Effective Half-Lives of Radiocesium in Terrestrial Plants Observed After Nuclear Power Plant Accidents

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Abstract Information on “how fast radiocesium could be removed from plants” is important to decide measures after contamination. To express the decreasing rate, effective half-life (T_{eff}) can be used; T_{eff} is defined as the time required for a 50 % decline of radiocesium in an individual plant or plant population in a natural ecosystem. There are four major radioactivity decrease process in plants when terrestrial environment is contaminated with radiocesium. (1) Removal of radiocesium attached to plant surface by rain and wind (weathering effect), (2) plant mass increase at growing season (dilution effect), (3) removal of radiocesium by shedding leaves and fruits (elimination effect) and (4) decrease of bioavailable radiocesium fraction in soil (aging effect). It is difficult to separate these processes, however, since radiocesium decreasing trend fits well with exponential curves (usually a combination of two exponential curves); effective half-lives (T_{eff}) are reported. This paper summarizes terrestrial plant T_{eff} data in woody and herbaceous plants observed after Fukushima nuclear accident and also compared the data observed after the Chernobyl power plant accident.

Keywords Effective half-life • Fukushima Daiichi Nuclear Power Plant accident • Chernobyl Nuclear Power Plant accident • Herbaceous plants • Woody plants

1 Introduction

The magnitude-9.0 (Richter scale) earthquake on March 11, 2011 triggered a huge tsunami that hit the Pacific Ocean coast of Eastern Japan. The tsunami destroyed the water circulation systems of the Fukushima Daiichi Nuclear Power Plants (FDNPP). Consequently, agricultural crops as well as many other plants became contaminated with radioactive fallout from the accident (Ministry of Health, Labour and Welfare

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(MHLW) 2016; Tagami et al. 2012). Several months later, radiocesium with relatively longer physical half-lives (T_{phy}), i.e., ^{134}Cs ($T_{phy} = 2.06$ years) and ^{137}Cs ($T_{phy} = 30.2$ years) were the only major radiation source to humans remaining in the environment. Although contribution to dose is negligible, it is also necessary to consider ^{135}Cs for its long half-life of 2.3×10^6 years (Zheng et al. 2014).

In the natural environment, decreases of radiocesium concentration in plants with time are usually found after the releases to the atmosphere by nuclear bomb testing and accidental releases (Cline 1981; Komamura et al. 2005; Pröhl et al. 2006; Paller et al. 2014). The radiocesium decreases in plants is usually faster than the physical half-lives of long-lived radiocesium isotopes such as ^{137}Cs and ^{135}Cs. Although the phenomenon is known, if there is not enough numerical information on the decreasing rates in plants, it is difficult to decide measures to recover from contaminated situations. For example, if the decreasing was a slow process then replacement of trees might be considered to decrease dose to humans, but if it was a fast process then it is possible to let the trees be in their original positions without doing any additional remediation acts.

There are four major radioactivity decreasing effects in plants as follows:

Effect-1 (Weathering): Radiocesium released to the environment through the air should directly deposit on plants in particulate, gaseous and/or ionic forms, and then, due to weathering (rain and wind, etc.), some portion of total deposited could be washed off from the plant surface. This process is found in both woody and herbaceous plants.

Effect-2 (Dilution): In a plant growing season, if radiocesium uptake is fast enough to meets plant growth rates, then apparent concentration decrease would not be found. However, generally, decrease in radiocesium concentration due to plant mass increase was found; that is, dilution effect. This process is also found in both wood and herbaceous plants.

Effect-3 (Elimination): Total amount of radiocesium in a plant decreases when the plant removes its tissues such as leaves, fruits, barks, stems and roots because these tissue parts also include radiocesium. This yearly tissue removal event is found in woody and perennial herbaceous plants.

Effect-4 (Soil aging): Soil can retain most radiocesium, but the bioavailability change with time; generally, decreasing trend can be observed after radioactive contaminants were added to soil, which is called as aging effect (declining bioavailable percentage to total radiocesium). It is also necessary to consider radiocesium leaching from surface to deeper layer, and weathering of soil (total radiocesium decrease) affects the radiocesium concentration change with time. Increase of root uptake is the process to slow the T_{eff}, but decrease provide opposite effect.

On the other hand, foliar uptake of re-suspended contaminated soil or dust, radiocesium concentration in plant would increase due to foliar uptake which might slow the radiocesium decrease from the plant; however, the latter effect was inconsequential relative to root uptake process (Hinton et al. 1995). It is also necessary to consider recycle of radiocesium in a plant especially for woody and perennial herbaceous plants, for example, radiocesium in leaves partially translated to stems/branches before shedding (Tagami and Uchida 2015a), which could slow the decrease of radiocesium in the plant. It is difficult to separate these effects by observing radiocesium concentration in plant in natural ecosystems; thus including all these effects, the environmental half-lives (T_{env}) are observed.

2 Effective Half-life

The effective half-life (T_{eff}), which is defined as the time required for a 50 % decline of radiocesium in an individual plant or plant population in a natural ecosystem. After atmospheric release of radionuclides and their deposition onto plant surfaces, T_{eff} could be calculated by observing concentration decrease with time in plants. For the case of radiocesium, it is reported that the decreasing trend with time in an individual plant or plant population fits well with a combination of two exponential curves as follows when observation time was longer than 1 year (Antonopoulos-Domis et al. 1996).

$$C_t = A\ \exp\left(-\lambda_a t\right) + B\ \exp\left(-\lambda_b t\right) \tag{1}$$

where C_t is the concentration of ^{137}Cs in plant at time t (d), A and B are constants for short and long components, respectively, and λ_a and λ_b are loss rates in a population for short and long components, respectively. Practically, however, a set of T_{eff}s was found when 6–8 years observation results were obtained after the radiocesium release to the atmosphere (Antonopoulos-Domis et al. 1996; Unlü et al. 1995). Indeed, in a short time period, ca. 3–4 years after the atmospheric releases, only the short components have been reported (Antonopoulos-Domis et al. 1991; Tagami and Uchida 2015b). Thus the equation in a short time period is simply used as follows.

$$C_t = A\ \exp\left(-\lambda_a t\right) \tag{2}$$

Thus, T_{eff} is calculated using the following equation:

$$T_{eff} = \ln\left(2\right) / \lambda_a \tag{3}$$

Thus, from fitting of the observation results of radiocesium concentrations in a plant or plant population with time using Eq. 2, effective half-lives (T_{eff}) are firstly calculated using Eq. 2. Relations of T_{eff}, T_{phy} and T_{env} are expressed by the following equation.

$$1/T_{eff} = 1/T_{phy} + 1/T_{env} \tag{4}$$

In this report, T_{eff} are reported using data ^{137}Cs concentrations in several plant species, especially focused on data for several terrestrial plants, i.e., woody and herbaceous plants, observed after Fukushima nuclear accident comparing the data observed after the Chernobyl nuclear accident.

3 Herbaceous Plants

3.1 Very Short-Term Decreasing Rate of Radiocesium in Herbaceous Plants

When radiocesium was released to the atmosphere, direct contamination of aerial part of plant surface should occur, that is, a part of radiocesium deposited on the ground partially intercepted (Carini et al. 2003; IAEA 2010). After the contamination, radiocesium concentration in plants should decrease due to the following two major effects as mentioned above, that is, Effect-1 (weathering) and Effect-2 (dilution). The latter effect is limited when the plant was in mature stage and no additional growing was expected. For the case of the FDNPP accident, heavy radioactive deposition occurred during March, 2011; at that time, most herbaceous plants in the wild was not germinated yet, but only perennial herbaceous plants which have leaves overwinter directly contaminated.

In agricultural fields, winter crops such as leafy vegetables and root vegetables as well as young shoots of wheat were planted in March in Japan, 2011. In order to avoid ingestion of radioactive materials, food monitoring was carried out and the data was applicable to know the radiocesium decreasing rates after the accident. Because the purpose of food monitoring to eliminate foods exceeding standard limit, thus crops measured were suitable size to harvest for markets. For leafy vegetables, such as spinach and lettuce, 2–3 months is necessary to harvest after sowing. Within 60 days after March 11, 2011, therefore, vegetable samples obtained for food monitoring were directly contaminated sometime between at their young to mature stages. Thus the decreasing rates of annual herbaceous plants mainly affected by weathering and mass increase could be calculated. This stage is, however, usually separately reported and not included in the Eq. 1; however, because the information is important, the data are summarized below.

Figure 1 shows, ^{137}Cs concentration change with time in leafy vegetables collected in Fukushima Prefecture and prefectures next to Fukushima Prefectures; the map is shown in Fig. 2. It should be noted that there were two types of reported radioactivity values in March and April 2011: total radiocesium (^{134}Cs + ^{137}Cs) or separately determined values (^{134}Cs and ^{137}Cs). If only a total radiocesium concentration was reported, then ^{137}Cs was calculated by subtracting the ^{134}Cs contribution assuming that the ^{134}Cs:^{137}Cs activity ratio was 1:1 on 11 March 2011. From the results, the ^{137}Cs concentration decreases were fitted well using Eq. 2 with p value of <0.001 by *t*-test in both areas. T_{eff} in both areas were calculated and the range was

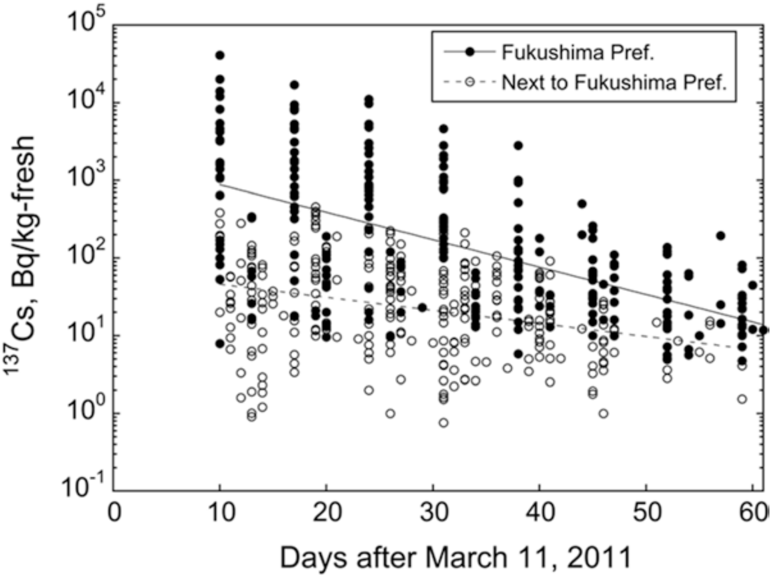

Fig. 1 ^{137}Cs concentration change in leafy vegetables collected in Fukushima Prefecture and prefectures next to Fukushima observed within 60 d after March 11, 2011 (Data adopted from MHLW 2016)

Fig. 2 Map of data collection sites in Japan for Fig. 1

from 8.5 to 17.9 days. For the case of Chernobyl accident, Mück (1997) reported initial T_{eff} values observed after the accident, that is, 4.2 days in lettuce and spinach, and 10.5 days in grass. From these results, it is concluded that weathering and mass increase were the important processes to fast decrease of radiocesium in herbaceous plants. On the other hand, the latter effect could not be found when the plant was in mature stage and no further weight increase was expected.

3.2 Short-Term T_{eff} of ^{137}Cs in Herbaceous Plants

After the contaminated plants were removed from the soil or dead on the soil, then the soil can be the major contamination source for the plants grown on the ground; from this stage, Eq. 1 was applied. For annual plants which were not germinated when the severe depositions occurred after the FDNPP, root uptake was the major radiocesium transfer pathway. It should be noted that only a small portion of total radiocesium in the soil was bioavailable, thus its transfer to plants through roots was generally small compared to the same group elements, e.g., K and Rb (IAEA 2010). Moreover, the bioavailable fraction of radiocesium generally decreases with time after its addition to soil (Cline 1981; Noordijk et al. 1992; Rosén et al. 1996; Tagami and Uchida 1996). Herbaceous plants, especially annual ones are sensitive to this factor; and when only a certain tissue of the plant was collected each year, e.g., cereal grains, beans, etc., then time dependence of the decreasing trend would be found. Unfortunately, however, not many continuous measurement results were found after the FDNPP accident.

Tagami and Uchida (2015b) measured ^{137}Cs in giant butterbur (*Petasites japonicas* (Siebold et Zucc.) Maxim.) and field horsetail (*Equisetum arvense* L.) from 2012 to 2014 grown in wild. Although these two species are perennial herbaceous plants, their above ground parts died every winter so that they can be indicators of radiocesium bioavailability in soil like annual plants. The T_{eff}s observed were ca. 450 (1.2 years) and 360 days (0.99 years), respectively. During this period, total ^{137}Cs concentration in soil and also vertical distribution did not change; thus probably because of the aging effect, bioavailable radiocesium decreased at these T_{eff} values. Smith et al. (1999) reported similar values for grasses with an average T_{eff} value of 1.5 years in UK, probably because grasslands were usually not ploughed so that the measurement conditions were similar to what was observed for wild plants in Japan. For the case of agricultural field, T_{eff} expected to be longer because of soil mixing accelerates the aging effect. Indeed, ^{137}Cs data in soybean (*Glycine max*) and buckwheat (*Fagopyrum esculentum*) grains collected in Fukushima Prefecture in 2012–2014 (detected data only from food monitoring data collated by MHLW 2016) showed longer T_{eff} as shown in Fig. 3. The calculated T_{eff}s were 858 (2.35 years) for soybean and 846 days (2.32 years) for buckwheat. Similar results were reported by Mück (1997) after the Chernobyl accident and the T_{eff} values were 1.4–2.7 years for vegetables and 3.0–3.4 years for cereals in Austria. This soil management difference was also be found in Russia in 1987–1989; T_{eff} for grains, potato and root crops ranged 1.2–2.9 while hay were slightly shorter, 0.9–2.3 (Fesenko et al. 1995).

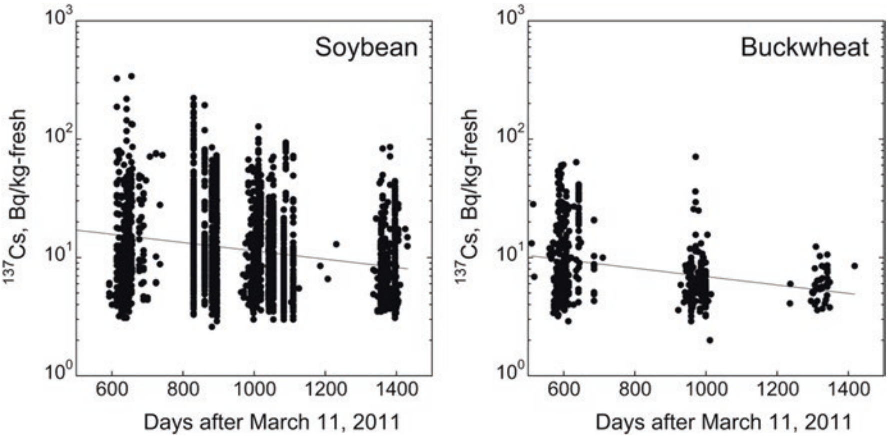

Fig. 3 Short-term ^{137}Cs concentration change with time in soybean and buckwheat collected in Fukushima Prefecture (Data adopted from MHLW 2016)

3.3 Long-Term T_{eff} of ^{137}Cs in Herbaceous Plants

For a longer period of time, leaching to deeper layer and translocation of radiocesium with soil particles need to be considered. Komamura et al. (2001) estimated the contribution of direct contamination from global fallout to the total ^{137}Cs concentration in polished rice grains and found after 1985, the contribution was less than 5 %, thus root uptake pathway was the major contributor after 1985. It should be noted that the effect from the Chernobyl accident was negligible for rice because its planting to the rice paddy field in Japan starts from early May to early June, that is, plant was small or not planted yet so that direct contamination was not important. However, wheat grains were affected because their harvest season is generally around the middle of May to June.

Using the rice grain data reported by Komamura et al. (2005) and additional data from the same group published by Ministry of Education, Culture, Sports, Science and Technology (MEXT 2016), Japan, T_{eff} affected by soil (Effect-4) was calculated. Fifteen sampling sites throughout Japan were used for ^{137}Cs measurements in polished rice grains from 1985 to 2005. For this case, long fraction, λ_b in Eq. 1, was calculated. The results are shown in Fig. 4 and the λ_b was calculated to be 0.0692 year^{-1}; the T_{eff} was 10 years, interestingly, the value was similar to herbaceous plants ($T_{eff}=13.4$ years on average) collected in Savanna River site in USA from 1974 to 2011 (Paller et al. 2014).

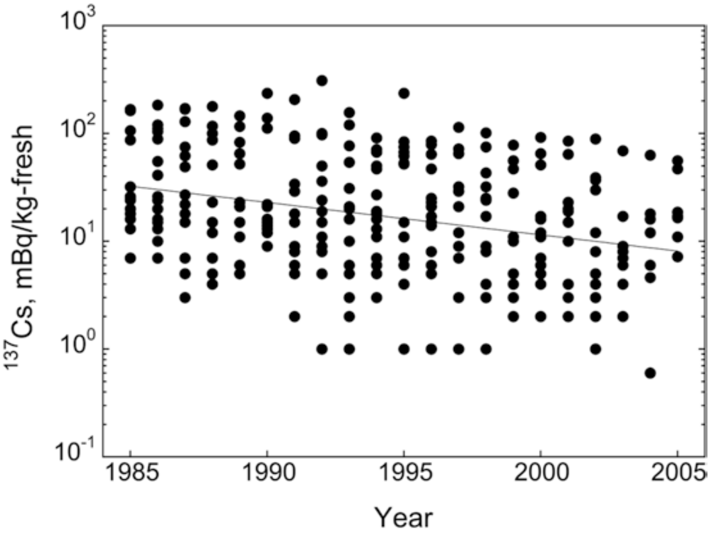

Fig. 4 Long-term ^{137}Cs concentration change in polished rice collected in Japan (Data adopted from Komamura et al. 2005)

4 Woody Plants

4.1 Short-term T_{eff} of ^{137}Cs in Fruit and Tea Plants

Trees are perennial woody plants; when the trees were directly contaminated with radiocesium, the concentration should rapidly decrease with time in a very short period of time as it was found for herbaceous plants, though the data were rarely found. After a certain period of time, radiocesium retained on the above ground parts would be gradually taken up by trees. Evergreen type trees are potentially most effective to take up radiocesium all the year around because leaf is the most important part to take up radiocesium among aerial parts of trees. Topçuoglu et al. (1997) reported that ^{137}Cs from the Chernobyl nuclear accident was stored into the stem of tea trees from old leaves and then translocated to new leaves. Deciduous trees are also possible to take up radiocesium from its aerial parts. Tagami et al. (2012) found radiocesium concentration in new shoots of deciduous trees were between those of evergreen tree and herbaceous plants. There were no emerged leaves of deciduous trees in March 2011 when heavy deposition was observed because it was before the growing season start for the case of the FDNPP accident. Due to the uptake of radiocesium by aboveground parts of trees, radiocesium concentrations in tree tissues in 2011 were highest and then, the concentrations have been decreasing (Kusaba et al. 2015a, b; Sato et al. 2015; Tagami and Uchida 2015c).

After the Chernobyl accident, effective half-lives were reported for trees. Pröhl et al. (2006) summarized T_{env}s of ^{137}Cs in terrestrial environment. T_{env} observed in pipfruit collected in Germany in 1965–1985 was 5.4 years and that in 1988–1999 was 6.3 years. Thus longer period of time, the T_{env} would be around his range. However, shortly after fallout, ca. within 2–4 years, T_{eff}s were usually shorter than those values. T_{eff} values observed after the Chernobyl accident were reported for fruit trees (Antonopoulos-Domis et al. 1991; Mück 1997) and tea (Unlü et al. 1995); the results are summarized in Table 1. Data obtained after the FDNPP accidents were also listed for comparison (Tagami and Uchida 2015c; Hirono and Nonaka 2016) together with calculated values from literature data (Kusaba et al. 2015a, b; MHLW 2016). In order to calculate T_{eff} from MHLW data, the same criteria reported in Tagami and Uchida (2015c) were used. In brief, the criteria were set that although distribution of radioactivity concentration in a local government area was not uniform, the amount of ^{137}Cs deposited to the land surface was assumed to be the same, and that more than three data per year should be reported in 2011–2013 by the local government. The result in Table 1 shows that the effective half-lives in fruit and tea trees were similar after the Chernobyl and Fukushima accidents. All T_{eff} values ranged from 0.34 to 1.64 years and these values log-normally distributed; thus geometric mean was calculated and the value was 0.86 years.

4.2 Short-Term T_{eff} of ^{137}Cs in Japanese Trees Obtained After the Chernobyl and Fukushima Accidents

However, unfortunately, these observations were carried out in different areas which made difficult to compare the T_{eff} values obtained after the Chernobyl and Fukushima accidents, because recent publication mentioned that T_{eff} increased with increasing annual precipitation (Rulík et al. 2014). For statistical analysis, it is necessary to obtain T_{eff} data from the same locations. In the outside of Japan, the effect of Fukushima was small; thus data obtained in Japan were surveyed using environmental radioactivity concentration database compiled by Nuclear Regulation Authority (2016). The database covers wide range of environmental samples, such as agricultural crops, milk, etc.; among them, indicator plants were selected for this analysis. An example is shown in Fig. 5 for ^{137}Cs concentration data in pine needles from 1984 to 2014 collected at one sampling site in Miyagi Prefecture. Using the data 1986–1988 and 2011–2013, T_{eff} in the same sampling site can be compared.

After the data survey, ^{137}Cs data in 2 years old leaves of Japanese black pine (*Pinus thunbergii*) collected at eight sampling sites were identified to be used this analysis. T_{eff} obtained are listed in Table 2. There were no correlations between the T_{eff} between observed after the Chernobyl and Fukushima accident by ANOVA test, and the geometric mean values of T_{eff} were 0.38 and 0.40 years, respectively.

In order to increase the number of T_{eff} values, monitoring sites were selected if the site had a series of ^{137}Cs data in plants in 1986–1988 or 2011–2013. Finally, T_{eff} data for Japanese black pine, tea (*Camellia sinensis*), citrus (*Citrus unshiu*), and Japanese cedar (*Cryptomeria japonica*) were obtained and the summary of the

Table 1 Reported T_{eff} of radiocesium in trees shortly after release

Plant name	Observation period	T_{eff} (in years)	References
Chernobyl			
Apple	1987–1990	0.86	Antonopoulos-Domis et al. (1991)
Apple	1987–1993	1.39	Mück (1997)
Apricot	1987–1990	0.77, 0.84	Antonopoulos-Domis et al. (1991)
Olive	1987–1990	0.83	Antonopoulos-Domis et al. (1991)
Peach	1987–1990	0.84	Antonopoulos-Domis et al. (1991)
Pear	1987–1990	0.69, 0.65	Antonopoulos-Domis et al. (1991)
Pear	1987–1993	1.23	Mück (1997)
Sweet cherry	1987–1990	0.66, 0.68	Antonopoulos-Domis et al. (1991)
Tea	1986–1988	0.34	Unlü et al. (1995)
Fukushima			
Apple	2011–2012	0.61	Renaud and Gonze (2014)
Apple	2011–2013	0.84, 0.96	This study (Data from MHLW 2016)
Blueberry	2011–2013	0.49	This study (Data from Kusaba et al. 2015b)
Chestnut	2011–2013	0.67	This study (Data from Kusaba et al. 2015a)
Chestnut	2011–2013	0.87	This study (Data from MHLW 2016)
Grape	2011–2012	0.57	Renaud and Gonze (2014)
Japanese apricot	2011–2012 2012–2013	0.47 0.76	Renaud and Gonze (2014)
Japanese apricot	2011–2013	0.50, 0.51, 0.54, 0.62, 0.73, 0.83	This study (Data from MHLW 2016)
Peach	2011–2012	0.51	Renaud and Gonze (2014)
Peach	2011–2013	0.63, 0.64, 0.68	This study (Data from MHLW 2016)
Pear	2011–2012	0.72	Renaud and Gonze (2014)
Persimmon	2011–2013	0.63, 0.83, 1.02, 1.13, 1.14, 1.20, 1.30	Tagami and Uchida (2015c)
Sweet cherry	2011–2012	0.52	Renaud and Gonze (2014)
	2012–2013	1.64	
Tea	2011–2013	0.36	Hirono and Nonaka (2016)
Yuzu	2011–2013	0.70, 0.84, 1.30	This study (Data from MHLW 2016)

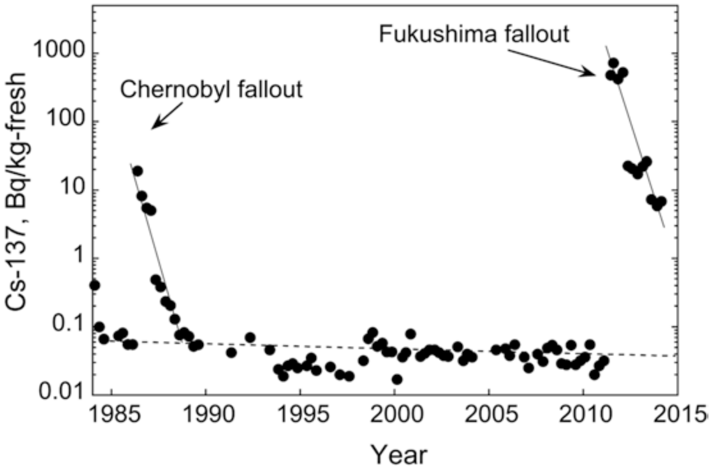

Fig. 5 ^{137}Cs concentrations in Japanese black pine needles collected in Miyagi Prefecture

Table 2 Comparison of T_{eff} of ^{137}Cs in pine leaves originated from the Chernobyl and Fukushima accidents

	After the Chernobyl accident (Observation period: 1986–1988)			After the Fukushima accident (Observation period: 2011–2013)		
Prefecture	Number of ^{137}Cs data	λ (year^{-1})	T_{eff} (years)	Number of ^{137}Cs data	λ (year^{-1})	T_{eff} (years)
Miyagi, A	11	2.34	0.30	11	2.02	0.34
Miyagi, B	22	2.00	0.35	12	1.85	0.37
Fukushima, A	8	1.32	0.52	9	2.74	0.25
Fukushima, B	9	1.22	0.57	11	1.89	0.37
Niigata, A	6	3.20	0.22	6	1.50	0.46
Niigata, B	11	1.98	0.35	11	1.25	0.55
Kyoto	3	1.45	0.48	3	0.98	0.71
Saga	11	1.74	0.40	7	2.15	0.32
Geometric mean		1.82	0.38		1.72	0.40

results are shown in Table 3. Significant difference was not observed among the data observed after the Chernobyl and Fukushima accidents by ANOVA test except for Japanese black pine ($p=0.023$, with logarithm data of T_{eff} values), although the geometric mean values differed only 0.07 years.

Table 3 Calculated T_{eff} of ^{137}Cs in leaves of Japanese black pine, Japanese cedar and tea trees, and fruit of *Citrus unshiu* collected in Japan

	After the Chernobyl accident (Observation period: 1986–1988)			After the Fukushima accident (Observation period: 2011–2013)		
Plant name	Number of T_{eff} data	λ (year^{-1})	T_{eff} (years)	Number of T_{eff} data	λ (year^{-1})	T_{eff} (years)
Japanese black pine	27	1.53	0.45 (0.22–0.79)	17	1.84	0.38 (0.25–0.71)
Tea	12	1.65	0.42 (0.26–0.76)	10	1.67	0.41 (0.33–0.68)
Citrus unshiu	8	1.28	0.54 (0.39–0.69)	2	1.25	0.54 (0.54–0.56)
Japanese cedar	3	1.38	0.50 (0.45–0.62)	1	1.90	0.36

5 Conclusions

Effective half-lives, T_{eff}, of radiocesium in herbaceous and woody plants were summarized in this paper comparing data obtained after the Chernobyl and FDNPP accidents, because T_{eff} is important to know how fast radiocesium removed from plants. T_{eff}s observed after these two accidents within 2–4 years for herbaceous plants as well as trees were similar. The T_{eff} data obtained after the Chernobyl accident were mostly obtained in temperate regions, that is, Germany, Austria, Greece, etc. The T_{eff} data obtained in Japan after the FDNPP accident were also temperate areas; under similar climate conditions, growing rates of plants would be the same, which caused the same range T_{eff} data. For a longer time period, however, radiocesium decline rates might change among different areas due to different precipitation rates and temperatures. It is also important to measure seasonal change to know main factor to eliminate radiocesium from plants. Thus, continuous measurement of radiocesium in plants is necessary for a long-time period after nuclear accident(s).

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