.9	–	–	0.51
			15–20	nd	nd	nd	–	–	–	–	–	–
		2015/5/2	0–1	nd	97.3	354	–	399	390	–	5.19	5.07
			1–3	nd	19.0	68.8	–	77.8	75.7	–	2.02	1.97
			3–5	nd	9.9	38.8	–	40.6	42.7	–	1.06	1.11
			5–10	nd	11.9	46.2	–	48.8	50.9	–	3.17	3.31
			10–15	nd	3.5	15.4	–	14.3	17.0	–	0.93	1.11
			15–20	nd	nd	nd	–	–	–	–	–	–
		2016/1/3	0–1	nd	126	608	–	651	680	–	8.46	8.86
			1–3	nd	32.6	149	–	168	166	–	4.37	4.32
			3–5	nd	3.7	19.5	–	19.1	21.8	–	0.50	0.57
			5–10	nd	nd	8.6	–	–	9.6	–	–	0.62
			10–15	nd	nd	7.9	–	–	8.8	–	–	0.57
			15–20	nd	nd	nd	–	–	–	–	–	–

(continued)

Table 3.3 (continued)

Site	Description	Sampling date	Depth, cm	Activity, Bq/kg						Inventory[a], kBq/m^2		
				^{131}I	^{134}Cs	^{137}Cs	^{131}I	^{134}Cs	^{137}Cs	^{131}I	^{134}Cs	^{137}Cs
				Detected			Decay corrected[b]			Decay corrected[b]		
		2016/8/13	0–2	nd	46.3	252	–	295	286	–	7.67	7.44
			2–4	nd	4.2	21.7	–	26.8	24.6	–	0.70	0.64
			4–6	nd	nd	8.3	–	–	9.4	–	–	0.24
			6–10	nd	1.5	9.5	–	9.6	10.8	–	0.50	0.56
			10–15	nd	nd	5.1	–	–	5.8	–	–	0.38
			15–20	nd	nd	nd	–	–	–	–	–	–

[a]Calculated assuming a density of soil is 1300 kg/m^3
[b]Activities were decay corrected to March 16, 2011
[c]Litter and fallen leaves
[d]Calculated assuming a density of litter is 800 kg/m^3
[e]Soil

Site B is a hilly area in Tochigi Prefecture at an elevation of approximately 500 m. Surrounded by paddy fields and pastures are scattered private houses. The soil sampling site is in the yard of a private house where six people live, including older adults and children. The yard has a small flower bed and a place to hang laundry, as well as playground equipment for the children. Since sampling was conducted 3 months after the FDNPP accident, ^{131}I had already disappeared. However, because a small gamma-ray peak of 39.6 keV emitted from the fission product ^{129}I (half-life: 1.57×10^7 years) was observed in the spectrum, the yard was undoubtedly radioactively contaminated with radioactive iodine as well as radioactive cesium. High activities of radioactive cesium were observed in the surface soil at Site B. Unlike Site D, radioactive cesium was also detected in the 1–5 and 5–10 cm layers. When collecting the core samples of the soil, the core sampler was inserted into the ground from the soil surface where radioactive contamination was extreme, so that the upper radionuclides may have adhered to the inner wall of the sampler and been carried to the lower layer. Care must be taken because such a displacement phenomenon gives false results as if there were contamination in the deep layer. However, the inventory of radioactive cesium in the 5–10 cm layer at Site B is approximately 2.2% of the total inventory, much greater than that expected to be carried to the deeper layer by such displacement. Therefore, at Site B, radioactive cesium had penetrated the soil at least deeper than 5 cm. It was not possible to determine whether the process by which radioactive cesium was transported to the depths originated from the soil properties at this site or whether the soil was physically mixed by human activity.

Site "i" is the site discussed in Sect. 3.7. The core sediments were collected at the Imperial Palace Outer Garden in central Tokyo. This site is approximately 1.2 km east-northeast from the Japanese Parliament. Many people visit this park, making it difficult to collect samples. Visitors may walk on the sampling point. A feature of this location is that the amount of radioactive iodine precipitated was much more significant than that of radioactive cesium (see Fig. 4.2). The site was estimated to have been radioactively contaminated on March 21, 2011, but the first core sampling was on April 10. At this time, both radioactive iodine and radioactive cesium were present in the surface 0–1 cm layer on the ground. By the 28th, ^{131}I and ^{134}Cs had penetrated into the 1–3 cm layer and accounted for 5.1% and 5.9% of the total inventory, respectively. However, some of the ^{137}Cs penetrated below the 1–3 cm layer. Perhaps the ^{137}Cs in the deeper layer originated from global fallout. As shown in Fig. 3.6, at Site "i", the radioactive cesium concentration differed each time sampling was performed. Human activity is thought to have greatly influenced the distribution of radioactive contamination on the ground surface of this site. The core collected in 2015 was only 50 cm from that collected in 2011, but the vertical distributions of the two are very dissimilar. It can be seen that by 2015, the radioactive cesium had diffused to the 10–15 cm layer, but in the case of the 2016 core, most of it remained in the layer shallower than 5 cm (see Table 3.3).

Radionuclides that polluted Japan's land in the FDNPP accident were precipitated from the atmosphere to the ground surface from radioactive plumes. Then, the radionuclides were adsorbed and retained on the ground surface, but their

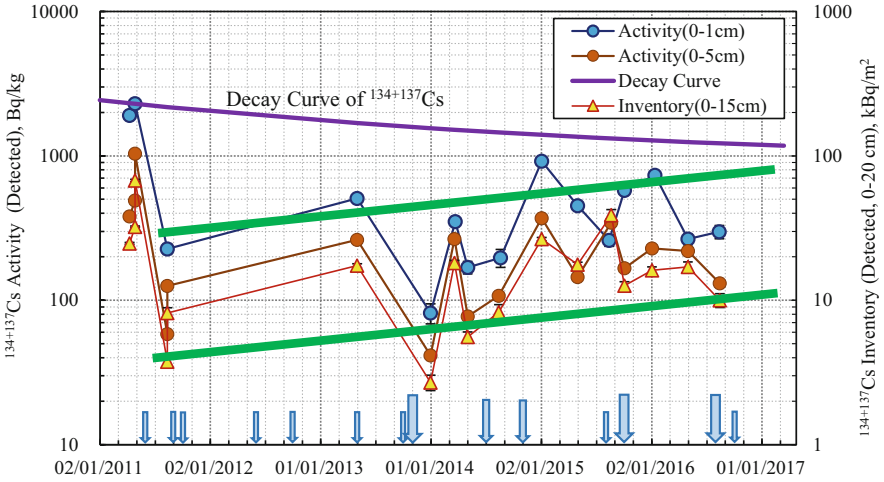

Fig. 3.6 Temporal changes in the concentration and inventory of $^{134+137}$Cs in the surface soil in the Imperial Palace Outer Garden. The radioactive decay curve of $^{134+137}$Cs is shown assuming that the activity of ^{134}Cs and ^{137}Cs are the same immediately after the accident. Vertical arrows showed the monthly rain precipitations. Small: over 200 mm. Medium: over 300 mm. Large: over 400 mm [48]

mechanism and behavior are very complicated. The vertical distributions of radionuclides in the soil and their changes significantly affect the behavior of radionuclides in the environment. Evaluating the vertical distribution of radionuclides in the soil is extremely important for management of radiation exposure and decontamination work for residents. If the inhomogeneity of the radioactive plume reflected the physical and chemical states of the radionuclides, its situation should have been recorded in the soil onto which they were precipitated. The soil itself has a heterogeneous composition and environmental form. Unfortunately, in Fukushima, decontamination work has been carried out without such evaluation of the soil characteristics; thus, it is challenging to manage radiation after decontamination work. As of 2020, the contaminated soil recovered from the decontamination work has been stored outdoors without proper protection. This neglect leads to a very high risk of providing a new source of environmental radioactive recontamination.

3.6 Contamination by Radioactive Nuclides in the Surface Soil in the Tokyo Metropolitan Area

Table 3.4 shows the concentrations and inventories of ^{131}I and radioactive cesium in soil samples collected at the sites in the Tokyo metropolitan area shown in Fig. 3.1 within 3 months after the FDNPP accident. Since the sampling dates differ for each site, the detected activities were corrected by radioactive decay to the values for

Table 3.4 Typical analytical results of the concentration and inventory of radionuclides in the soil collected in the Tokyo metropolitan area and Fukushima Prefecture

Prefecture	Site	Description	Sampling date	Taken, cm	Decay corrected activity[a], Bq/kg				Ratio for ^{137}Cs		Inventory[c], kBq/m^2		
											This work[a]		MEXT[b]
					^{131}I	^{134}Cs	^{137}Cs	$^{134+137}$Cs	^{131}I	^{134}Cs	^{131}I	^{134} +^{137}Cs	^{134} +^{137}Cs
Fukushima	A	Schoolyard	2011/3/19	0–1	122,000	13,900	14,100	28,000	8.65	0.986	1590	363	300–600
			2011/4/27	0–1	142,000	21,400	21,900	43,300	6.48	0.977	1850	563	
Tochigi	B	Pasture[e]	2011/6/4	0–1	43,600	11,100	11,300	22,400	3.86	0.982	566	290	60–100
		Garden	2011/6/13	0–1	24,500	3020	3150	6170	7.78	0.959	318	80.2	
		Farmland	2011/6/13	0–1	91,800	6730	6810	13,500	13.5	0.988	1190	176	
Gunma	C	Paddy field	2011/4/30	0–1	5640	1830	1850	3680	3.05	0.989	73.3	47.9	30–60
	D	Forest	2011/3/26	0–1	582	67.4	70.7	138	8.23	0.953	7.6	1.8	10–30
		Farmland	2011/4/29	0–1	1630	183	184	367	8.85	0.996	21.2	4.8	
				1–3	nd	nd	4.5	4.5	–	–	–	0.1	
	E	Farmland[g]	2011/4/29	0–1	4360	1760	1720	3480	2.53	1.02	56.7	45.1	
	F	Plateau forest[h]	2011/5/1	0–1	2050	824	810	1630	2.53	1.02	26.6	21.2	
	H	Road side	2011/4/28	0–1	3860	740	745	1490	5.18	0.993	50.1	19.3	<10
Nagano	G	Plateau forest[f]	2011/4/28	0–1	513	550	557	1110	0.920	0.986	6.7	14.4	<10
		Hotel garden	2011/4/28	0–1	3350	726	755	1480	4.44	0.962	43.6	19.3	
Ibaraki	L	Garden	2011/4/20	0–1	4850	225	237	462	20.4	0.949	63.1	6.0	<10
	M	Garden	2011/4/20	0–1	8420	105	108	213	78.1	0.977	109	2.8	10–30
Chiba	N	Garden	2011/4/10	0–1	582	59.7	69.2	129	8.41	0.863	7.6	1.7	<10
	a	Road side	2011/4/11	0–1	7020	672	672	1340	10.4	1.00	91.3	17.5	10–30
	b	Road	2011/4/11	0–1	12,100	2460	2510	4970	4.82	0.980	157	64.6	

(continued)

Table 3.4 (continued)

Prefecture	Site	Description	Sampling date	Taken, cm	Decay corrected activity[a], Bq/kg				Ratio for ^{137}Cs		Inventory[c], kBq/m^2		
											This work[a]		MEXT[b]
					^{131}I	^{134}Cs	^{137}Cs	$^{134+137}$Cs	^{131}I	^{134}Cs	^{131}I	^{134} +^{137}Cs	^{134} +^{137}Cs
	c	Garden	2011/5/19	0–2	40,700	5300	5330	10,600	7.64	0.994	1060	276	60–100
		Road sludge	2011/6/11	0–1	114,000	33,900	33,900	67,800	3.36	1.00	1480	881	
		Road side	2011/6/11	0–1	28,500	17,700	17,700	35,400	1.61	1.00	371	460	
	d	Road side	2011/5/19	0–1	26,100	3210	3190	6400	8.18	1.01	340	83.2	
	e	Garden	2011/5/19	0–2	25,200	1060	1080	2140	23.3	0.981	655	55.6	10–30
Tokyo	f	Road side	2011/5/19	0–1	56,900	3470	3410	6880	16.7	1.02	740	89.4	10–30
	g	Garden (1)	2011/4/16	0–1	25,700	1630	1610	3240	16.0	1.01	334	42.1	
			2011/4/27	0–1	77,500	3560	3600	7160	21.5	0.989	1010	93.1	
		Garden (2)	2011/5/25	0–1	27,700	1710	1740	3450	15.9	0.983	360	44.8	
	h	Road side	2011/4/10	0–1	10,100	588	580	1170	17.4	1.01	131	15.2	<10
	i	Garden[d]	2011/4/10	0–1	18,500	966	962	1930	19.2	1.00	240	25.1	
			2011/4/28	0–1	53,700	1180	1180	2360	45.5	1.00	698	30.6	
				1–3	1360	43.6	62.5	106	21.8	0.698	35.0	3.0	
	j	Garden	2011/4/10	0–1	20,000	644	683	1330	29.3	0.942	259	17.2	
	k	Road side (1)	2011/6/2	0–2	18,200	1780	1760	3540	10.3	1.01	474	92.2	
		Road side (2)	2011/6/2	0–2	7780	475	473	948	16.4	1.00	202	24.6	
		Road side (3)	2011/6/2	0–2	11,600	514	536	1050	21.7	0.959	302	27.2	
	l	Road side	2011/6/2	0–2	11,100	1230	1220	2450	9.10	1.01	290	63.8	
	m	Garden	2011/5/19	0–2	6740	432	442	874	15.2	0.976	175	22.8	

Saitama	I	Rode side	2011/4/28	0–1	nd	151	154	305	–	0.979	–	4.0	<10
	J	Road side	2011/4/28	0–1	3130	204	207	412	15.1	0.986	40.7	5.4	
	K	River bank	2011/4/10	0–1	6130	248	252	500	24.3	0.981	79.6	6.5	
							Average		14.3 ± 13.9	0.978 ± 0.053			

Detection limit: ^{131}I <2 Bq/kg, ^{134}Cs and ^{137}Cs <0.6 Bq/kg
nd: not detected
[a]Decay-corrected value to March 16, 2011
[b]Airborne monitoring results by the MEXT ([14] a)
[c]Calculated assuming 1300 kg/m^3 for the soil density
[d]Measurements continued from April 10, 2011, to August 16, 2016. The results are shown in Fig. 3.6
Elevation [e]500 m, [f]1430 m, [g]1100 m, [h]1390 m

March 16, 2011. The background of Fig. 3.1 is the geographic distribution of $^{134+137}$Cs precipitation obtained by airborne monitoring. Figure 3.2 has the same background as Fig. 3.1. This map was released on November 11, 2011, by the MEXT. The distribution map of the pollution by radioactive cesium shown here is called a revised version. The MEXT released a pollution map of the same area on October 21, 2011, but a municipality where the nuclear plant was installed protested that the pollution level was too high. Therefore, the Japanese government changed the processing method for the background radioactivity in creating an airborne monitoring map and reprocessed the data to reduce the apparent contamination concentration. The original contamination map released on October 21 is shown in Fig. 9.5. The change in the background processing method decreased the radioactive cesium inventory by dozens of kBq/m^2. This treatment also significantly reduced the reliability of the airborne monitoring data.

The author's measurement sites do not cover the entire Tokyo metropolitan area. However, the measured data showed that the geographic distribution of radioactive cesium contamination was in good agreement with the MEXT pollution map published at a later date. Radioactive cesium was detected in surface soil at all stations sampled between March 19 and June 13, 2011. Despite a very short half-life of 8.04 days, ^{131}I was also discovered in all surface soils except at Site "I" in Saitama Prefecture (see Fig. 3.1). This result suggests that the radioactive contamination of the ground due to the FDNPP accident spread widely throughout the Tokyo metropolitan area at the beginning of the accident.

The inventories of samples collected to a surface depth of only 1 cm may have been estimated slightly lower than the actual values, considering the vertical distribution of radionuclides in soil shown in Table 3.4. However, it is believed that the difference is at most within 5%. In many areas shown in Table 3.4, the $^{134+137}$Cs inventory estimated by MEXT from airborne monitoring is lower than the value measured in this study. In particular, the tendency is remarkable in the urban areas of Chiba Prefecture and Tokyo. This difference in the inventories suggests that the estimation of radioactive contamination levels by airborne monitoring is limited in densely populated areas. The likely underestimation of the inventories in an urban area by airborne monitoring affects local and long-term estimates of external exposure for the inhabitants.

3.6.1 *Inventory of ^{131}I in the Tokyo Metropolitan Area*

The ^{131}I inventory in the Tokyo metropolitan area converted to the value on March 16, 2011, was nd (not detected: <0.03 kBq/m^2)—1480 kBq/m^2. Since the inventory of Fukushima City (Site A) was 1590–1850 kBq/m^2, ^{131}I deposition was smaller in all areas of the Tokyo metropolitan area than in Fukushima City. However, the ^{131}I inventories at Site B in Tochigi Prefecture, Site "c" in Chiba Prefecture, and Site "g" in Tokyo exceeded 1000 kBq/m^2. Site B is close to Fukushima, where significant plumes released from the FDNPP have passed, and it is easy to deduce that large

amounts of radioactive material were deposited. Site "c", on the other hand, features an area where hot spots were formed immediately after the accident. Although this site is approximately 200 km from the FDNPP, its inventory is higher than that of the surrounding area. Later, MEXT airborne monitoring (Fig. 3.2) confirmed that the area was a highly radioactively contaminated zone. In Tokyo, Site "g" in the east and Sites "I" and "j" in the city center showed high inventories. The inventories in central Tokyo were higher than those at other sampling sites in the surrounding area. This geographic distribution of ^{131}I in the Tokyo metropolitan area provides evidence that the FDNPP accident caused high concentrations of ^{131}I to develop central Tokyo.

The radioactive iodine detected in tap water supplied from the Kanamachi water purification plant in Tokyo on March 23, 2011, was the first event of radioactive iodine contamination in Tokyo from the FDNPP accident. In Japan at this time, the standard for radioactive iodine in drinking water was less than 300 Bq/kg based on the food and drink intake limit set by the Japan Nuclear Safety Commission (currently the NRA: Nuclear Regulatory Authority, Japan). In addition, the Food Sanitation Law of the Ministry of Health, Labor, and Welfare specified a provisional standard value of 100 Bq/kg in beverages for infants. The radioactive iodine concentrations of tap water at this water purification plant greatly exceeded this value. The radioactive iodine concentration of tap water at the Kanamachi water purification plant, announced by the Tokyo metropolitan government, were 210 Bq/kg on March 22 ([36] a) and 190 Bq/kg on March 23. No radioactive cesium values were listed in these announcements ([36] b). The Edogawa River water is used by the Kanamachi water purification plant to produce tap water. The Edogawa River is discussed in Chap. 6 of this book as a source of radioactive contamination for Tokyo Bay. Rain fell on the 21st in the Tokyo area. As predicted by Dr. Ishida in Fig. 2.1, radioactive plumes with extremely high ^{131}I concentrations had invaded eastern Tokyo and northern Chiba Prefecture on the 21st. Most likely, the Tokyo Metropolitan Government Bureau of Waterworks used the polluted raw water from the Edogawa River at this time. The radioactive materials released in the FDNPP accident were being transported to Tokyo, and the people of Tokyo learned this fact for the first time through the press. Site "f" was located approximately 1 km from the Kanamachi water purification plant. Its ^{131}I inventory was 740 kBq/m^2. This value is approximately half that in Fukushima City, but it is estimated that ^{131}I at the same level as Fukushima was deposited in the catchment area of the Kanamachi water purification plant. This information raised worries about the effects of radioactive contamination of tap water due to the FDNPP accident. Since March 28, ^{131}I has not been detected in the tap water at this water treatment plant (nd: $<$20 Bq/kg) ([36] c), but the concern about radioactive contamination of tap water due to the FDNPP accident has not disappeared.

3.6.2 Inventories of ^{134}Cs and ^{137}Cs in the Tokyo Metropolitan Area

The geographic distribution of radioactive cesium inventories also depended heavily on the soil sampling site. As with ^{131}I, its broad distribution was similar to that determined by the MEXT airborne monitoring. However, the ratio of ^{131}I to radioactive cesium inventories varied significantly from sampling point to sampling point. This difference is discussed in detail in Chap. 4.

The $^{134+137}Cs$ inventory at Site A in Fukushima City showed good agreement between the MEXT estimates and the measured values. However, farther south in Tochigi Prefecture, the measured value was significantly higher than the estimated MEXT value. In Gunma, Nagano, Ibaraki, and Saitama prefectures, the estimated MEXT values, and the measured values were almost the same. Many of these sampling areas were in the countryside. On the other hand, in Chiba Prefecture, the measured values at all sites except Site N were much higher than the MEXT estimates. The measured values at all sites in Tokyo were also greater than the estimated MEXT values.

The $^{134+137}Cs$ inventory at site "c" in Kashiwa City, Chiba Prefecture was 276–881 kBq/m^2, which was almost the same level as 363–563 kBq/m^2 in Fukushima City. Site "c" was located on the side of the road next to a park so that it may have been affected by the trees planted in the park. It is highly probable that rainwater containing massive amounts of radionuclides that had been transported down tree trunks was flowing into the road sludge with the highest inventory of $^{134+137}Cs$ at 881 kBq/m^2. This site also had an extremely high ^{131}I inventory. However, in the roadside soils approximately 3 m away, the $^{134+137}Cs$ inventory was only halved, despite ^{131}I decreasing to one-quarter of that in the road sludge. In the garden soil in the park, ^{131}I decreased to approximately 70% of the value in the road sludge, while $^{134+137}Cs$ decreased significantly to approximately 30% of the value in the road sludge.

Thus, the concentrations and inventories of radioactive iodine and radioactive cesium fluctuated in a complicated manner, even at very closely spaced sampling points. Since various factors are presumed to have been involved in the behavior of these radionuclides precipitated from plumes to the ground surface, it would be challenging to generalize their behavior for each radionuclide. However, estimating the behavior of radionuclides from the environmental conditions at each sampling point is imperative, and determining the environmental conditions at the sampling points and collecting samples that meet the objectives of the analysis are also essential. Unnecessarily collecting and analyzing samples is not scientific.

The $^{134+137}Cs$ inventory in central Tokyo was lower than that in northern Chiba. However, it a value of approximately 100 kBq/m^2 was observed in eastern Tokyo adjacent to this highly polluted area. Sites "g"–"l" are in not only the center of Tokyo but also the center of legislation, administration, and economics in Japan. It is incredibly alarming that such important areas of Japan are exposed to radioactive contamination. The high contamination by ^{131}I attenuates rapidly over time.

However, radioactive cesium, especially ^{137}Cs, has a long half-life of 30.1 years and will remain in this area for a long time. Radioactive cesium deposited on office buildings and infrastructure is likely to be washed away by rain and re-adsorbed and fixed in the surrounding soil. Attention must be paid to the behavior of radioactive cesium derived from urban structures.

In the immediate aftermath of the FDNPP 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. The decay-corrected inventories of ^{131}I and $^{134+137}$Cs on March 16, 2011, in these areas, were 35–1480 kBq/m^2 and 15.2–881 kBq/m^2, respectively, which are almost the same levels as those in Fukushima City. As described in Chap. 5, the radioactive cesium concentration in the sludge incineration ash was higher in the urbanized eastern part of Tokyo and was a correlated with changes in rainfall in this area. It is possible that the radioactive cesium precipitated on concrete and asphalt is more likely to be discharged than that on the soil. These results suggest that the Tokyo metropolitan area continues to be affected by radioactive contamination caused by the FDNPP accident. The densely populated central regions of Japan have been exposed to massive radiation contamination. It is unprecedented worldwide that the FDNPP accident has caused large-scale radiation exposure to the general public. One hopes that long-term health impact assessments and monitoring will be conducted not only for Fukushima residents but also for residents of the Tokyo metropolitan area.

3.7 Temporal Changes in Radioactive Cesium in Park Soil from Central Tokyo

The soil at Site "i", located in the Imperial Palace Outer Garden in the center of Tokyo, was continuously surveyed for radioactivity concentrations and inventories. As shown in Photo 3.1, the soil collection site is located in a part of the park where people can freely move around. The first sampling took place on April 10, 2011, when ^{131}I (2150 Bq/kg) and $^{134+137}$Cs (1910 Bq/kg) were detected with high radioactivity, but ^{131}I disappeared quickly due to radioactive decay. However, as shown in Table 3.3, most of the radioactive cesium accumulated on the soil surface throughout the monitoring period and was not detected in layers deeper than 15 cm. Figure 3.6 shows the temporal changes in the average $^{134+137}$Cs concentrations in the layers 1 and 5 cm below the surface. The inventory of $^{134+137}$Cs shows the cumulative values for layers up to 15 cm deep where $^{134+137}$Cs was detected. The decay curve of radioactive cesium, assuming that the amounts of ^{134}Cs and ^{137}Cs deposited immediately after the accident were the same, is also shown in Fig. 3.6. On April 28 and August 13, 2011, 2 cores were collected from the same site, approximately 10 cm apart, to verify the accuracy of the inventory. One of these cores was used to measure the vertical distribution, and the analytical results were expressed as vertically cumulative values to calculate the inventory. The other core was sliced

to a thickness of 5 cm and analyzed for the inventory. The $^{134+137}$Cs inventories of these 2 cores, standardized on the date of collection, were 32.6 and 67.5 kBq/m^2 on April 28 and 4.1 and 8.2 kBq/m^2 on August 13. These differences in inventories for adjacent core samples are attributed to the fact that the horizontal distribution of radioactive cesium infiltrated into the soil results in highly localized contamination, rather than to differences in sampling methods. Most likely, this speculation seems reasonable. Furthermore, in the cores collected on March 23, 2014, on August 14, 2015, and on May 1, 2016, the concentrations of $^{134+137}$Cs in the 0–1 cm layer and the 0–5 cm layer were close or reversed. In these cores, radioactive cesium penetrated deep into the soil without being retained on the soil surface. It was not possible to determine whether such distributions of radioactive cesium were due to local physical vertical mixing of the soil at the sampling point, or whether ultrafine contaminated particles in the sediment had advected deeper. The spatial-temporal distribution of radioactive cesium in the soil of urban areas is discussed below.

From April 10–28, 2011, after the FDNPP accident, the activities and inventories of ^{131}I and $^{134+137}$Cs increased sharply. If this increase was not the result of heterogeneous distribution of the radionuclides in the soil, the diffusion of radionuclides from the FDNPP to the Tokyo area was probably ongoing [37]. However, on August 13, the $^{134+137}$Cs radioactivity decreased to approximately 10% of its initial value, and ^{131}I was not found. That is, it can be assumed that the new emission of radionuclides from the FDNPP reactors to the Tokyo area had subsided by that time. Chapters 5 and 7 emphasize the phenomenon that the concentrations of radioactive cesium that had polluted the environment suddenly decreased immediately after the FDNPP accident. The changes in radioactive cesium concentrations in the sludge incineration ash at the Tokyo Water Reclamation Center, which are described in Chap. 5, also decreased sharply to approximately one-tenth of the initial value in the 9 months after the accident [38]. However, the concentration of $^{134+137}$Cs in the park soil decreased sharply and seemed to have increased gradually since August 2011. As shown in Table 3.3, no significant penetration of radioactive cesium into the subsurface was observed over time. Presumably, radioactive cesium that was initially lost from the park soil in Tokyo was not an ionic species that is absorbed by clay minerals and fixed to the soil. Perhaps it was present as an ultrafine particle species that could move through soil voids with rainwater, which can be inferred from various situational evidence obtained by analyzing many samples [39–47].

The adsorption behavior of radionuclides on clay minerals in the soil has been studied for many years, and it is well known that radioactive cesium penetrates clay mineral layers where it undergoes strong ionic adsorption and binds to soil particles (References 19–30 in Chap. 1). Heavy rain associated with a category five super typhoon drenched the Tokyo metropolitan area from May 28–30, 2011, totaling 108 mm of rain in central Tokyo [48]. The radioactive cesium on the soil of the Imperial Palace Outer Garden might have been washed out in this heavy rain. Perhaps, the ultrafine radioactive cesium particles called hot particles and the radioactive cesium adsorbed on fine clay particles or plant litter and humus were washed away from the soil surface. As a result, there is no denying that the concentration and inventory of radioactive cesium may have decreased. However,

as shown by the vertical arrows in Fig. 3.6, torrential rains have occurred in the Tokyo metropolitan area many times since then. In other words, soil runoff due to rainfall alone cannot explain the rapid decrease in radioactivity immediately after the accident. This fact also implies that radioactive cesium deposited immediately after the FDNPP accident may have contained a large amount of mobile cesium in a highly migratory form. In other words, radioactive cesium that disappeared from the soil early in the accident is thought to have existed as a chemical species that was not easily retained in the soil. The concentration and inventory of radioactive cesium in the soil of the park at Site "i" seem to have fluctuated significantly since the second year of the accident, but compared to the expected decrease in radioactivity intensity due to the radioactive decay of $^{134+137}$Cs, its value appears to be steadily increasing over time. These spatiotemporal variations in radioactive cesium in soil, as shown in Fig. 3.6, indicate that the recycling system for the transport and supply of radioactive cesium at this location may not have been balanced. It is possible that the supply was much lower than the amount lost due to movement on the ground surface. The Tokyo metropolitan area is surrounded by many highly contaminated areas, not just that around the FDNPP reactors. Thus, the possibility that radioactive cesium continued to enter Tokyo with atmospheric dust cannot be denied.

Most likely, the temporal changes in radioactivity intensity in the Imperial Palace Outer Garden soil may reflect the effect of uneven distribution depending on the sampling area. To overcome this difficulty, Chap. 5 investigates the radioactive contamination of sludge incineration ash at a sewage treatment plant, which captured temporal fluctuations in radioactive contamination in Tokyo.

Many of the author's data discussed in this chapter have been published in PLOS ONE [49]. The data were revised and recalculated as needed before inclusion in this book.

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