VARIATION OF RADIOCESIUM CONCENTRATIONS IN CEDAR POLLEN IN THE OKUTAMA AREA SINCE THE FUKUSHIMA DAIICHI NUCLEAR POWER PLANT ACCIDENT

H. Tsuruoka[1], K. Inoue[1], Y. Sakano[1], M. Hamada[1], H. Shimizu[1], M. Fukushi[1,*]

[1]Department of Radiological Sciences, Tokyo Metropolitan University, 7-2-10 Higashiogu, Arakawa-ku, Tokyo 116-8551, Japan

Received month date year, amended month date year, accepted month date year

Due to releases of radionuclides in the Fukushima Daiichi Nuclear Power Plant Accident, radiocesium (^{134}Cs and ^{137}Cs) has been incorporated into large varieties of plant species and soil types. There is a possibility that radiocesium taken into plants is being diffused by pollen. Radiocesium concentrations in cedar pollen have been measured in Ome City, located in the Okutama area of metropolitan Tokyo, for the past 3 years. In this research, the variation of radiocesium concentrations was analyzed by comparing data from 2011 to 2014. Air dose rates at 1 m above the ground surface in Ome City from 2011 to 2014 showed no significant difference. Concentration of ^{137}Cs contained in the cedar pollen in 2012 was about half that in 2011. Between 2012 and 2014, the concentration decreased by approximately one fifth, which was similar to the result of a press release distributed by the Japanese Ministry of Agriculture, Forestry and Fisheries.

INTRODUCTION

In the Fukushima Daiichi Nuclear Power Plant (F1-NPP) accident, large amounts of artificial radionuclides such as ^{131}Xe, ^{131}I, ^{134}Cs and ^{137}Cs were released to the environment(1). Among them, cesium belongs to the group of elements known as the alkali metals. Therefore, radiocesium (^{134}Cs+^{137}Cs) has similar properties to its congener potassium which plants require to grow. It is well known that radiocesium is absorbed by the roots from the soil after deposition, and specifically in cedar trees, it is known to migrate to the male flowers of the trees. Furthermore, the migrated radiocesium is re-distributed in the environment because of cedar pollen scattering(2). Cedar pollen particles are approximately 30 μm in diameter and they weigh approximately 12 ng, making them particularly lightweight(3). Occasionally, pollen particles remain airborne for distances of more than 200 km(4). Thus, citizens of the metropolitan Tokyo area may have been internally exposed to radiocesium by inhaling cedar pollen from trees growing in Tokyo and nearby prefectures areas, including especially Fukushima Prefecture. In order to estimate the amount of internal exposure from cedar pollen, since the accident in 2011, the Ministry of Agriculture, Forestry and Fisheries, has been continuously measuring the concentration of radiocesium contained in male cedar flowers at the 24 places in Fukushima Prefecture. The highest radiocesium concentration in Fukushima Prefecture was 253 kBq kg^{-1} that was observed in Komaru, Namie-Town, Futaba-gun in 2011(5). Survey results have been published by the Ministry of Agriculture, Forestry and Fisheries in a press release announced on February 8, 2012, for Hachioji City and Nishitama-gun in Tokyo(6), but no continuous radiocesium concentration measurements for cedar pollen have been carried out in Tokyo.

In this study, variation of the concentration of radiocesium contained in cedar pollen released from a cedar forest of Tokyo's Okutama area were investigated during the three years of 2011-2014. Ome City in the Okutama area was selected for collection of male cedar flowers; it satisfied the conditions of having extensive cedar tree plantations and having high deposition (Bq m^{-2}) of ^{134}Cs and ^{137}Cs in the soil, based on measurement results of airborne monitoring in the outskirts of Tokyo carried out in September 2011 by the Ministry of Education, Culture, Sports, Science and Technology(7).

MATERIALS AND METHODS

Well-grown cedar trees with male flowers that were found in a cedar forest in Ome City (Figure. 1) were selected for the study and four measurement locations were set in. At the measurement locations, male cedar flower parts were collected. Male cedar flowers had already matured by this period, and become a dormant state, showed a concentration similar to the pollen to be scattering. The male flowers were dried in a microwave oven over 15 minutes, keeping in mind that they were not to be overheated, then they were ground into a powder in a mortar. After weighing the ground flowers, the weighed powder was packed in a U-8 container.

*Corresponding author: fukushi@tmu.ac.jp

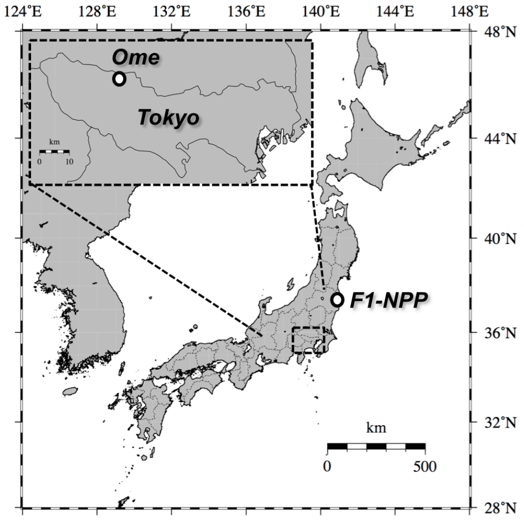

Figure 1. Map showing Ome City, located in the Okutama area of Tokyo where measurements were made.

Radioactivity concentration measurements were carried out for 30000 s using a germanium semiconductor detector (GMX10P, ORTEC, Oak Ridge, TN, USA). At the measurement locations where the male cedar flowers were collected, effective dose rate (μSv h^{-1}) was measured using a NaI(Tl) scintillation spectrometer (identi FINDER, ICX Technologies, Oak Ridge, TN) at a height of 1 m from the ground surface. Air dose rate (nGy h^{-1}) was calculated using a dose coefficient of 0.748 Sv Gy^{-1}[(8)]. For the measurement results, the mean value of 10 measurements made for 120 s consecutively at 10 s intervals, excluding the maximum and minimum values of the 12 obtained data were adopted.

RESULTS

The variation of the mean measured radioactivity concentrations of ^{134}Cs and ^{137}Cs in cedar pollen are shown in Figure 2. The sum of the radioactivity concentrations of ^{134}Cs and ^{137}Cs decreased 53% between December 2011 and December 2012 and the mean radioactivity concentration of January 2014 was reduced 78% compared to December 2011.

The variation of the air dose rates (nGy h^{-1}) at the locations where the male cedar flowers were collected is shown in Figure 3. The air dose rate was reduced by 17% between December 2011 and December 2012 and the air dose rate of January 2014 was reduced 27% compared with December 2011. No significant decrease was observed in comparison with the change in radioactivity concentration of cedar pollen shown in Figure 2.

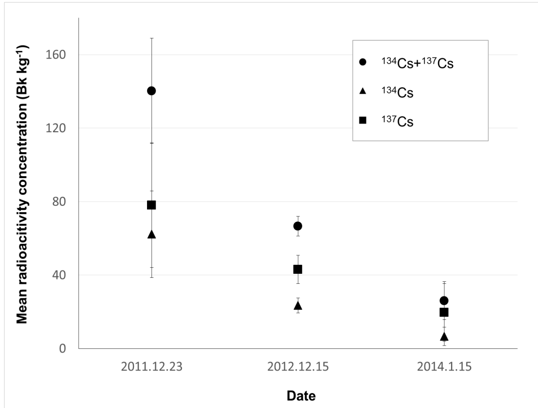

Figure 2. Variation of mean radioactivity of cesium isotopes.

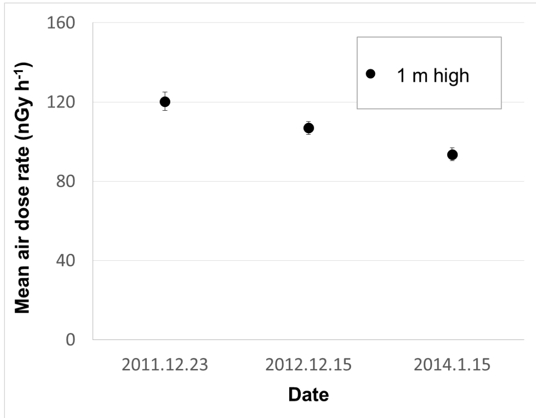

Figure 3. Variation of mean air dose rate at measurement locations.

DISCUSSION

Generally, it has been reported that the environmental half-life in a forest are longer than in farmland, since radiocesium stays in the soil for a long time, 10 to 100 years[(9)]. However, referring to the variation of radioactivity concentration in cedar pollen measured from 2011 to 2014 (Figure 2), attenuation was earlier than would be expected for the physical half-life of ^{134}Cs (2.06 years) and of ^{137}Cs (30.17 years). Then, the decay constant of ^{134}Cs+^{137}Cs and their environmental half-live (year) were calculated with Equation (1).

$$D = D_L\,(exp\,(-\,\lambda_L t)),\ \ T = 0.693\,/\,\lambda_L \quad (1)$$

In Equation (1), D represents the air absorbed dose rate (nGy h^{-1}) due to the artificial radionuclides. D_L represents the primal air absorbed dose rate due to the long-half-life radionuclides (^{134}Cs+^{137}Cs) λ_L represents the decay constant. *t* represents the years elapsed from the primal date when the radioactive plume from the F1-NPP arrived at Ome City (March 21, 2011[10]). As a result, the decay constant was 0.0016 and the environmental half-life was 1.16 years. Compared to the survey results on the variation of radioactivity concentrations in cedar pollen measured in Namie Town, that the Forestry Agency announced, the value was decreased by 64% from 253 kBq kg^{-1} in 2011 to 90.5 kBq kg^{-1} in 2012, then it declined 77% to 59 kBq kg^{-1} in 2013 from the 2011 value. Calculating the environmental half-life by Equation (1) showed the value in Namie Town Fukushima was 1.27 years which was similar to the value calculated in Ome City. It was assumed that the reasons why these environmental half-life terms were similar, that both Tokyo and Fukushima Prefecture belong to the same climate region[11], and they have a lot in common in the vegetation environments of their cedar forests referring to the locations shown in the press release published by Forestry Agency, Japan[12].

Figure 3 shows the air dose rates at locations were cedar pollen was collected; no significant variation was observed in comparison with the variation of radioactivity concentrations of cedar pollen. Also, referring to the air dose rates of a vehicle-borne survey published on the Extension Site of Distribution Map of Radiation Dose, etc. measured by Japan Atomic Energy Agency, Sector of Fukushima Research and Development[7], the values at 1 m high from the ground surface were below 0.1 μSv h^{-1} at the points and around the dates we have collected the cedar male flowers. Cesium belongs in a group known as alkali metals, easily becomes cation Cs^{+}, losing orbital electron of the outermost shell, and it is adsorbed to the parts with the negative charge in the soil. Therefore, cesium landed on the surface of soil are without being affected by the rainfall to penetrate deep into the soil, it remains in the vicinity of the surface layer[13]. According to the soil sampling results of after the F1-NPP accident, in the Andosol farmlands of the National Institute for Agro-Environmental Sciences, located in the Tsukuba, Ibaraki Prefecture it has been reported that, even though there were rain falls several times before sampling, most of ^{137}Cs had remained in the surface of the within soil several cm[13]. Thereafter, most of the radiocesium adsorbed on the soil surface and held in the soil is strongly bound with clay minerals, and the amount of radiocesium that leaches from the soil into rainwater and groundwater decreases with time. Mainly, plants are absorbing nutrients dissolved in water through the roots, so the amount of radiocesium that plants absorb is also decreasing at the same time as the leached radiocesium is decreasing[14]. It was assumed that, since the amount of radiocesium being absorbed into the plants leaching into rainwater and groundwater over time was decreased, the radioactivity concentration of cedar pollen was significantly reduced compared with the air dose rate.

Then, from the results above, the exposure dose was calculated from February to May (120 days) which is the scattering period of pollen. Radioactivity concentrations of ^{134}Cs and ^{137}Cs in the cedar pollen were: ^{134}Cs, 13.7 Bq kg^{-1}; ^{137}Cs, 40.6 Bq kg^{-1}; the highest reported cedar pollen number concentration scattered in the atmosphere of the Kanto region in the past was 2207 m^{-3}[15]. Assuming each cedar pollen particle weights 12 ng, the respective concentrations of ^{134}Cs and ^{137}Cs included in the cedar pollen scattered into the atmosphere were 3.63×10^{-7} Bq m^{-3} and 1.08×10^{-6} Bq m^{-3}. Subsequently, considering the air intake per day for a healthy adult individual as (22.2 m^3 d^{-1}) the effective dose coefficients were: ^{134}Cs, 0.020 μSv Bq^{-1}; ^{137}Cs, 0.039 μSv Bq^{-1}. Respective exposure doses per day of ^{134}Cs and ^{137}Cs were estimated to be 1.61×10^{-7} μSv d^{-1} and 9.31×10^{-7} μSv d^{-1}. For this reason, total exposure dose (^{134}Cs+^{137}Cs) received from the pollen scattering period (120 days) was estimated to be 0.13 nSv per pollen season. Under the same conditions, the total exposure was calculated as 0.14 μSv by referring to the highest radioactivity concentration in the cedar pollen measured in Fukushima Prefecture, and assuming an adult inhaled: 1.7×10^4 Bq kg^{-1} of ^{134}Cs and: 4.2×10^4 Bq kg^{-1} of ^{137}Cs.

Comparing the result measured in Ome City, the value measured in Fukushima was 1077 times higher. Radioactivity concentration (^{134}Cs+^{137}Cs) of male cedar flowers in Ome, Tokyo has decreased year by year.

The environmental half-life obtained in this study was used to estimate the radiocesium concentration in the cedar pollen and it would be under the detection limit (6.67×10^{-3} Bq kg^{-1}) within 1.7 years from September 2015.

REFERENCES

1. Hosoda, M., Tokonami, S., Sorimachi, A., Monzen, S., Osanai, M., Yamada, M., Kashiwakura, I. and Akiba, S. *The time variation of dose rate artificially increased by the Fukushima nuclear crisis*. Sci. Rep. 1, **87** (2011). doi: 10.1038/srep00087.
2. Forestry Agency, Japan Press release. *Survey result of the concentration of radioactive cesium contained in cedar male flower*. (2014). Available on http://www.rinya.maff.go.jp/j/press/kenho/pdf/130208_2-01.pdf (accessed 28 July 2014).
3. Forestry Agency, Japan Press release. *Survey result of the concentration of radioactive cesium contained in cedar male flower*. (2011). Available on http://www.rinya.maff.go.jp/j/press/hozen/pdf/111227-03.pdf (accessed 28 July 2014).
4. Tokyo Metropolitan Institute of Public Health, *Tokyo Metropolitan Institute of Public Health pollinosis note*. (2014). Available on http://www.tokyo-eiken.go.jp/kj_kankyo/kafun/ (accessed 22 September 2014).

5. Forestry Agency, Japan Press release. *Survey result of the concentration of radioactive cesium contained in cedar male flower (Interim report)*. (2011). Available on http://www.rinya.maff.go.jp/j/press/hozen/pdf/111227-01.pdf (accessed 28 July 2014).
6. Japan Atomic Energy Agency. *Outline version of the report of the research on radioactive materials second-order distribution associated with the Fukushima Daiichi nuclear power plant accident*. (2013). Available at http://www.jaea.go.jp/fukushima/kankyoanzen/mapping_report/2nd-japanese/summary.pdf (accessed 31 August 2014).
7. Ministry of Education, Culture, Sports, Science and Technology - Japan. *Extension site of distribution map of radiation dose, etc*. (2014). Available on http://ramap.jmc.or.jp/map/eng/ (accessed 31 August 2014).
8. Moriuchi, S., Tsutsumi, M. and Saito, K. *Examination on conversion factors to estimate effective dose equivalent from absorbed dose in air for natural gamma radiations*. Jap. J. Health. Phy. **25**, 121-128 (1990).
9. Shaw, G. *Radionuclides in forest ecosystems,* Radioactive. Environm. **10**, 127-155 (2007).
10. Morino, Y., Ohara, T. and Nishizawa, M. *Atmospheric behavior, deposition, and budget of radioactive materials from the Fukushima Daiichi nuclear power plant in March 2011*. Geophys. Res. Lett. **38**, L00G11 (2011). doi: 10.1029/2011GL048689.
11. Koizumi, K. and Kato, H. *Climatic division of Japan depending on the spatial variation pattern of climatic elements*. (2012). Available on http://www.chs.nihon-u.ac.jp/institute/nature/kiyou/2012/pdf/2_6.pdf (accessed 22 September 2014).
12. Forestry Agency, Japan Press release. *Survey result of the concentration of radioactive cesium contained in the 2013 male cedar*. (2014). Available on http://www.rinya.maff.go.jp/j/press/ken_sidou/pdf/140131-01.pdf (accessed 28 July 2014).
13. Yamaguchi, N., Takata, Y., Hayashi, K., Ishikawa, S., Kuramata, M., Eguchi, S., Yoshikawa, S., Sakaguchi, A., Asasda, K., Wagai, R., Makino, T., Akahane, I. and Hiradate, S. *Behavior of radiocaesium in soil-plant systems and its controlling factor*. National Institute for Agro-Environmental Sci. Rep. **31**, 75-129 (2012).
14. Tsukada, H., Takeda, A. and Hasegawa, H. *Uptake and distribution of ^{90}Sr and ^{137}Cs in rice plants*. 16th Pacific Basin Nuclear Conference. Aomori, Japan. **39**, 149-160 (2008).
15. Ministry of the Environment. *Ministry of the environment pollen observation system*. (2011) Available on http://kafun.taiki.go.jp/ (accessed 31 August 2014).