別紙様式１（課程博士申請者用）

博 士 学 位 論 文

Dynamic survey of radioactive cesium concentration in cedar pollen after Fukushima Daiichi nuclear power plant accident in Japan

日本における福島第一原子力発電所事故後のスギ花粉中放射性セシウム濃度の動態調査

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- Abstract -

Due to the impact of the Tsunami caused by the Great East Japan Great Earthquake that reached the magnitude of 9.0 which occurred on March 11, 2011, ^{134}Cs and ^{137}Cs were diffused in the environment by Fukushima Daiichi power plant accident. Cesium is known to have properties similar to potassium. Cesium is selectively absorbed into plants by misidentifying cesium as potassium which is a cognate element. Radioactive cesium scattered by the accident is absorbed through plant roots by being redistributed after deposition in soil. I conducted a unique survey similar to that of the Ministry of Agriculture, Forestry and Fisheries in the Cryptomeria forest in Ome City, Tokyo, and analyzed the radioactivity concentration of cesium contained in cedar pollen. We carried out this survey from December 2011 to December 2017 for about seven years and examined the change in cesium radioactivity concentration in cedar pollen in Ome city.

The total value of ^{134}Cs and 137 Cs decreased from the average 140.3Bq kg^{-1} in December 2011. Radioactive cesium concentration in cedar pollen in Ome City decreased to 47% on average to 66.6 Bq kg^{-1} in December 2012 compared to December 2011. In January 2014, the average was 26.1 Bq kg^{-1} and it was 19%. In December 2014, the average value was 7.2 Bq kg^{-1}, which was 5%, and in October 2015 it was 1%, an average of 1.9 Bq kg^{-1}. After 2016, the measured value is less

than 0.0 Bq kg^{-1} and it seems to have reached the plateau. There is a difference between the dynamics of radioactive cesium derived from the Chernobyl nuclear accident that has been published so far and the dynamics in Japan. It is thought to be due to climate and terrain peculiar to Japan. In terms of accidents in Fukushima, it can be considered that direct soil was not contaminated as a result of vegetation of cedar forest, which worked in a good direction from the viewpoint of decontamination work.

Contents

Chapter 1 Introduction

1.1 Research background

Due to the impact of the Tsunami caused by the Great East Japan Great Earthquake that reached the magnitude of 9.0 which occurred on March 11, 2011, many of the facilities were destroyed at the Fukushima Daiichi Nuclear Power Station and the whole power supply was lost. Due to this influence, the cooling function of the reactor core was lost, causing a core melting accident at the nuclear power plant. A vent work carried out to depressurize the pressure vessel and the like due to the accident and a hydrogen explosion in the reactor building damaged the nuclear reactor and the building and a large amount of artificial radionuclide such as ^{131}Xe, ^{131}I, ^{134}Cs and ^{137}Cs were diffused in the environment[1]. In addition to the atmospheric releases, the direct discharges of highly contaminated water from the damaged reactor buildings resulted in contamination of radionuclides in the sea.

As shown in Table 1-1-1, it was difficult to accurately grasp the amount of radioactive cesium at the beginning of the accident, and there was a difference in the presentation of the total release volume by each research institution[2]. Since this was an announcement in the emergency phase, it was unavoidable that

accurate amounts were not grasped. Even if we compare the announcement of each institution again, it can be said that there was not a large gap between the numerical values.

Table 1-1-1. Evaluation results announced by Nuclear Safety Organizations Immediately after the accident[2)]

Organization	Publication Date	Evaluation period	Released Amount（PBq）				
			Noble gases	^{131}I	^{134}Cs	^{137}Cs	INES evaluation
Japan Atomic Energy Agency Nuclear Safety Commission	2011/4/12/ 2011/5/12/	2011/3/11/ to 2011/4/5/	-	150	-	13	670
Japan Atomic Energy Agency Nuclear Safety Commission	2011/8/22/	2011/3/12/ to 2011/4/5/	-	130	-	11	570
Japan Atomic Energy Agency	2012/3/6/	2011/3/11/ to 2011/4/10/	-	120	-	9	480
Nuclear and Industrial Safety Agency - Japan	2011/4/12/	-	-	130	-	6.1	370
Nuclear and Industrial Safety Agency - Japan	2011/6/6/	-	-	160	18	15	770
Nuclear and Industrial Safety Agency - Japan	2012/2/16/	-	-	150	-	8.2	480
Institut de radioprotection et de sûreté nucléaire (IRSN)	2011/3/22/	2011/3/12/ to 2011/3/22/	2,000	200	30		-

Among these, radioactive iodine (^{131}I) and radioactive cesium (^{134}Cs and ^{137}Cs) are the main nuclides that may migrate into plants. It is said that its release amount is about 10% at ^{131}I compared with the occurrence of a Chernobyl nuclear power plant accident, and about 38% and about 17% for ^{134}Cs and ^{137}Cs, respectively[3) ~ 5)].

Table 1-1-2. Comparison of the radionuclide quantity released into the environment due to the Chernobyl nuclear accident and the Fukushima Daiichi nuclear power plant accident[2)]

Property and nuclide name	Physical half-life	Total release volume due to Chernobyl nuclear accident PBq (10^{15} Bq)*1	Release amount by Fukushima Daiichi nuclear power plant accident (～3/15) PBq (10^{15} Bq)*2
Volatile element			
tellurium 129m	33.6 d	240	3.3
tellurium 132	3.20 d	~1,150	88
Iodine 131	8.02 d	~1,760	160
Iodine 133	20.8 h	910	42
cesium 134	2.065 y	~47	18
cesium 137	30.17 y	~85	15
Elements with intermediate volatility			
Strontium 89	50.53 d	~115	2.0
Strontium 90	28.79 y	~10	1.4
Ruthenium 103	39.26 d	>168	0.0000075
Ruthenium 106	373.6 d	>73	0.0000021
Barium 140	12.75 d	240	3.2

Cont.

Nonvolatile (including fuel particles)			
Zirconium 95	64.03 d	84	0.017
Molybdenum 99	2.75 d	>72	0.0000067
Cerium 141	32.51 d	84	0.018
Cerium 144	284.9 d	~50	0.011
Neptunium 239	2.356 d	400	0.076
Plutonium 238	87.7 y	0.015	0.000019
Plutonium 239	24.110 y	0.013	0.0000032
Plutonium 240	6.564 y	0.018	0.0000032
Property and nuclide name	Physical half-life	Total release volume due to Chernobyl nuclear accident PBq (10^{15} Bq)*1	Release amount by Fukushima Daiichi nuclear power plant accident (～3/15) PBq (10^{15} Bq)*2
Curium 242	162.8 d	~0.4	0.00001

*1 IAEA, Environmental Consequences of the Chernobyl accident and their remediation: twenty years of experience / report of the Chernobyl Forum Expert Group 'Environment', IAEA Vienna, 2006.
*2 Excerpted from the Ministry of Economy, Trade and Industry "Nuclear Safety and NISA" Partial error of radioactive material release data ".

It is already known that radionuclides brought about by nuclear tests in the atmosphere at the past and accidents at the Chernobyl nuclear power plant in the past exist in the environment and eventually in the soil, and the amount has been clarified by known reports[6].

Regarding the total emission amount of radionuclide immediately after the accident, refer to Table 1-1-2 Ministry of Economy, Trade and Industry

announcement figures[5]. Among the various kinds of radionuclides released with accidents, ^{131}I, which is a nuclide with a relatively short half-life, is supposed to be about 1/16 after one month if the release from Fukushima Daiichi NPS is stopped, and it was forecasted to decrease to about 1/2000 after 3 months[7]. However, for radioactive cesium (^{134}Cs and ^{137}Cs), there was no change in total radiation dose even 3 months after the accident.

As for cesium, it is vaporized by the low temperature 944 K as it is the metal, which is next to 630 K of mercury, so it easily vaporizes due to the heat provided by the accident and it has the property that it easily diffuses in the wind. In addition, it was concerned that it will stay for a long time in the environment and have a long influence on the surrounding environment because of its long physical half-life of (^{137}Cs in 2.0648 year and ^{137}Cs, 30.1671) year. Since April 2011, the emission of radionuclides into the atmosphere is limited and additional contamination to the terrestrial environment is also minimized[5], though the range contaminated with radioactive material due to accident is extensive.

While the decontamination work by the government steadily progresses, as shown in Figure 1-1, as of January 2018 the evacuation direction area was designated in the range of 20 km radius around the Fukushima Daiichi nuclear power plant and the range extending to the northwest direction about 40 km, many residents are in a situation where their homes are restricted[8]. Although the ^{137}Cs

concentration in the sediment within the radius of 20 km from Fukushima Daiichi nuclear power plant was several 1000 Bq kg^{-1}-dry in April 2011 immediately after the accident[9], in January 2015 it decreased to about 10 times the concentration before the accident (in the year 2000; ~2.0 Bq kg^{-1}-dry[10]). In this way, it was able to quickly respond by setting appropriate evacuation areas and evacuation areas immediately after the Fukushima Daiichi nuclear power plant accident because of many models and research results existed from the experience of the Chernobyl accident. It was able to take advantage of that experience in Fukushima Daiichi nuclear power plant accident.

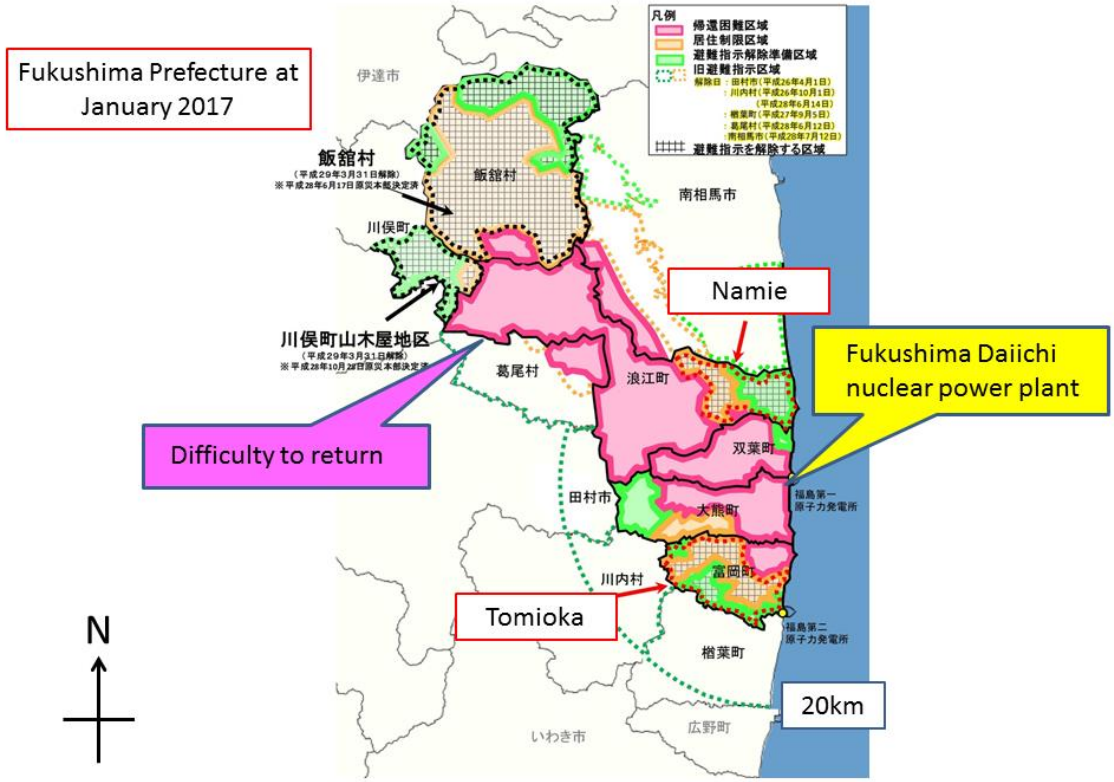

Figure 1-1. The latest conceptual diagram of evacuation instructed area in Fukushima[8].

* Pink frame - Difficulty to return, Yellow frame -Habitable restricted area, Green frame -Evacuation direction cancellation preparation area, Dotted line – Ex-evacuation Instruction Area

Remediation of terrestrial areas has been widely applied to areas contaminated by nuclear accidents, in particular after nuclear power plant accidents at Windscale, Chernobyl and Fukushima. Here we compare Chernobyl nuclear power plant accident with Fukushima Daiichi nuclear power plant accidents. A comparison of remediation after the Chernobyl and Fukushima Daiichi nuclear power plant accidents are shown in Table 1-2-1 to Table 1-2-5 below[11]. Radioactive iodine and radioactive cesium isotopes were important dose-forming radionuclides for both of

these accidents. Since radioactive iodine attenuated after sufficient time from the occurrence of the accident, radioactive cesium isotopes are the cause of environmental pollution[12]. Residents' exposures to radiation emitted from these radioactive cesium and migration to agricultural crops are still serious problems[13][14].

Table 1-2-1. Chernobyl and Fukushima comparison- contamination scenario[11]

	Chernobyl	**Fukushima Daiichi**
Factor	**Similarities**	
Deposition	Heterogenous at a small and large scale	
	Differences	
Context	NPP based accident	Followed a major earthquake and tsunami
^{134}Cs : ^{137}Cs ratio	1:1.6 – slower combined physical decay	1:1 – faster combined physical decay
Timing	At start of growing season	Before growing season
Population intensity	Moderate, no pressure to use land	High, pressure on available land
Terrain	Flat, forested and agricultural	Mountainous: forested slope and coastal catchment
Intensity of agriculture	Low – medium	High
Key products	Milk, meat, grain, potatoes	Rice, fruit, leafy and root crops, grain, flowers
Extent of lateral movement	Low	Potentially high

Table 1-2-2. Chernobyl and Fukushima comparison of exposure pathways[11)]

	Chernobyl	**Fukushima Daiichi**
Factor	**Similarities**	
	Soils having clay minerals with frayed edge sites lind radiocaesium and reduce mobility rapidly	
	Soils with low exchangeable K have relatively high radioceasium transfer to crops	
	Differences	
Fraction of soils with high OM content	High	Low
K fertilliser usage	Low	High
RCs availability for root uptake	Moderate to very high	Very low to moderate
Transfer to Animal products	High	Low
Intake of local food	High to very high	Low
Intake of wild food	High to very high	Low to moderate
Intensity of agriculture	Low – medium	High

Table 1-2-3. Chernobyl environmental remediation classification*[11)]

^{137}Cs [kBq m^{-2}]	**Classification**
Below 37	Not contaminated
37 – 185	Remediation for areas with "sensitive soils" (eg wet peat, acid sandy)
185 – 555	Remediation applied for sandy soils and light loam soils
555 – 1480	Full scale remediation
>1480	No economic activity

*After the division of the USSR(1991) different policies were adopted in Ukraine and Belarus

Table 1-2-4. Chernobyl and Fukushima comparison of radiological criteria[11)]

	Chernobyl	**Fukushima Daiichi**
Factor	**Similarities**	
	Long term goal of effective annual dose 1 mSv	
	Differences	
Temporary permissible levels for effective annual dose	1986 – 100 mSv 1987 – 30 mSv 1988-1989 – 25 mSv 1991 – 1 mSv	March 2011 – 5 mSv Sep 2011 – 1 mSv
Ambient dose rate μSv h^{-1}	2.2corresponding to lifetime additional dose of 350 mSv (applied in 1989)	0.19(excl. natural background)
Agricultural land	Varies with soil type	5000 Bq kg^{-1} dw
Changes with time in food standard limits	Down in CIS countries, stable in EU countries	Down(decreasing)

A wide range of aircraft surveys including the Kanto region were held for the first time after the accident, and Figure 1-2 shows the results of the Ministry of Education, Culture, Sports, Science and Technology announcement of the total deposited amount of radioactive cesium (^{134}Cs and ^{137}Cs) diffused by the Fukushima Daiichi Nuclear Power Plant accident[15)]. This is converted from the data obtained by the aircraft survey to the value as of September 18, 2011. Radioactive cesium diffuses widely from the Fukushima Daiichi Nuclear Power Station in north-northwest and south-west directions, and it seems that radioactive cesium is forming hot spots in a part of the Kanto region. This is due to the influence of the wind direction at the time of large-scale leak occurred on March 15, 2011 and the air

current by the topography of the inland area. The total deposited amount of ^{134}Cs and ^{137}Cs at the measurement point in Ome city in Tokyo which we investigated in this study is from 10 kBq m^{-2} to 30 kBq m^{-2}. Total deposits in Tokyo in the west of Ome City and around the Edogawa River basin were all less than 10kBq m^{-2}, indicating the highest total deposit amount of 60kBq m^{-2} to 100kBq m^{-2} in the Okutama region in western Tokyo from Ome City to Chichibu Mountains. A part of the radioactive plume transported to the central part of the Kanto region in the morning of March 15, 2011 was transported to the middle part of the Kanto region in the afternoon. Also, on the afternoon of March 20, About 40 km of radioactive plume has been observed from the eastern coast of Ibaraki prefecture to the western part of Saitama due to the wind closer to the east and it is estimated that it stayed at high concentration in the western part from the nighttime to the morning of 21st[3)]. High concentrations of radioactive plumes are considered to have been transported to the middle Kanto district through the atmosphere and deposited on the ground by precipitation. The characteristics of the precipitation distribution are consistent with the dose distribution on the surface.

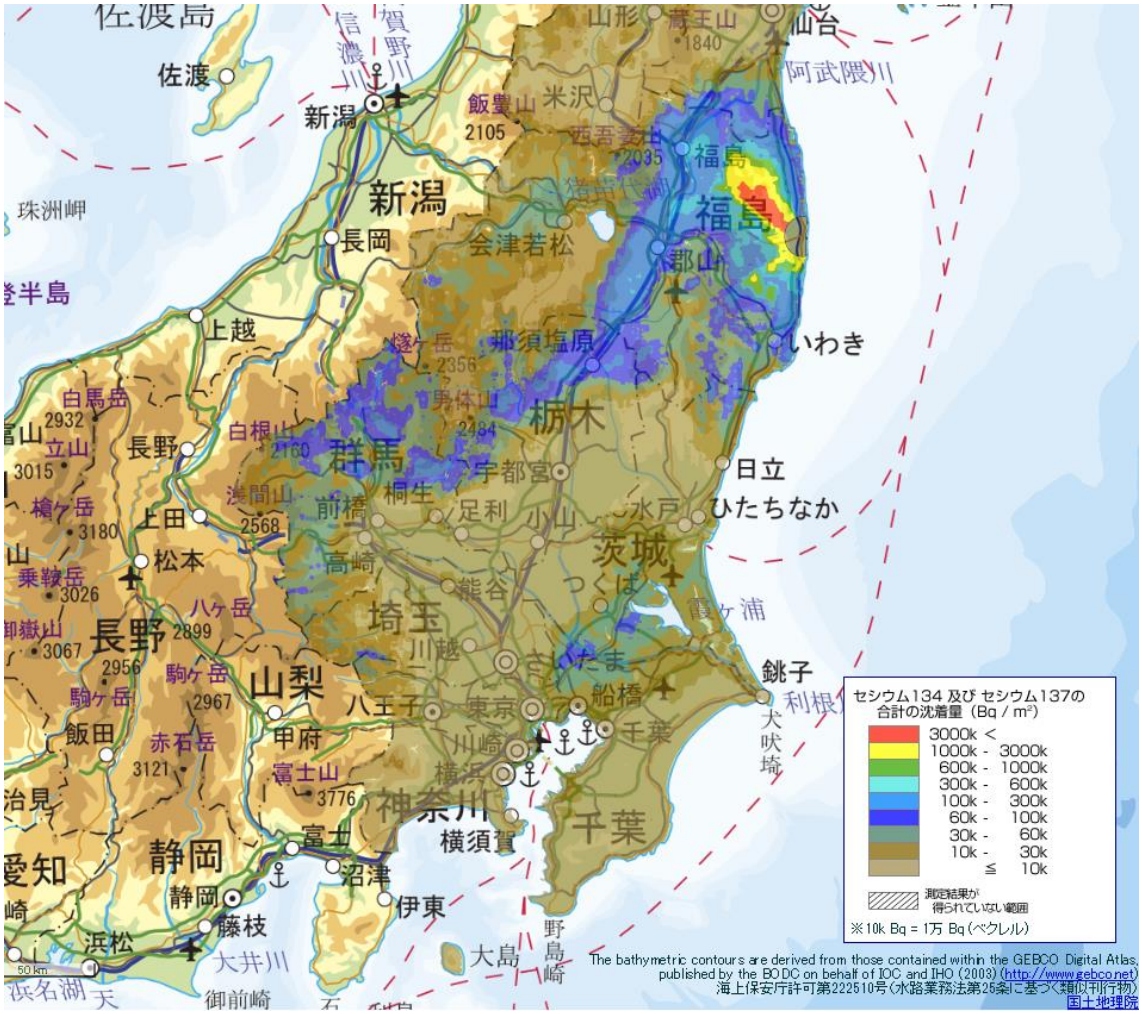

Figure 1-2. Airborne survey map by Ministry of Education, Culture, Sports, Science and Technology – Japan. total deposition amount of radioactive cesium (^{134}Cs + ^{137}Cs) on 18th of September 2011[15].

1.2 Transfer of cesium and its behavior in plants

1.2.1 Transfer of cesium into plants

Cesium is known to have properties similar to potassium, a cognate element, called alkali metal. Potassium is counted as one of the basic three elements of plant fertilizer[16], in addition to nitrogen and phosphate, and potassium present in the soil is selectively taken into plants. Cesium is also actively absorbed into plants as a result of misidentification of cesium as potassium, which is a cognate element. Radioactive cesium scattered by accident is absorbed through plant roots by being redistributed after deposition in soil[12]. At the same time, it is known that radioactive cesium on the wind is deposited as it is trapped by leaves of plants and falls off from plants by weathering, while it is absorbed into plants by absorption on the leaves[17]. Regarding absorption from the surface of plants, it is said that absorption rate from leaves is particularly high[1]. As the concentration of radioactive cesium in the atmosphere decreases, the proportion of leaf absorption decreases and the absorption from the root increases[18]. When radioactive cesium is taken up in Japanese cedar trees (Cryptomeria japonica), radioactive cesium absorbed via roots and leaves specifically moves to male flowers of cedar trees. As comparing the concentrations of radioactive cesium (^{134}Cs and ^{137}Cs) contained in male flowers and pollen inside them is about the same level[19], we decided to estimate the radioactive

cesium concentration in cedar pollen by measuring the radioactive cesium concentration of male flowers of cedar trees.

1.2.2 Respreading of radioactive cesium by cedar pollen

Since cedar male flower is a flower that carries pollen by the wind, radioactive cesium descended to the forest was concerned that it would be rediffused as cedar pollen scatters[20]. The size of cedar pollen is about 30 μm in diameter, and it has small protrusions called papilla which exhibit a shape close to a sphere. Because its weight is as light as 12 ng[21], depending on the weather conditions though, it catches the wind and scatters more than 200 km[22], As a result, residents of the metropolitan area were concerned about internal radiation exposure with radioactive cesium by inhaling cedar pollen which flew from Fukushima prefecture and cedar forest near Tokyo. Figure 1-3-1 shows distribution of cedar forest mainly in Kanto district[23]. It is understood that the density of cedar forest is high in the southern part of Fukushima Prefecture and the northern part of Ibaraki Prefecture, and in the western part of Saitama and Tokyo. Figure 1-3-2 shows the distribution of major source areas of pollen bombarded by people living in the metropolitan area based on the results of the pollen scattering peak of 2008[23]. Red and yellow areas indicate sources that have a strong influence on the metropolitan area (the portion

surrounded by the blue line).

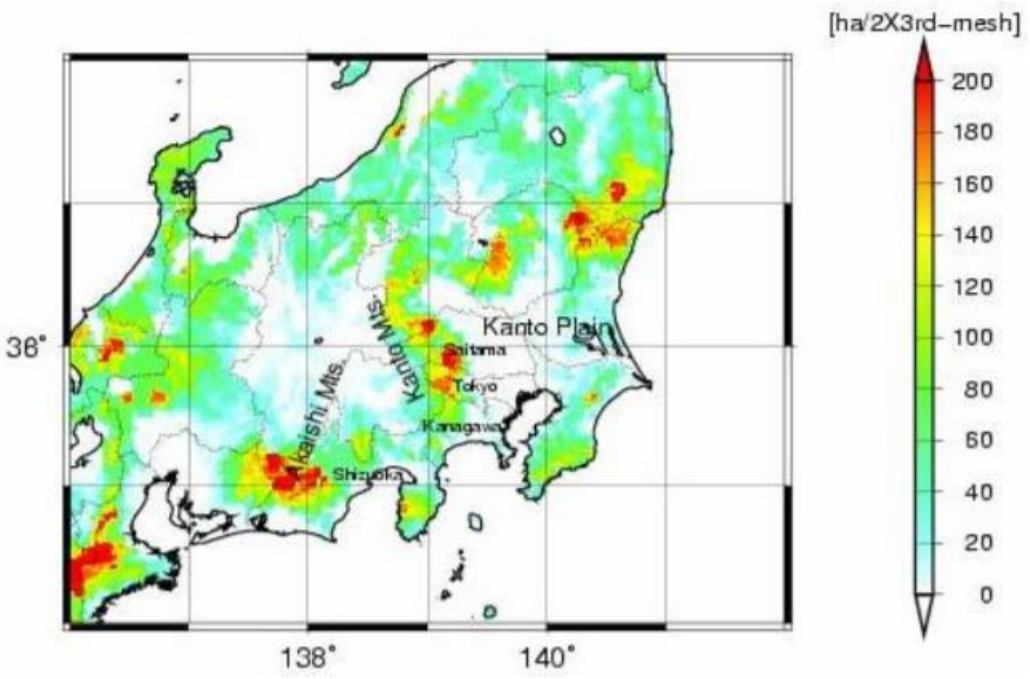

Figure 1-3-1. Distribution of sources of Japanese cedar pollen (Cryptomeria japonica forests over 26 years old) in Honshu central area[23)]
* Red and yellow indicate areas with many cedar forests

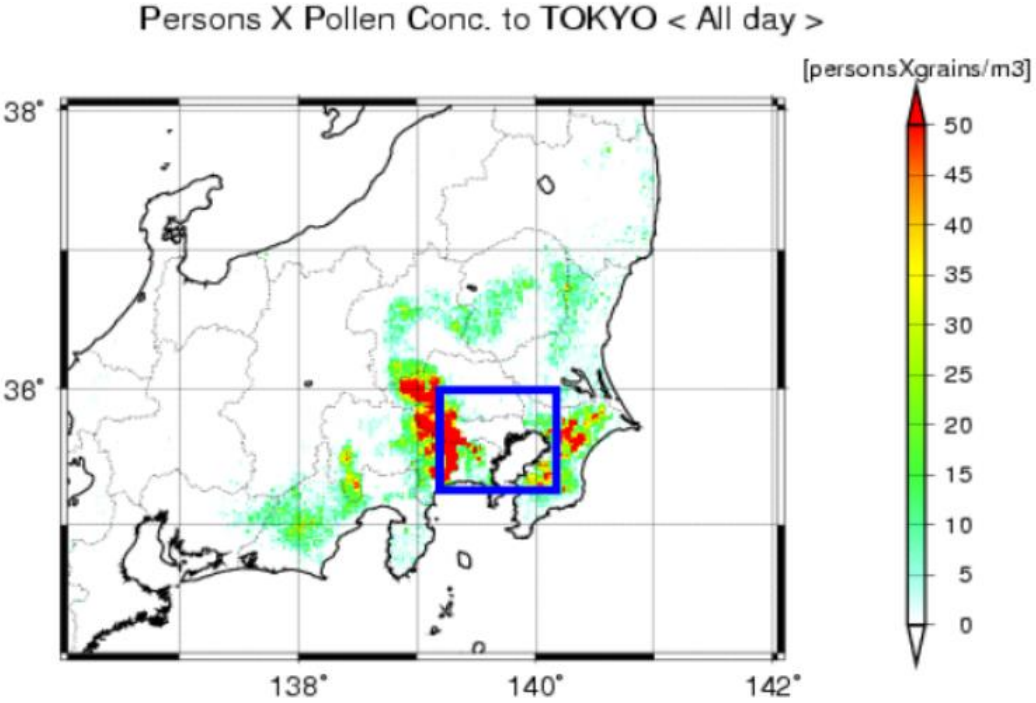

Figure 1-3-2. Distribution of major sources of pollen exposed to people living in the metropolitan area region distribution[23)]

1.3 The purpose of this research

In order to estimate the amount of internal exposure by pollen, the Ministry of Agriculture, Forestry and Fisheries continued to investigate the concentration of radioactive cesium ^{134}Cs and ^{137}Cs contained in male cedar flower at 24 measuring points in Fukushima Prefecture from 2011 to 2014 after the accident. 253,000 Bq kg^{-1} of radioactive cesium was observed in Namie Town, Futaba-gun, Fukushima Prefecture, which showed the highest value in 2011[24]. As a survey conducted in Tokyo, survey results conducted in Nishitama-gun, Tokyo and Hachioji city in Tokyo Metropolitan Government were press released on February 8, 2012 press release. A partial excerpt of the result is shown in Table 1-3[25]. The survey conducted by the Ministry of Agriculture, Forestry and Fisheries based on conditions such as the vegetation of Cryptomeria forest is limited to the west area from Hachioji, Tokyo. In this table, the value of radioactive cesium concentration per dry amount [kg] is not detected, and the measurement result of cedar in Tanasawa Okutama Town, Nishi-Tama-gun, Tokyo Metropolitan area is a value of 100 Bq kg^{-1} or less. In the other two points, they measure 398 Bq kg^{-1} in Kobotoke Uratakao town in Hachioji city, and 223 Bq kg^{-1} in Hinohara village Kurakake, Nishitama - gun, Tokyo. However, there have been no research studies that continued the measurement of radioactive cesium concentration in cedar pollen in Tokyo afterwards.

Table 1-3. Survey results and locations of radioactive cesium contained in cedar male in Tokyo metropolitan area[25].

Measurement point number	Location of cedar forests	Concentration of radiocesium contained in the male cedar flower [Bq kg^{-1} dry weight]
175	Kobotoke Uratakao cho Hachioji, Tokyo	398
176	Tanazawa Okutama Nishitama, Tokyo	ND
177	Kurakake Hinohara Nishitama, Tokyo	223

Note: ND represents value was below 100 Bq kg^{-1}

Therefore, we also conducted a unique survey similar to the Ministry of Agriculture, Forestry and Fisheries in the Cryptomeria forest in Ome City, Tokyo, and analyzed the radioactivity concentration of cesium contained in cedar pollen. We carried out this survey from December 2011 to December 2017 for about seven years and examined the change in cesium radioactivity concentration in cedar pollen in Ome city. In addition, based on the obtained results, the ecological half-life, which is an index when radioactive cesium decays from a specific environment, was estimated by a calculation formula. The ecological half-life is a half-life taking into consideration climatic conditions and topographical environmental conditions and its deposition process in addition to the physical half-life unique to the radionuclide[26]. The ecological half-life is an important factor in estimating the exposure dose of the surrounding residents in the area, since it is a measure of attenuation of the radioactive half-life per land. 66% of the area of Japan is forest

and 71% in Fukushima prefecture is also forest[28]. Even in Tokyo, where the capital city of Japan, its forest ratio reaches 36%. Investigating the dynamics of radioactive cesium in the forest is an important research for Japan. There have been many Western studies that investigated the impact of the Chernobyl nuclear power plant accident in the past, but research that investigated the radioactive cesium dynamics in the forest in Japan is a new attempt to start after the Fukushima Daiichi nuclear power plant accident occurred. I designed the present study since there have not been any studies investigating attenuation of radioactive cesium concentration in cedar pollen in Japan, We would like to consider this study.

Chapter 2　Survey method

2.1　Measurement location and timing of male cedar flower sampling

2.1.1　Selection of male cedar flower sampling area

The point where male cedar flower is collected is based on the measurement result of aircraft monitoring in Tokyo suburbs conducted by the Ministry of Education, Culture, Sports, Science and Technology, September 2011. We selected Tokyo Metropolitan Okutama Area as the subject of investigation based on the condition that the total deposit amount [Bq m^{-2}] of ^{134}Cs and ^{137}Cs in Tokyo is high and there is a wide range of planted plants of cedar trees[15]. Figure 2-1-1 shows the positional relationship between Ome city that I set measurement points and Fukushima Daiichi nuclear power plant in Japan. The measurement point of Ome city and the straight distance to Fukushima Daiichi nuclear power plant are 242 km. Ome city is a city in the northwest part of the Tama area of Tokyo, and it is the fourth largest in the municipalities of Tokyo, excluding the islands. A village developed at the fan tip where the Tama river flows from the Kanto Mountains to Musashinoidai area became the base of Ome city center. Tamagawa River flows from west to east near the center of the city area, and Kasumigawa and Mogi River swhich are tributaries of the Iruma River (Arakawa water system) flow from the

west to the east in the north. It is largely changed from a flat land in the eastern part to the western part in hilly and mountainous regions. Annual average temperature is about 13~14 degree in Celsius. The climate is mixed with the Pacific Coast climate and the Central Highlands climate, it is colder than in the city center in winter and the lowest temperatures in January and February are recorded below freezing almost every day.

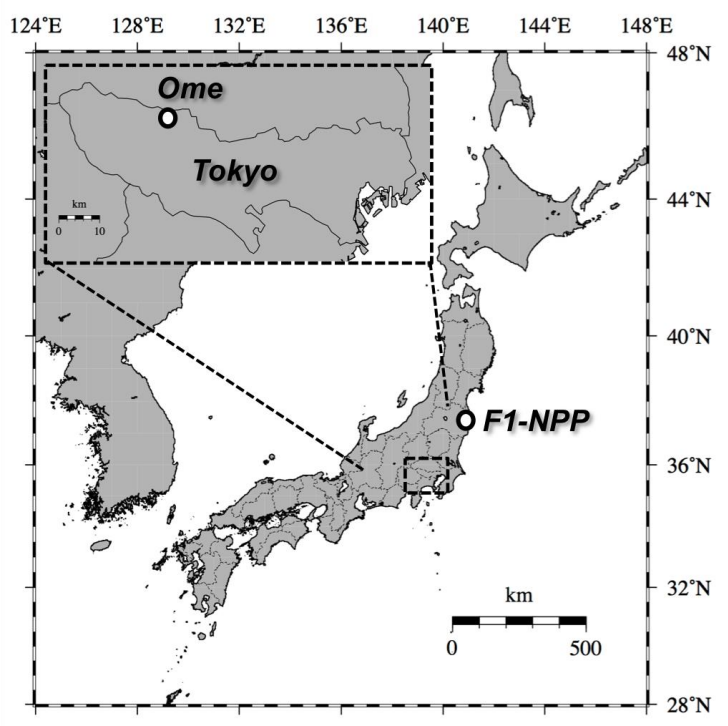

Figure 2-1-1. Measurement point Yuzuki-cho in Ome city Tokyo.

Figure 2-1-2 shows detailed geographical features of radioactive cesium and the detailed geography of Yuzuki-cho Ome City, the measurement points are exactly located[15]. Four measurement points were set in the yellow circled area. Close to the north the Tamagawa River flows

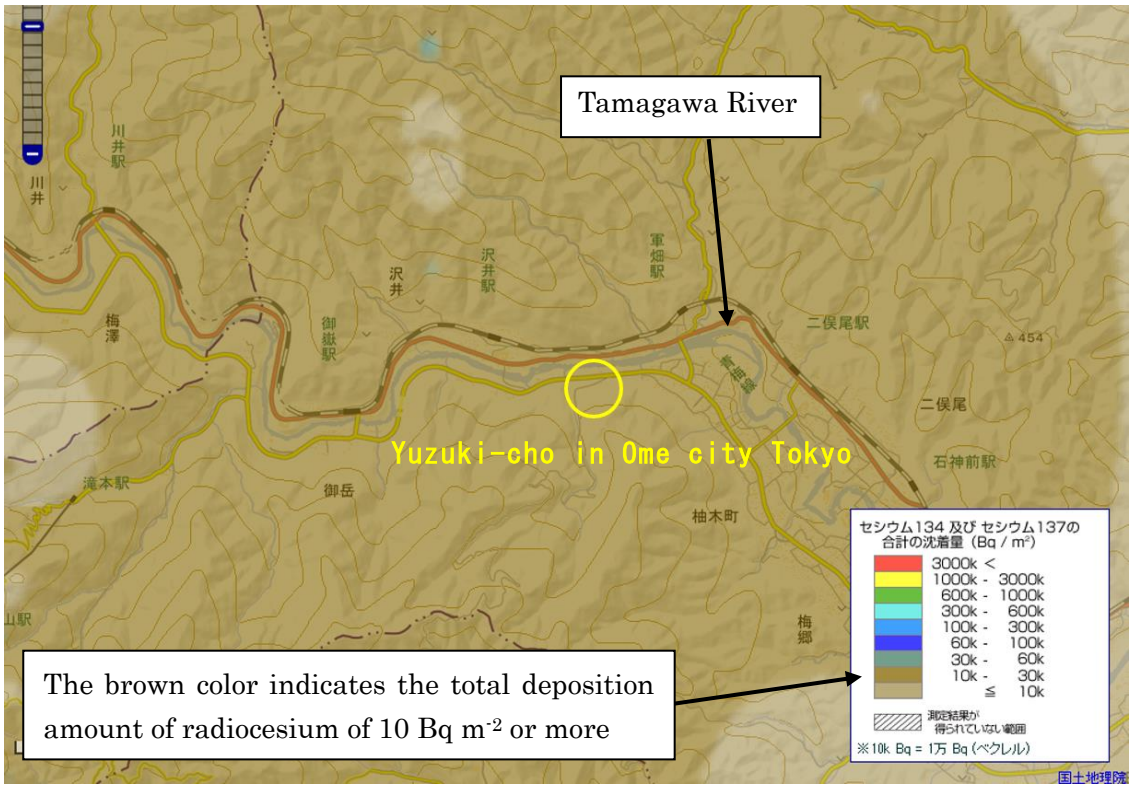

Figure 2-1-2. Measurement point location and total deposition amount of radioactive cesium at Yuzuki-cho in Ome city Tokyo on 18th of September 2011[15].

2.1.2 Male cedar flower measurement point

Several cedar trees wearing well mature male flowers were selected from cedar forest near Yunoki cho 3 in Ome city, Tokyo. Among them, the male flower was further planted at a position where it can be harvested, and four points from the tree showing standard size and branching were set as measurement points. A detailed latitude and longitude of a cedar tree selected as a survey tree in Ome city is shown in Table 2-1. I set points 1, 2, 3 and 4 in order from the trees located on the south side of the collected cedar trees. Figure 2-1-3 shows the vegetation of the

cedar at the measurement point. Cedar is vegetated on slopes, and there are no people who normally enter these cedar forests. Cedar has not been care for special pruning etc.

Table 2-1. Measurement points' latitude and longitude

Measurement point	Latitude	Longitude
1	35°48'10.60"	139°12'04.93"
2	35°48'10.72"	139°12'02.94"
3	35°48'12.21"	139°12'04.41"
4	35°48'12.15"	139°12'06.05"

Figure 2-1-3. Cedar vegetation of Ome city measurement point (2015)

2.1.3 Timing to sample cedar male flowers

Cedar is a flower medium, male flowers that blow cedar pollen occur during the summer from July to August, making pollen inside while matured in the autumn. Male flowers are completed by around November, after which the dormancy state is suspended, with decreasing temperature and shortening day time. Male flowers awake from dormancy by being exposed to low temperature for a certain period of time and enter the preparation period of flowering. Then in the spring of March to spring, it will bloom and pollen will be scattered[29]. From this, it is considered that the male flower is mature enough to be collected from November to February that is in the state before starting to scatter the pollen. Actual sampling of male flowers of cedar began in December 2011 when cedar male flowers matured, which were first made after the accident, matured, and then collected around December or January every year thereafter.

2.2 Measurement of radioactive cesium concentration in cedar male flower

At the measurement point, only the branches and leaves with dense mature cedar male flowers were cut off and collected. Figure 2-2-1 shows the appearance of the male flower collected at the measurement point.

Figure 2-2-1. Appearance of the male cedar flower collected in Ome city (2015)

The collected male flowers were dried using a general microwave oven considering the low vaporization temperature of cesium. They were dried for more than 15 minutes, paying attention not to make them reach extremely high temperature, and processed into powder in a mortar. Weight of powdered male flower was measured, and it was

packed in U-shaped screw vessel U - 8 (Sekiya Rikagaku Co., Ltd., polystyrene, height: 68 mmH, inner diameter: 56 mmφ) to prepare a measurement sample. Gamma ray measurement was performed for 30,000 seconds using a germanium semiconductor detector (ORTEC GMX 10 P, AMETEK, Inc., United States of America). Radioactivity concentrations were calculated from gamma ray peaks of ^{134}Cs at 605 keV and 796 keV, and ^{137}Cs at 662 keV. As an example of the sample measurement result of 2014 which measured the count above the detection limit as shown in Figure 2-2-2. As peak search method is smoothed second order differentiation peak search, peak center calculation method is second order differential coefficient 3 point parabolic approximation method, Full width at half maximum (FWHM) × 1.00 were set toindentify nuclide.

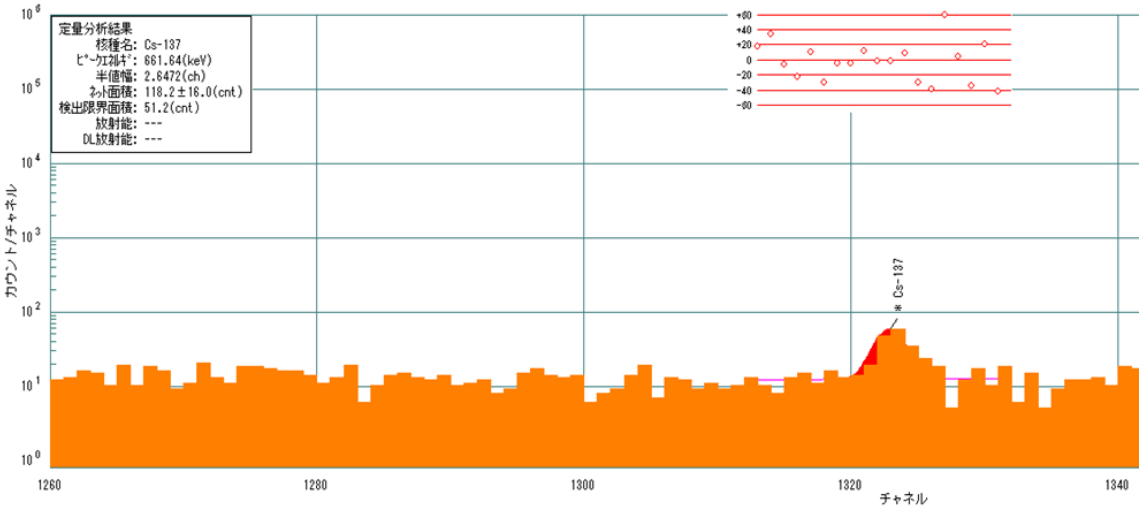

Figure 2-2-2. Peak count measurement result of cedar male flower sample (Measurement point 2 in 2014)

2.3 Ambient dose equivalent ratio measurement around cedar male flower sampling point

Ambient dose equivalent rate was measured at the height of 1 m above ground surface and recorded. For the measurement, a NaI (Tl) detector (radiation identi FINDER Ultra K-NG, ICX technologies, Oak Ridge, USA) was used. This device is capable of nuclide identification of gamma rays and spectrum measurement of gamma rays. Measurements were continuously made at 10-second intervals for 2 minutes, and the average value of the ten data excluding the maximum value and the minimum value was obtained from the calculated 12 pieces of data to derive the average air dose rate. Data measured by the Fukushima Technical Headquarters of the Japan Atomic Energy Agency entrusted by the Ministry of Education[30], Culture, Sports, Science and Technology, and the results of the travel survey published in the Extensin Site of Distrubution Map of Radiation Dose, etc. are shown in Figure 2-3-1to 2-3-3[15]. The area surrounded by a red circle is the area where male flowers were collected in this study. It can be seen that the space dose rate at the height of 1 m above the ground surface was less than 0.1μ Sv h^{-1} before and after the day when we collected cedar male flower, and it was consistent with the value we measured in the field.

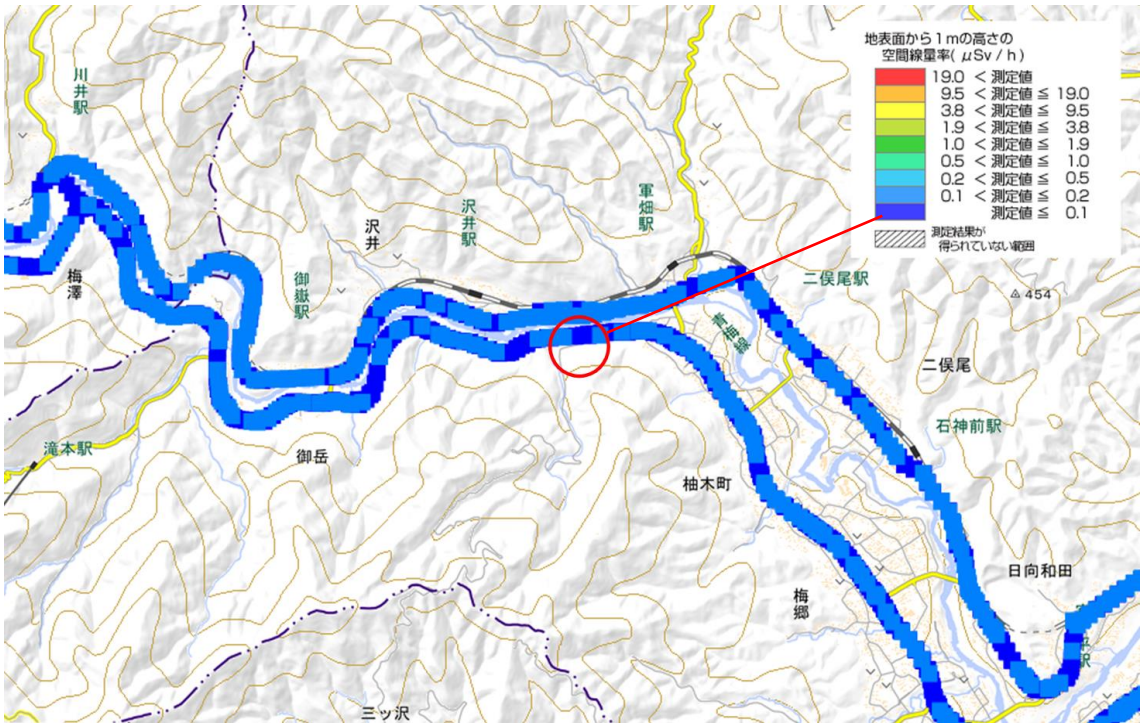

Figure 2-3-1. Air dose rate at the point we collected male cedar flower in Ome city by Education, Culture, Sports, Science and Technology - Japan (December 2011) [15)]

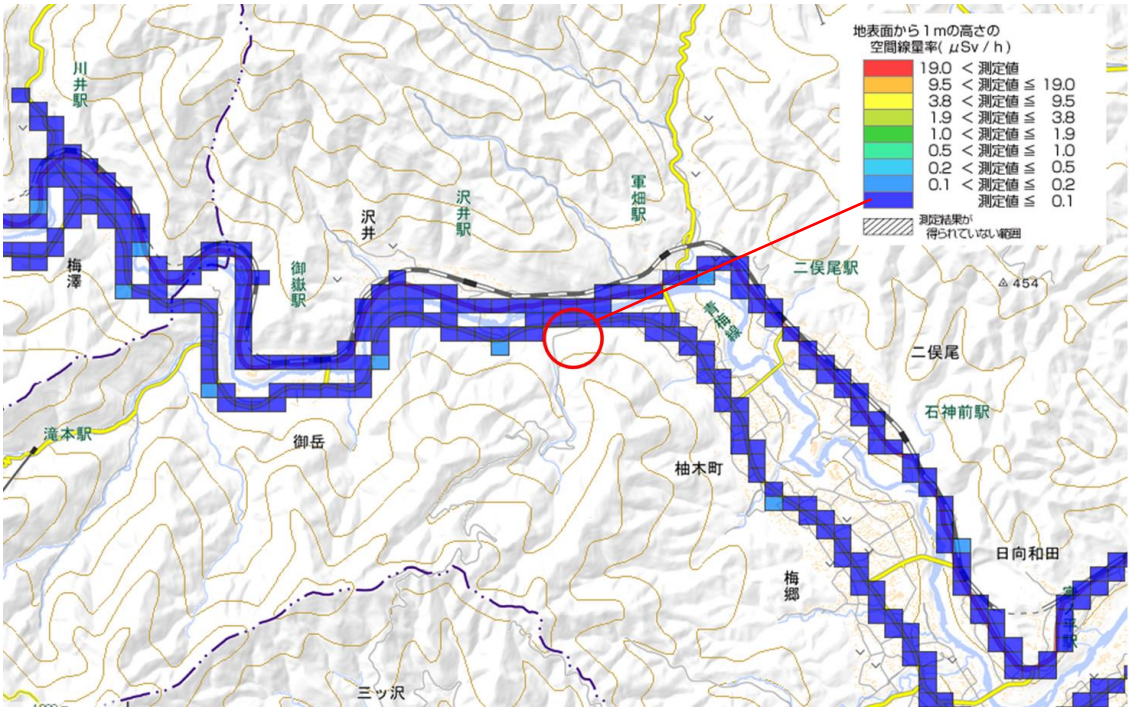

Figure 2-3-2. Air dose rate at the point we collected male cedar flower in Ome city by Education, Culture, Sports, Science and Technology – Japan (November to December 2012) [15)]

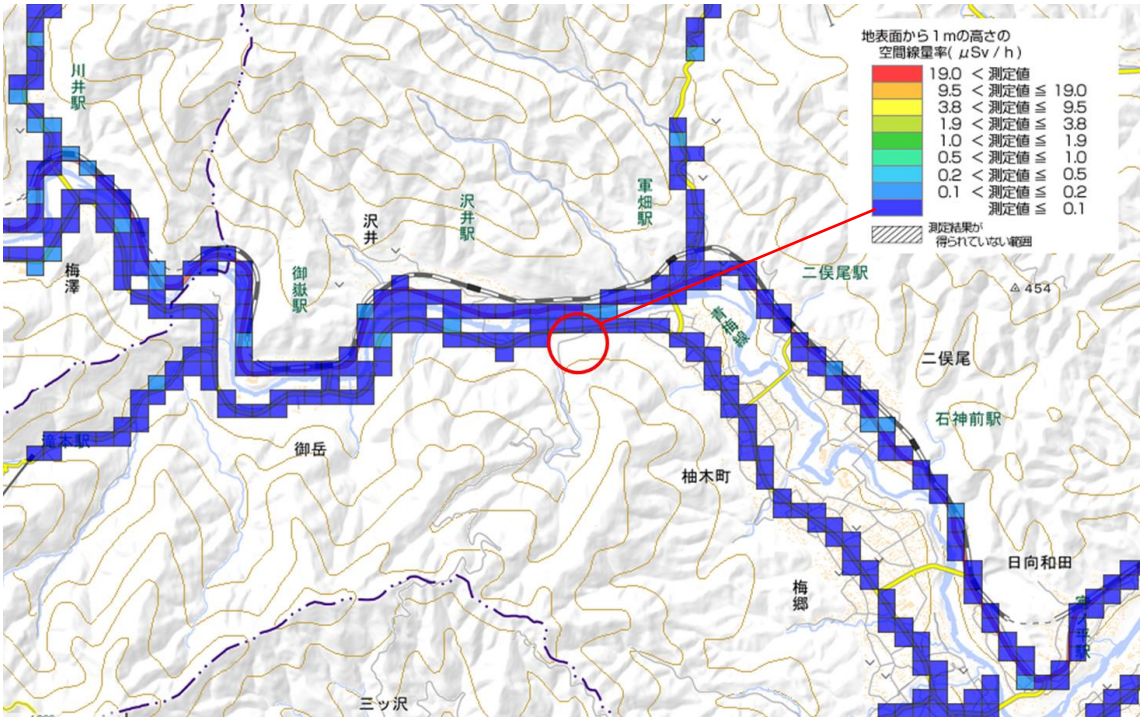

Figure 2-3-3. Air dose rate at the point we collected male cedar flower in Ome city by Education, Culture, Sports, Science and Technology – Japan (November to December 2013) [15)]

Chapter 3 Survey results

3.1 Measurement results of cedar male flower samples

3.1.1 Radioactive cesium concentration in cedar male flowers

The results obtained by measuring the male flower sample of cedar male flower radioactivity concentration were summarized for each radioactive cesium nuclide and shown in Table 3-1-1 to Table 3-1-7 in order of sample collection. 201.4 Bq kg^{-1} recorded in point 1 in 2011, which showed the highest value in the 7-year measurement. The radioactive cesium concentration in pollen obtained at each measurement point decreased every year, and as of January 2014 the radioactivity concentration of the sample at point 4 was below the detection limit of the device. However, the measurements made in December, radioactive cesium was detected from the sample at point 4 again. Though the trees to be sampled are the same, it is considered that the cause is that the measurement results are slightly different depending on the growth situation of each year and the difference of the branch that sampling is performed. In the measurements after December 2016, the radioactive cesium concentration was 0.0 Bq kg^{-1} or less in all samples. Although it is not able to be said that the numerical value was completely less than the detection limit, it can be said that it was attenuated to a value close to 0.0 Bq kg^{-1} as compared with 201.4

Bq kg^{-1} recorded at the beginning of the measurement. Even if the concentration of radioactive cesium in cedar pollen became 0.0 Bq kg^{-1} or less, the investigation was continued, because Ministry of Agriculture, Forestry and Fisheries continued to investigate the concentration of radioactive cesium ^{134}Cs and ^{137}Cs contained in male cedar flower in Fukushima. There was a purpose to make public the survey results in Tokyo, as a comparison target of Fukushima. Furthermore, there was a purpose to investigate whether the numerical value which once became less than 0.0 Bq kg^{-1} rises again due to environmental changes and cesium behavior in cedar trees. As a result, the survey result of 2017 were less than 0.0 Bq kg^{-1} at all points like 2016.

Table 3-1-1 Radioactive cesium concentration measurement details (2011.12.23)

Measurement point and nuclides		Measured value [Bq kg^{-1}]	Error	Detection limit [Bq kg^{-1}]	Radioactive Cesium (^{134}Cs + ^{137}Cs) average [Bq kg^{-1}]	Error average
1	^{134}Cs:605keV	83.9	3.1	5.2	201.4	6.3
	^{134}Cs.:796keV	89.0	3.9	6.7		
	^{137}Cs	114.9	3.8	5.5		
2	^{134}Cs.:605keV	64.7	2.8	5.0	137.1	5.4
	^{134}Cs.:796keV	61.5	3.4	6.2		
	^{137}Cs	74.0	3.2	5.5		
3	^{134}Cs.:605keV	71.4	3.0	5.4	173.6	5.8
	^{134}Cs.:796keV	79.1	3.6	6.0		
	^{137}Cs	98.3	3.5	4.9		
4	^{134}Cs.:605keV	23.7	1.9	4.3	49.1	3.9
	^{134}Cs.:796keV	24.4	2.5	6.1		
	^{137}Cs	25.0	2.3	5.4		

Table 3-1-2 Radioactive cesium concentration measurement details (2012.12.15)

Measurement point and nuclides		Measured value [Bq kg^{-1}]	Error	Detection limit [Bq kg^{-1}]	Radioactive Cesium (^{134}Cs + ^{137}Cs) average [Bq kg^{-1}]	Error average
1	^{134}Cs.:605keV	24.1	2.2	5.1	66.3	4.5
	^{134}Cs.:796keV	22.6	2.8	7.3		
	^{137}Cs	42.9	2.8	5.9		
2	^{134}Cs.:605keV	16.8	2.2	5.9	49.1	4.3
	^{134}Cs.:796keV	18.4	2.7	7.0		
	^{137}Cs	31.5	2.6	5.8		
3	^{134}Cs.:605keV	25.7	2.8	7.1	76.7	5.6
	^{134}Cs.:796keV	21.7	3.2	8.4		
	^{137}Cs	53.0	3.6	7.5		
4	^{134}Cs.:605keV	29.3	2.3	5.4	74.4	4.6
	^{134}Cs.:796keV	29.3	2.3	5.4		
	^{137}Cs	45.1	2.8	5.8		

Table 3-1-3 Radioactive cesium concentration measurement details (2014.1.15)

Measurement point and nuclides		Measured value [Bq kg^{-1}]	Error	Detection limit [Bq kg^{-1}]	Radioactive Cesium (^{134}Cs + ^{137}Cs) average [Bq kg^{-1}]	Error average
1	^{134}Cs.:605keV	13.8	2.8	8.1	54.3	5.9
	^{134}Cs.:796keV	13.5	3.5	3.5		
	^{137}Cs	40.6	3.8	3.8		
2	^{134}Cs.:605keV	2.0	0.6	1.7	6.0	1.0
	^{134}Cs.:796keV	1.0	0.5	1.3		
	^{137}Cs	4.2	0.6	1.7		
3	^{134}Cs.:605keV	8.6	2.2	6.6	44.2	5.0
	^{134}Cs.:796keV	13.0	3.1	9.0		
	^{137}Cs	33.4	3.2	7.6		
4	^{134}Cs.:605keV	0.0	NA	7.6	0.0	NA
	^{134}Cs.:796keV	0.0	NA	8.6		
	^{137}Cs	0.0	NA	8.6		

Table 3-1-4 Radioactive cesium concentration measurement details (2014.12.21)

Measurement point and nuclides		Measured value [Bq kg^{-1}]	Error	Detection limit [Bq kg^{-1}]	Radioactive Cesium (^{134}Cs + ^{137}Cs) average [Bq kg^{-1}]	Error average
1	^{134}Cs.:605keV	0.0	NA	4.2	2.9	NA
	^{134}Cs.:796keV	5.8	1.8	5.2		
	^{137}Cs	0.0	NA	4.6		
2	^{134}Cs.:605keV	7.0	1.5	4.2	6.6	NA
	^{134}Cs.:796keV	6.2	1.7	4.8		
	^{137}Cs	0.0	NA	4.5		
3	^{134}Cs.:605keV	6.3	1.5	4.2	7.1	NA
	^{134}Cs.:796keV	7.9	1.7	4.8		
	^{137}Cs	0.0	NA	4.5		
4	^{134}Cs.:605keV	6.3	1.4	4.2	12.3	NA
	^{134}Cs.:796keV	0.0	NA	8.6		
	^{137}Cs	9.1	1.5	3.9		

Table 3-1-5 Radioactive cesium concentration measurement details (2015.10.31)

Measurement point and nuclides		Measured value [Bq kg^{-1}]	Error	Detection limit [Bq kg^{-1}]	Radioactive Cesium (^{134}Cs + ^{137}Cs) average [Bq kg^{-1}]	Error average
1	^{134}Cs.:605keV	0.0	NA	6.5	0.0	NA
	^{134}Cs.:796keV	0.0	NA	8.3		
	^{137}Cs	0.0	NA	6.3		
2	^{134}Cs.:605keV	0.0	NA	5.9	7.7	NA
	^{134}Cs.:796keV	0.0	NA	7.2		
	^{137}Cs	7.7	2.3	6.8		
3	^{134}Cs.:605keV	0.0	NA	8.2	0.0	NA
	^{134}Cs.:796keV	0.0	NA	1.0		
	^{137}Cs	0.0	NA	8.3		
4	^{134}Cs.:605keV	0.0	NA	4.6	0.0	NA
	^{134}Cs.:796keV	0.0	NA	5.0		
	^{137}Cs	0.0	NA	4.6		

Table 3-1-6 Radioactive cesium concentration measurement details (2016.12.5)

Measurement point and nuclides		Measured value [Bq kg^{-1}]	Error	Detection limit [Bq kg^{-1}]	Radioactive Cesium (^{134}Cs + ^{137}Cs) average [Bq kg^{-1}]	Error average
1	^{134}Cs.:605keV	0.0	NA	0.0	0.0	NA
	^{134}Cs.:796keV	0.0	NA	0.0		
	^{137}Cs	0.0	NA	0.0		
2	^{134}Cs.:605keV	0.0	NA	0.0	0.0	NA
	^{134}Cs.:796keV	0.0	NA	0.0		
	^{137}Cs	0.0	NA	0.0		
3	^{134}Cs.:605keV	0.0	NA	0.0	0.0	NA
	^{134}Cs.:796keV	0.0	NA	0.0		
	^{137}Cs	0.0	NA	0.0		
4	^{134}Cs.:605keV	0.0	NA	0.0	0.0	NA
	^{134}Cs.:796keV	0.0	NA	0.0		
	^{137}Cs	0.0	NA	0.0		

Table 3-1-7 Radioactive cesium concentration measurement details (2017.12.14)

Measurement point and nuclides		Measured value [Bq kg^{-1}]	Error	Detection limit [Bq kg^{-1}]	Radioactive Cesium (^{134}Cs + ^{137}Cs) average [Bq kg^{-1}]	Error average
1	^{134}Cs.:605keV	0.0	NA	0.0	0.0	NA
	^{134}Cs.:796keV	0.0	NA	0.0		
	^{137}Cs	0.0	NA	0.0		
2	^{134}Cs.:605keV	0.0	NA	0.0	0.0	NA
	^{134}Cs.:796keV	0.0	NA	0.0		
	^{137}Cs	0.0	NA	0.0		
3	^{134}Cs.:605keV	0.0	NA	0.0	0.0	NA
	^{134}Cs.:796keV	0.0	NA	0.0		
	^{137}Cs	0.0	NA	0.0		
4	^{134}Cs.:605keV	0.0	NA	0.0	0.0	NA
	^{134}Cs.:796keV	0.0	NA	0.0		
	^{137}Cs	0.0	NA	0.0		

3.1.2 Transition of radioactive cesium concentration in cedar male flowers

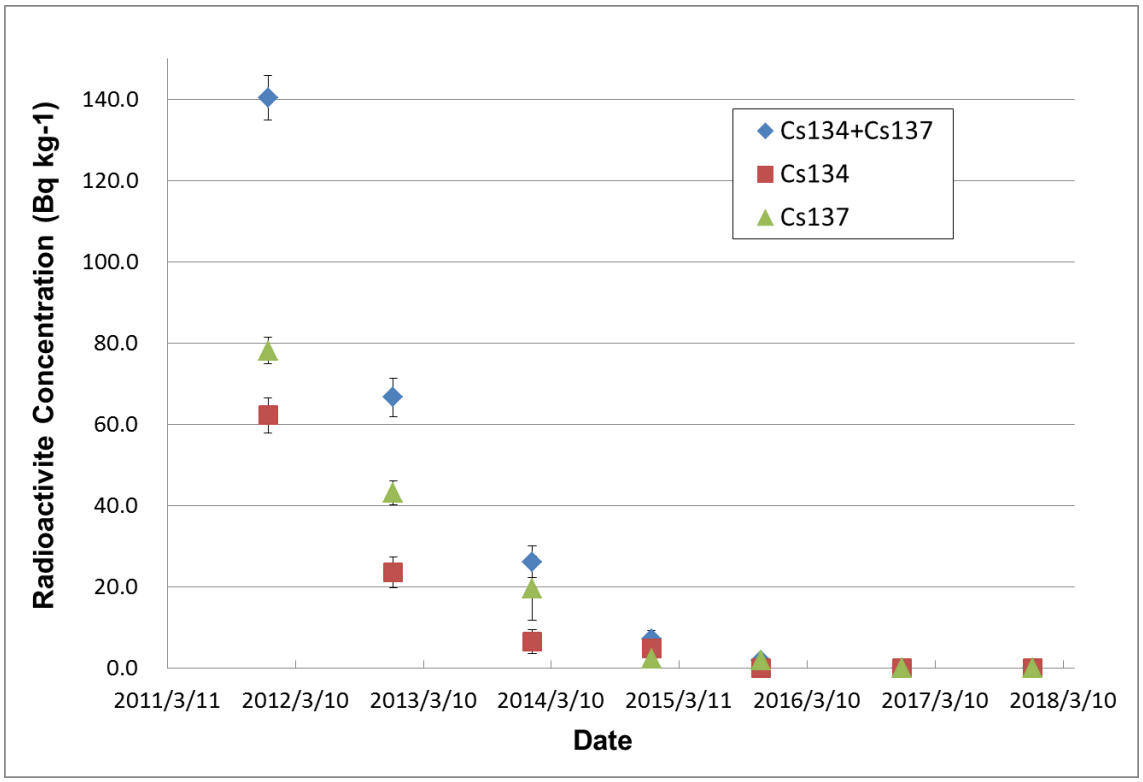

Figure 3-1-1. Variation of average radioactivity of cesium isotopes

Figure 3-1-1 shows a graph of changes in the measured radioactivity concentration of radioactive cesium in cedar pollen. The four measurement data obtained are averaged for each radioactive cesium nuclide, and the error on the measurement instrument is shown as an error bar. The total value of ^{134}Cs and 137 Cs decreased from the average 140.3 Bq kg^{-1} in December 2011 to the average 66.6Bq kg^{-1} in December 2012, to 47%, and in January 2014 the average decreased to 26.1 Bq kg^{-1} that is 22% of December 2011.

Next, a graph standardized with the value of December 2011 obtained from cedar

male flower matured formed and matched for the first time after the Fukushima Daiichi nuclear accident as 1 is shown in Figure 3-1-2. The total radioactivity concentration of ^{134}Cs and ^{137}Cs is halved after 1 year compared to the first year after the accident and it is reduced by 1/5 after 2 years. Then it decreased to 1/20 when 3 years passed and decreased to 1/75 when 4 years passed. The numerical value became almost 0 when more than 5 years passed. Radioactive cesium concentration in cedar pollen has halved faster than 2.0648 years, which is the physical half-life of ^{134}Cs and 30.1671 years, which is the physical half-life of ^{137}Cs. From this, it can be said that the ecological half-life of radioactive cesium in Ome city environment is shorter than the physical half-life. Looking at the transition for each nuclide, attenuation of ^{134}Cs is faster than ^{137}Cs. This is considered to be because of the fact that the physical half-life of ^{134}Cs is as short as 2.0648 years compared to the physical half-life of ^{137}Cs of 30.1671 years. From both Figure 3-1-1 and Figure 3-1-2 it can be seen that the radioactive cesium concentration in cedar pollen decreases exponentially

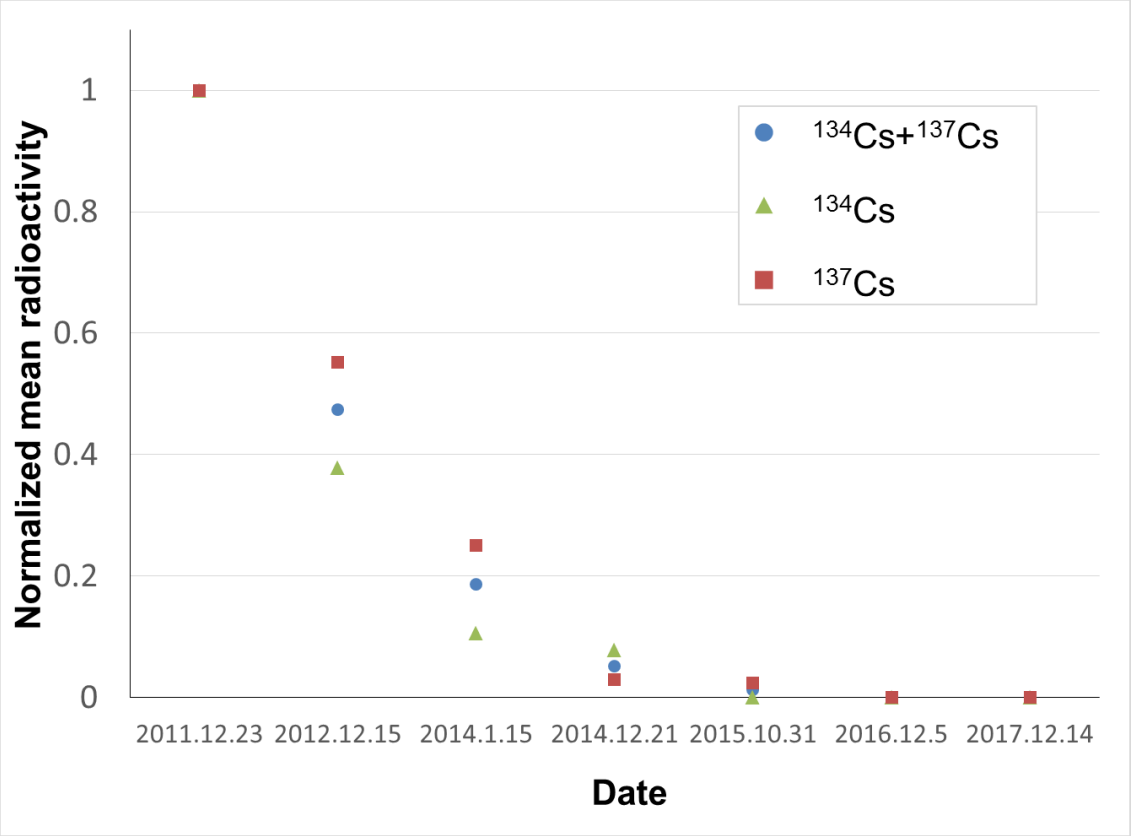

Figure 3-1-2. Variation of normalized average radioactivity of cesium isotopes

3.2 Average air dose rate and average ambient dose equivalent rate of cedar male flower sampling place

3.2.1 Average air dose rate

Four sites, where cedar male flower was collected, were taken as measurement points and the average air dose rate was measured at a height of 1 m above the ground. After averaging them again, table 3-2 shows the results expressed in [μSv h^{-1}]. Refering to the results of the travel survey published by the Ministry of Education, Culture, Sports, Science and Technology as mentioned above, the air dose rate of 1 m high on the ground surface near the cedar forest selected for the measurement point was the result that it was less than 0.1μSv h^{-1} all day before and after the schedule when we collected cedar male flower[15]. It shows the same result as our survey. Figure 3-2-1 shows a graph showing the average air dose rate at measurement points for each fiscal year.

Table 3-2. Average air dose rate at measurement points (1m high)

Measurement date	1m high [μSv h^{-1}]
2011.12.23	0.10
2012.12.15	0.09
2014.1.15	0.07
2014.12.21	0.06
2015.10.31	0.05
2016.12.5	0.05

* No survey has been made on 2017.12.4.

3.2.2 Average ambient dose equivalent rate

The distribution of incident direction of radiation resulting from the source distribution in the environment is roughly divided into three types of global isotropic, hemispherical isotropic, and horizontal isotropic incidence. In either case, the direction of human body is not fixed, and the azimuth angle can be regarded as isotropic as a group as an average. Under such a source distribution state, the effective dose equivalent has no large energy dependency in the energy range of 50 keV to 3 MeV as compared with the air absorbed dose. Thus, it is possible to evaluate the effective dose equivalent without great error via the conversion factor from the air absorption dose even if the energy is unknown. Furthermore, when the energy spectrum does not change very much like natural gamma rays, it can be converted and evaluated with one conversion factor[31]. Since this study targeting environmental radiation, we converted it into the air dose rate [nGy h^{-1}] using the effective dose conversion factor 0.748 Sv Gy^{-1} evaluated by Moriuchi et al[32]. Figure 3-2-2 shows a graph of the average air dose equivalent rate for each fiscal year. Although there is a moderate declining trend from FY 2011 to FY 2013, it decreased by half by 2015 and there was no significant change in the average air dose equivalent rate at the measurement point thereafter.

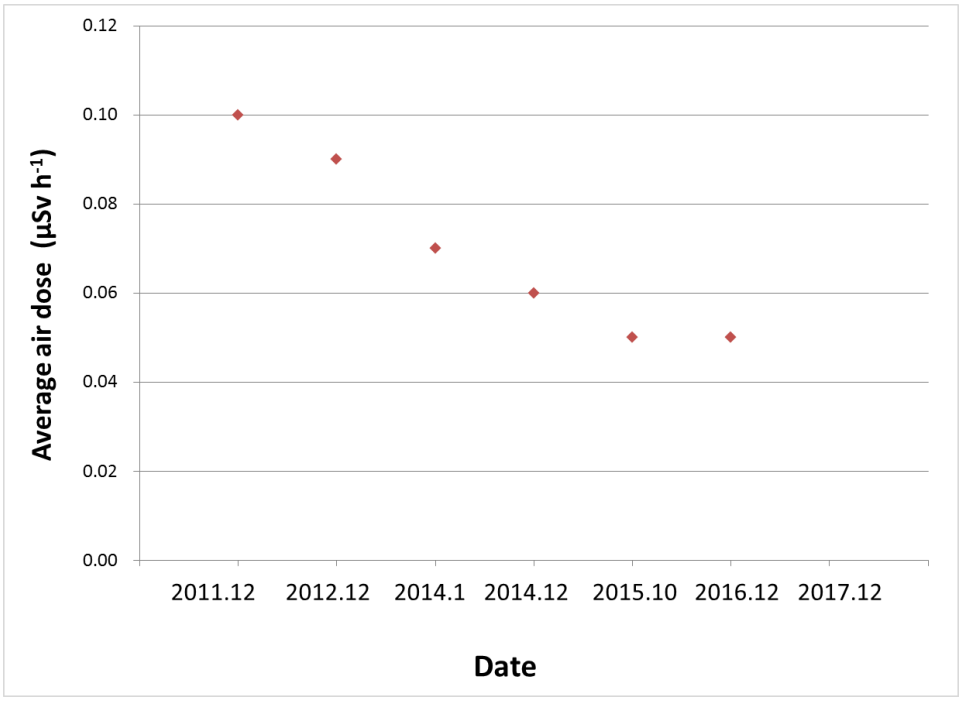

Figure 3-2-1 Variation of average air dose rate at measurement points

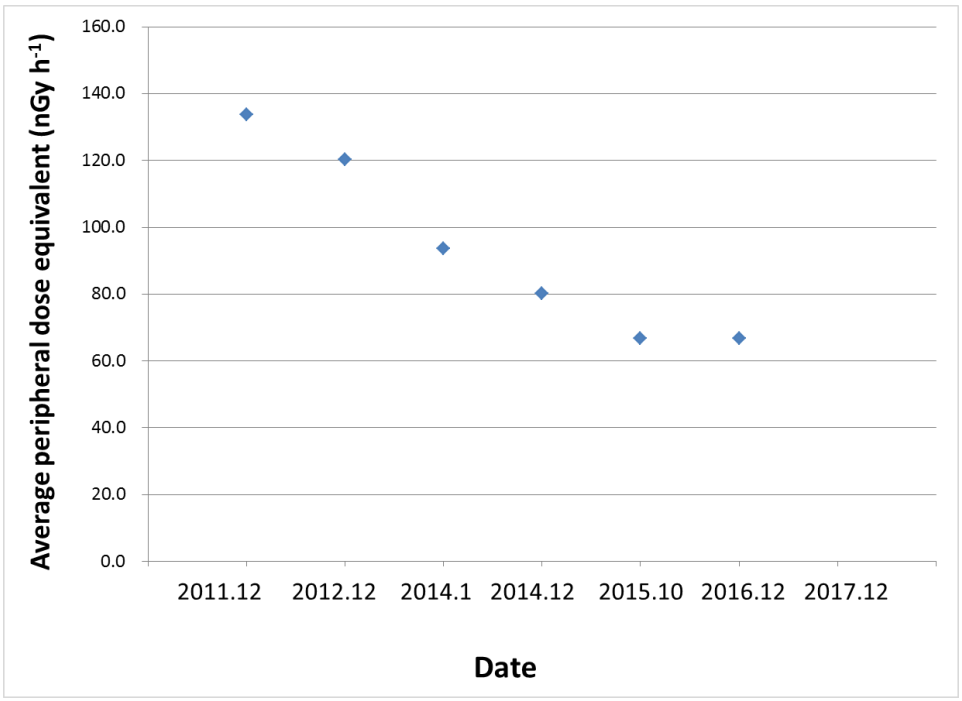

Figure 3-2-2 Variation of average air dose equivalent at measurement points

3.3 Calculation of radiation dose from cedar pollen

3.3.1 Radioactive cesium concentration in cedar pollen dispersed in the atmosphere

Based on the obtained results, the exposure dose per adult in the pollen scattering period, which is the period from February to May, was calculated and was used as the pollen exposure dose for each fiscal year. Assuming that the male flowers and pollen of the survey results for 2011, which had the highest value, had the same radioactive cesium concentration, radioactivity concentrations of ^{134}Cs and ^{137}Cs in cedar pollen are 86.5Bq kg^{-1} at ^{134}Cs and 114.9 Bq kg^{-1} at ^{137}Cs. There is a record of 2,207 m^{-3} as the highest monthly atmospheric concentration of cedar pollen measured in the Kanto district atmosphere in the past[33]. This was measured at the forest tree breeding field in Gunma prefecture in March 2008. Considering the possibility of cedar pollen scattering at the same concentration also in Tokyo, although it is data in the Kanto mountains area region, it was adopted as the value of concentration of cedar pollen atmospheric concentration so as not to underestimate the radiation dose. Here, when the weight of individual cedar pollen is 12 ng on average[28], the concentrations of radioactive cesium contained in cedar pollen that scatter in the atmosphere are calculated below.

In ^{134}Cs

$$86.5\ [Bq\ kg^{-1}] \times 2{,}207\ [m^{-3}] \times 12\ [ng] = 2.29 \times 10^{-6}\ [Bq\ m^{-3}] \quad (3\text{-}1)$$

In ^{137}Cs

$$114.9[Bq\ kg^{-1}] \times 2{,}207\ [m^{-3}] \times 12\ [ng] = 3.04 \times 10^{-6}\ [Bq\ m^{-3}] \quad (3\text{-}2)$$

3.3.2 Adult respiration rate and radioactive cesium effective dose coefficient

Then it is necessary to multiply the daily air inhalation amount of adults and the effective dose coefficient of radioactive cesium. The amount of air that an average adult inhales in a day's life depends on race, age, sex, weight, and activity. This involves breathing rate, which is the respiration rate per unit time, and it is an important factor in evaluating internal exposure by inhalation ingestion. For the average daily respiration rate, various data are indicated[34], among the International Commission on Radiological Protection publication (ICRP publication), there are differences in parameters used for calculation and settings of multiple breathing rates that differ depending on the subject (table 3-3-1) [35]. As the average respiration rate of Japanese men and women, there is a representative one as a parameter of dose evaluation of radon by the National Institute of Radiological

Sciences (NIRS), and its value is 17.3m^3 d^{-1} [36]. This is obtained by multiplying the behavioral respiratory rate data of ICRP publication 23 by 0.9 and weighting by the time zone of living. Besides this, there is a seminar presentation by Honda that calculated age-specific pulmonary ventilation by calculation, yet both are less than the air inhalation amount 22.2 m^3 d^{-1} for an adult per day for ICRP publication 71[37]. These results suggest that the respiration rate of Japanese may be lower than that of Westerners, because the mass of Japanese target tissue is smaller compared with Westerners, it is thought that the dose per unit intake is not much affected[38]. Therefore, in order not to underestimate the exposure dose, and also because the Forestry Agency is using "22.2 m^3 d^{-1} for air adsorption per day for adults as the numerical value of the International Radiological Protection Committee (22.2 m^3)," so this study as well used it for calculation.

Table 3-3-1 Respiration rate configurations in ICRP Publ.23, 68, 71

Source	Respiration rate	Remarks	Target
ICRP Publ.23 (1975)	20 L min^{-1} (=1.2 m^3 h^{-1})	normal person at light work	Worker
	2.3×104 L/day (=0.96 m^3 h^{-1})	Daily Respiratory volume of a normal person (8h of light work, 8h of daily activity, 8h at rest)	Resident
ICRP Publ.68 (1994)	1.2 m^3 h^{-1}	Average value 2.5h of seating (Respiratory volume 0.54 m^3 h^{-1}) and 5.5h of light work (Respiratory volume 1.5 m^3 h^{-1}) (2.5×0.54+5.5×1.5) / (2.5+5.5) = 1.2 (m^3 h^{-1})	Worker
ICRP Publ.71 (1995)	22.2 m^3 / d^{-1} (= 0.93 m^3 h^{-1})	Respiratory volume average of sleeping (0.45 m^3 h^{-1}) , seating (0.54 m^3 h^{-1}) , light work (1.5 m^3 h^{-1}) , hard work (3.0 m^3 h^{-1}) 8.0×0.45+6.0×0.54+9.75×1.5+0.25×3.0 = 22.2 m^3 d^{-1} 22.2 m^3 d^{-1} / 24h d^{-1} = 0.925 m^3 h^{-1}	Resident

The effective dose coefficient of radioactive cesium was calculated using (^{134}Cs: 0.020×10^{-3} Sv Bq^{-1}, ^{137}Cs: 0.039×10^{-3} Sv Bq^{-1} [39]), which is the effective dose coefficient due to adult inhalation. Then, we calculated with consideration the adult respiration rate and effective dose factor. The radiation dose rate per day with radioactive cesium is below.

In ^{134}Cs

$$2.29 \times 10^{-6}\ [\mathrm{Bq\ m^{-3}}] \times 22.2\ [\mathrm{m^3\ d^{-1}}] \times 0.020\ [\mathrm{Sv\ Bq^{-1}}] \quad (3\text{-}3)$$

$$= 1.02 \times 10^{-6}\ [\mu\mathrm{Sv\ d^{-1}}]$$

In ^{134}Cs

$$3.04 \times 10^{-6}\ [\mathrm{Bq\ m^{-3}}] \times 22.2\ [\mathrm{m^3\ d^{-1}}] \times 0.039\ [\mathrm{Sv\ Bq^{-1}}] \quad (3\text{-}4)$$

$$= 2.63 \times 10^{-6}\ [\mu\mathrm{Sv\ d^{-1}}]$$

3.3.3 Radiation dose per adult by inhalation of cedar pollen

Daily exposure dose for each nuclide obtained from Eq. (3) and Eq. (4) was summed up and multiplied by 120 days from February to May of pollen scattering, to be the radiation dose by inhalation of cedar pollen.

$$(1.0 \times 10^{-6}\ [\mu\mathrm{Sv\ d^{-1}}] + 2.63 \times 10^{-6}\ [\mu\mathrm{Sv\ d^{-1}}]) \times 120\ [\mathrm{d}] \quad (3\text{-}5)$$

$$= 4.38 \times 10^{-4}\ [\mu\mathrm{Sv}]$$

Therefore, the exposure dose (^{134}Cs+^{137}Cs) received per adult by inhalation of Japanese cedar pollen during the splashing period of cedar pollen was 4.38×10^{-4} μSv. Table 3-3-2 shows the trial calculation of radiation dose per pollen season in 2011. The exposure dose rate per unit time is the exposure dose rate per day divided by 24.

Table 3-3-2 Estimated exposure dose by cedar pollen in a healthy adult (with data measured in 2011)

Classification		^{137}Cs	^{134}Cs
Concentration of radioactive cesium contained in the cedar pollen		114.9 Bq kg^{-1}	86.5 Bq kg^{-1}
Atmospheric concentrations of airborne cedar pollen		2,207 m^{-3}	
Weight per piece of cedar pollen		1.20×10^{-11} kg	
Concentration of radioactive cesium contained airborne cedar pollen in the air		3.04×10^{-6} Bq m^{-3}	2.29×10^{-7} Bq m^{-3}
Radiation dose inhaling the atmosphere above in a healthy adult	Hour	1.52×10^{-7} μSv h^{-1}	
	Total period (February to May)	4.38×10^{-4} μSv	

Assuming the same conditions as above, the radiation dose when adults in Fukushima inhale pollen is calculated by quoting 1.08×10^{5} Bq kg^{-1} of ^{134}Cs and 1.45×10^{5} Bq kg^{-1} of ^{137}Cs which is the highest concentration of radioactive cesium in

cedar male flower measured in Fukushima Prefecture in 2011[19], and it is 0.551 μSv, that is 4210 times higher than the result of trial calculation I did in Ome city in the same year. Likewise, when comparing by calculating from 5.8×10^4 Bq kg^{-1} of ^{134}Cs and 1.7×10^4 Bq kg^{-1} [40] of ^{137}Cs which are the highest radioactive cesium concentration in male cedar flower measured in Fukushima Prefecture, total radiation dose was 0.205 μSv, which was 1,140 times higher than my calculation Ome city in Tokyo based on my measurement in the same year. Moreover, when comparing it by calculating from 4.5×10^4 Bq kg^{-1} of ^{134}Cs and 1.7×10^4 Bq kg^{-1} of ^{137}Cs[42] those are the highest concentrations of radioactive cesium in male cedar measured in Fukushima prefecture in fiscal 2013, total radiation dose was 0.140 μSv, which was 1,060 times higher than my calculation on Ome city in Tokyo based on my measurement.

Chapter 4　Consideration

4.1　Calculation of the ecological half-life of radioactive cesium in the Ome City cedar forest

According to the description in 3.1.2 of Chapter 3, the radioactivity concentrations of ^{134}Cs and ^{137}Cs in cedar pollen were halved after 1 year compared with the first year after the accident, and it was found to be reduced by 1/5 after 2 years. Since it attenuated earlier than the physical half-life of ^{134}Cs and ^{137}Cs, the disintegration constant and ecological half-life of ^{134}Cs + ^{137}Cs in the cedar forest in Ome city, Tokyo were calculated using the following expression[42] .

$$D_{total} = D_S\,(exp\,(-\lambda_S t)) + D_L\,(exp\,(-\lambda_L t)) \quad (1)$$

$$T_{eco} = 0.693 / \lambda \quad (2)$$

Here, D_{total} is the air absorption dose rate by artificial radiation nuclides, Ds is the initial air absorption dose rate by short half-lives nuclides such as ^{131}I, D_L is the initial air absorbed dose rate due to long half-life nuclides such as ^{134}Cs + ^{137}Cs, λs is the disintegration constant of short half-life nuclides, λ_L is the disintegration constant of the long half-life nuclide, t is the number of years elapsed since March

21, 2011[43] which is the day when the radioactive plume (plume) arrived in Kanto area. Plume refers to the flow of air masses containing gas and dust existing in nature from ancient times[7] . It is considered that a large amount of radioactive material declined in the land area on March 21 due to the large precipitation on this day[44] . Here, as of December 2011 when I started investigating cedar pollen in this study, it is considered that the contribution of short half-life nuclides was negligibly small. Considering only long half-life nuclides, I determined the disintegration constant and the ecological half-life from the following formula.

$$D_{total} = D_L\,(exp\,(-\,\lambda_L t)) \tag{1'}$$

$$T_{eco} = 0.693 / \lambda_L \tag{2'}$$

As a result, the disintegration constant and ecological half-life were 0.0016 and 1.16 years, respectively. Referring to the results of the survey at Namie Town in Fukushima Prefecture announced by the Forestry Agency, the trend of radioactive cesium concentration in cedar pollen decreased by 25% from 253 kBq kg^{-1} in 2011 to 90.5 kBq kg^{-1} in 2012, 59 kg^{-1} in 2013, which is 77% lower than the value in 2011[25] [40] [41] . From these report results, the ecological half-life is calculated using the

formulas (1 ') and (2') to be 1. 27 years, which shows the ecological half-life close to the result in Ome city obtained in this study.

4.2 Consideration on behavior of radioactive cesium in Ome city

4.2.1 Soil fixation of radioactive cesium

Cesium belongs to first group element, easily loses orbital electrons of the outermost shell and becomes an ionic state of Cs^+, so it is adsorbed to a site with negative charges present in the soil. This is why the cesium descended to the soil surface layer tends to stay near the surface layer without penetrating into the deep soil even by rainfall[45]. Most of the radioactive cesium adsorbed to the soil becomes strongly bonded with the clay mineral and many of them are bound to the silicon atom and are held in the soil and the amount of radioactive cesium that dissolves in water decreases with the passage of time[46]. Since plants absorb nutrients dissolved in water mainly through roots, it is believed that the amount of radioactive cesium absorbed by plants also decreases at the same time[47]. Generally, it is said that the ecological half-life of radioactive cesium in forests is on the order of 10 to 100 years compared with farmland etc. because of its long term retention[48]. However, these research results are aimed at the dynamics of Russia's vast coniferous forest (Taiga)

which exists in the Eurasian Continent. The ecological half-life of the radioactive cesium concentration in cedar pollen of Ome City and Namie Town obtained from this survey is 1.16 years and 1.27 years respectively, which was shorter than the data by Western literature which had been reported so far. Therefore, we focused on Japanese geography, soil composition and climate.

4.2.2 Soil composition in Ome city Tokyo and Fukushima prefecture

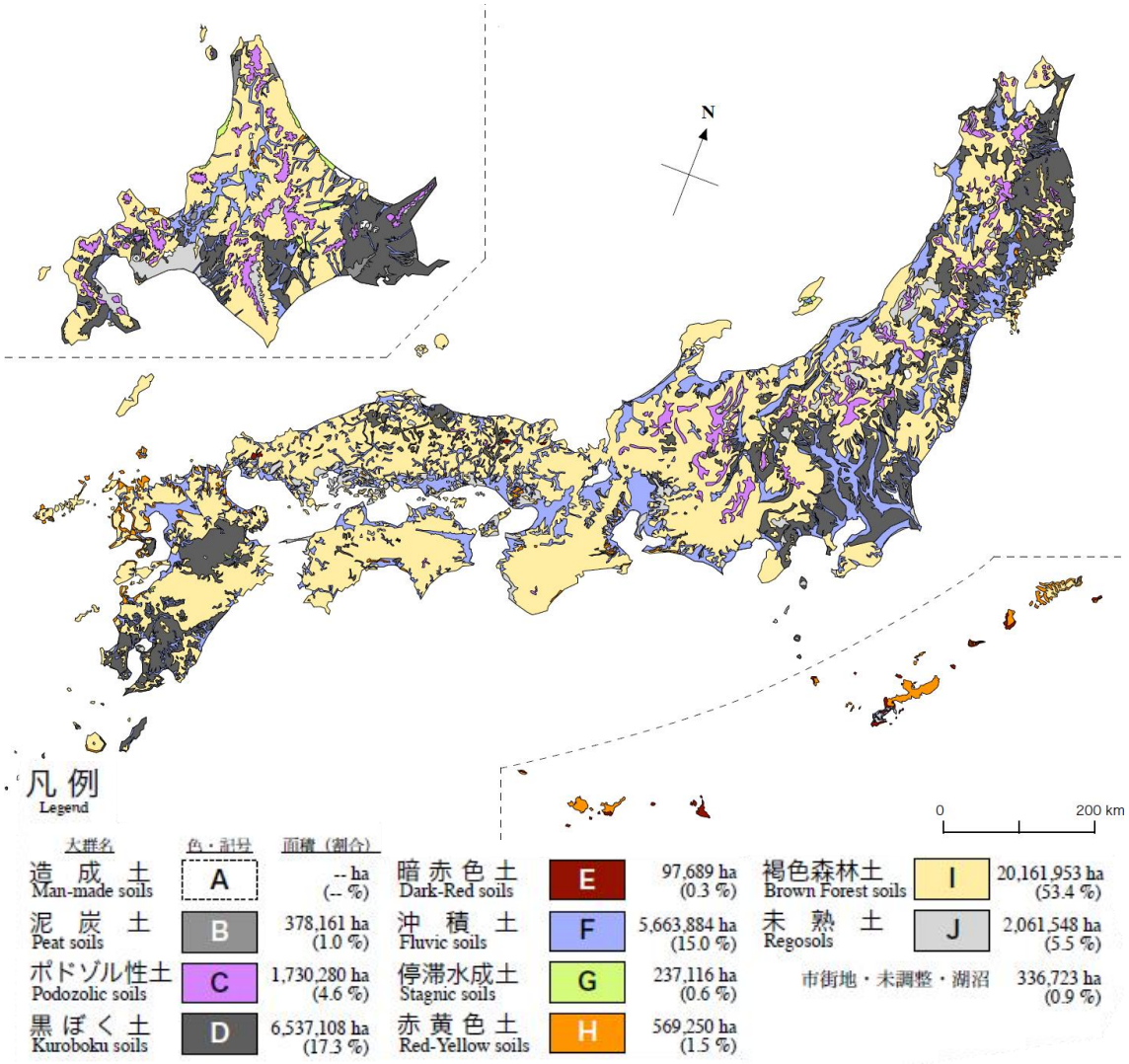

Figure 4-2-1 Japanese soil composition[49)]

Figure 4-2-1 shows a map of Japan's soil composition classified into 10 soil types as a map[49)]. Kuroboku soil and Fluvic soil are present around the Tokyo metropolitan soil composition, brown forest soil spreading to the west side of the Tokyo metropolitan area around Ome city as a border. Brown forest soil and

Kuroboku soil are mixed in soil composition in Fukushima prefecture. Figure 4-2-2 is a part of the soil map of Ome City, as indicated by the Economic Planning Agency. What flows in the center of the map is the upper stream area of the Tama River. The area indicated by the symbol Sng indicated on the coast of the river represents an area classified as soil consolidation called Sengase-to, the area indicated by the symbol Ong represents an area classified as soil consolidation called Ongata-to[50]. Among them, the Sengase-to area is composed of gravel-brown forest soil clearly mixed with Kuroboku soil. The fine grain filling the gap between the gravels is blackish brown and the inclusion of Kuroboku soil is clear, however relatively hard granular to small agglomerate structures are developed and there is no light and soft grain structure characteristic of Kuroboku soil. The Akiru-to soil consolidation which also contains contamination of black soil is different depending on whether it is gravel or not. It is slightly wider along the Tama River in Ome City, and it is distributed in a narrow band along the Hirai River in the Akiru terrace. In addition, Ongase-to is brown forest soil on the terrace surface where all layers or less than 30 cm is gravel. Gravel is mainly circle · half-pointed small gravel to intermediate gravel, and the narrow filling clay is mainly clayish earth or mucilage. It emerges in the bottom level surface (Aoyagi surface and lower terrace surface) without influence of volcanic ash along the Tamagawa upstream, Hirai River, Akikawa River, Asakawa River[51].

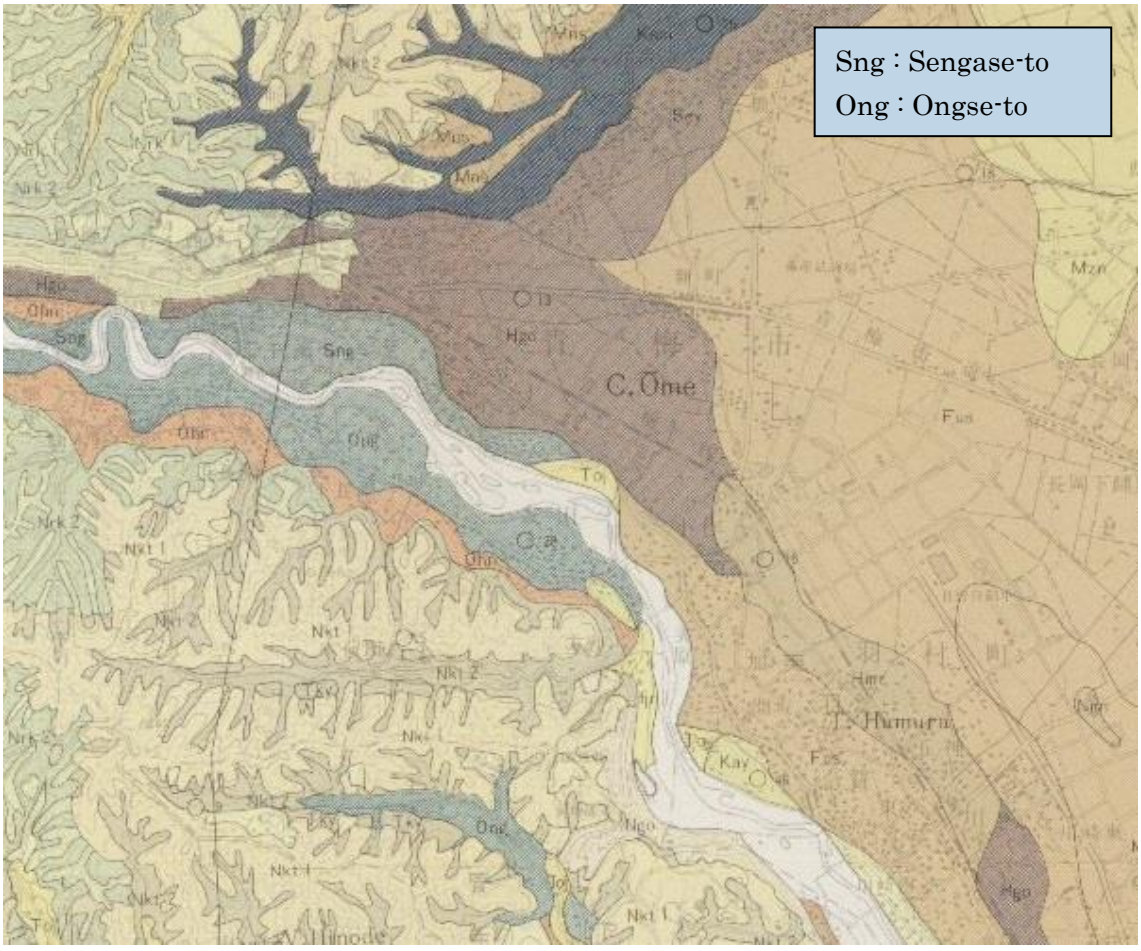

Figure 4-2-2 Soil map of Ome city in Tokyo Okutama area[50].

Then, the photographs of the soil around the trees from which we collected the cedar male flowers of are shown in Figure 4-2-3 to 4-2-6. All 4 pictures shows soil compositions containing brown forest soil on Kuroboku soil that tends to contain gravel. From the mixed situation of Kuroboku soil, it is judged that the area that was the subject of the survey this time belongs to the Sengase-to area.

Figure 4-2-3 Soil at point 1

Figure 4-2-4 Soil at point 2

Figure 4-2-5 Soil at point 3

Figure 4-2-6 Soil at point 4

4.3 Radioactive cesium behavior in Kuroboku soil

According to the soil sampling result after the Fukushima Daiichi nuclear power plant accident in the Kuroboku soil field in the Agricultural Environment Technology Research Institute, despite several rainfalls before sampling it has been reported that ^{137}Cs remained mostly within a few cm of soil surface layer[52]. Radioactive cesium brought by the Chernobyl accident also has many reports that

the lower penetration rate is extremely slow and remains near the surface layer[53]. Also in the cedar forest that investigated in this study is highly likely which radioactive cesium remained at the very surface layer of the soil, and it was considered to be in a state susceptible to the influence of wind and rain erosion. Firstly, the influence of windy eating where dry soil is carried by the wind is considered. In addition, the cedars targeted for survey were vegetated on steep slopes. On the Pacific side of Japan, it is susceptible to rainfall such as rainy season and typhoon, so most of radioactive cesium that remained on the surface of the soil was washed off with rainy soil watering. It is considered that the washed soil goes down the slope, and finally it flows into the Tama River which flows 70 meters north of the measurement point. Radioactive cesium takes two forms of dissolved and suspended in river water[55]. Radioactive cesium moved into water contaminates plants by its root absorption[56].

Chapter 5 Conclusion

5.1 Conclusion

Radioactive cesium concentration in cedar pollen in Ome City decreased to 47% on average to 66.6 Bq kg^{-1} in December 2012 compared to December 2011. In January 2004, the average was 26.1 Bq kg^{-1} and it was 19%. In December 2014, the average value was 7.2 Bq kg^{-1}, which was 5%, and in October 2015 it was 1%, an average of 1.9 Bq kg^{-1}. After 2016, the measured value is less than 0.0 Bq kg^{-1} and it seems to have reached the plateau.

This result is similar to the over time change of average value of the survey results in Fukushima Prefecture announced by the Ministry of Agriculture, Forestry and Fisheries. According to the press release announced on January 31, 2014, when comparing the measured values in Fukushima prefecture as a whole, the concentration declined to about a half of that was in fiscal 2011 in fiscal 2012, furthermore, it is announced that it has decreased to about one fifth in fiscal 2013[41]. According to the announcement in 2016, it is reported that it is reduced to 10% in 2014 and it has decreased to 5% in 2015[57]. The results of the survey in 2016 and 2017 have not yet been published (as of the end of December 2017). This is the result of describing the average value of the whole area of Fukushima Prefecture,

now we would like to focus on the result of Namie town where radioactive cesium concentration in cedar pollen shows the highest value every year. Table 5-2-1 shows the trend of radioactive cesium concentration in Namie Town from 2011 to 2015, indicating the following trend. When comparing the highest value in 2011 with the highest value in each year afterwards, it is about 35% in 2012. Then it was 23% in 2013, 8% in 2014, and in 2015 it fell to a concentration of about 3%. Focusing only on this one point, it seems that it is different from the attenuation tendency of radioactive cesium concentration in cedar pollen in Tokyo. The actual survey site is selected so that it is evenly distributed from the high air dose rate to the low point. Because there are 24 measurement points throughout Fukushima prefecture, the average value of all of them has a similar tendency in case compared with Tokyo.

Table 5-2-1. Radioactive cesium concentration in cedar tree of Namie town[q)]

Measurement year	Radioactive cesium concentration in cedar pollen [Bq kg^{-1}]
2011	253,000
2012	90,500
2013	59,000
2014	20,500
2015	8,800

A river or a waterway flows in all the districts in 24 areas indicated as the location of the cedar forest where the Ministry of Agriculture, Forestry and Fisheries conducted the survey site, and the main Japanese cedar tree is vegetated

on inclined land[41]. As Japanese cedar prefers water and nutrient environments such as along the creek, it is because it is often planted in a slightly damp place from the valley and the slope to the middle part during artificial afforestation. Moreover, the soil composition in Fukushima prefecture is mainly brown forest soil and Kuroboku soil[58]. From the viewpoint of weather conditions Fukushima Prefecture and Tokyo belong to the same Pacific climate classification[59]. Even in the survey conducted by the Ministry of Agriculture, Forestry and Fisheries, they changed the sample collection part slightly from the vegetation situation of cedar trees, abandon sampling due to difficulty in entering to the forest, the situation shows the difficulty of investigation against the natural environment.

There is a difference between the dynamics of radioactive cesium derived from the Chernobyl nuclear accident that has been published so far and the dynamics in Japan. It is thought to be due to climate and terrain peculiar to Japan. In terms of accidents in Fukushima, it can be considered that direct soil was not contaminated as a result of vegetation of cedar forest, which worked in a good direction from the viewpoint of decontamination work. Decontamination becomes difficult if radioactive cesium adheres to the soil, but radioactive cesium descending to the leaf surface of cedar is diffused by pollen and it can be said that it worked in the direction of decreasing ambient air dose rate. Cesium and potassium have similar properties though, cesium is not an essential element for plants. Therefore, it seems

that the plant tried to lower the concentration of cesium in the body by using pollen as a kind of excretory organ. It also supports this that cesium contained in trees easily migrates to new growth sites such as fruits, sprouts, and young leaves[60]. It can also be said that cedar trees planted before the accident indirectly worked as phytoremediation.

Research on whether radioactive cesium that was diffused by pollen or flowed into the river due to soil watering or wind erosion might return to the Japanese cedar forest over the larger circulation would require a longer span investigation. We would like to continue the survey in the future.

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Achieved papers

1. H. Tsuruoka, K. Inoue, Y. Sakano, M. Hamada, H. Shimizu, M. Fukushi
   VARIATION OF CESIUM CONCENTRATIONS IN CEDAR POLLEN AT OKUTAMA AREA AFTER THE FUKUSHIMA DAIICHI NUCLEAR POWER PLANT ACCIDENT
   Radiation Protection Dosimetry pp. 1–4 (2015)

2. H. Tsuruoka, Kazumasa Inoue, Shota Hosokawa, and Masahiro Fukushi
   Measurement of radon and thoron concentrations in the Tokyo Metropolitan University Arakawa Campus building
   The Journal of Japan Academy of Health Sciences p40~48 (2016)

3. 雀岡　大
   奥多摩地区における福島第一原発事故後のスギ花粉中セシウム量の推移
   首都大学東京大学院 修士論文 (2015)

4. Tan Van Le, Kazumasa Inoue, Hiroshi Tsuruoka, Makoto Fujisawa, Moeko Arai, Linh Dai
   EFFECTIVE DOSE DUE TO TERRESTRIAL GAMMA RADIATION ESTIMATED IN SOUTHERN VIETNAM BY CAR-BORNE SURVEY TECHNIQUE
   Radiation Protection Dosimetry pp. 1–8 (2017)

5. 清水秀雄，石田和雄，前寺郁彦，雀岡大、井上一雅、福士政広
   福島第一原子力発電所周辺における空間線量率とモクズガニ甲殻へのセシウム集積との関係性の検討
   医療保健学研究　7 号 p1~6 (2016)

6. Kazumasa Inoue, Hiroshi Tsuruoka, Tan Le Van, Moeko Arai a, Kyoko Saito, Masahiro Fukushi
   Impact on ambient dose rate in metropolitan Tokyo from the Fukushima Daiichi Nuclear Power Plant accident
   Journal of Environmental Radioactivity p158~159 (2016)

7. Kazumasa Inoue, Hiroshi Tsuruoka, Tan Le Van, Masahiro Fukushi
Contribution ratios of natural radionuclides to ambient dose rate in air after the Fukushima Daiichi Nuclear Power Plant accident
Journal of Radioanalytical and Nuclear Chemistry (2015)

8. F. Maedera, K. Inoue, M. Sugino, R. Sano, H. Shimizu, H. Tsuruoka, and M.Fukushi.
NATURAL VARIATION OF DOSE RATE IN AIR AT IZU-OSHIMA ISLAND AFTER THE FUKUSHIMA DAIICHI NUCLEAR POWER PLANTS ACCIDENT
Radiation Emergency Medicine Vol.4 No.1 (2015)