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Comparisons of effective half-lives of radiocesium in Japanese tea plants after two nuclear accidents, Chernobyl and Fukushima

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ABSTRACT

The time-dependence of ^{137}Cs in new shoots of tea (*Camellia sinensis* (L.) Kuntze) following a ^{137}Cs-deposition was analyzed and quantified in terms of effective half-lives. The underlying monitoring studies were performed after the accidents in the Chernobyl and the Fukushima Daiichi nuclear power plants for tea plants growing in Japan. The major transfer route for atmospherically deposited radiocaesium to the first new shoots sampled after the accidents were different: for the Fukushima accident, it was mainly translocation of radiocaesium deposited onto old leaves and twigs to the new growth, while direct deposition on the new leaves was the major source after the Chernobyl accident. The effective half-lives in new tea leaves representing the fast and slow components of the decline did not significantly differ between these accidents. Geometric means (ranges) of fast and slow effective half-lives of ^{137}Cs after the Chernobyl accident were 66 d (25–125 d) and 902 d (342–15900 d), respectively, and those after the Fukushima accident were 50 d (26–105 d) and 416 d (222–1540 d), respectively. From these results, ^{137}Cs declines in new tea leaves were similar although contamination conditions were different for these two accidents.

1. Introduction

Green tea, produced from tea plants (*Camellia sinensis* (L.) Kuntze), is considered to have positive health effects, including even the possible reduction of risk for some types of cancers (Boehm et al., 2009; Zhou et al., 2016; Xing et al., 2019). Thus, its consumption and production are increasing worldwide (FAO, 2015). Japan is one of the biggest green tea production countries; according to the most recent report by the Ministry of Agriculture, Forestry and Fisheries (MAFF), 80,000–86,000 tons are harvested annually in 2009–2018 (MAFF, 2019a). A new shoot with 2–3 young new leaves is generally harvested because it is the best size to produce green tea drinks; therefore, commercial products identified as "green tea leaves" in the market includes both new leaves and stems. Hereafter, commercial product "green tea leaves" and new shoots harvested for monitoring (non-commercial use) are termed as "new shoots of tea plants" because these are the same tissue part of tea plants.

As a consequence of the 2011 accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP), green tea production in Japan was partly affected. Direct deposition of a small portion of radionuclides to new shoots occurred in 2011 because new shoots started to grow from the middle of April, and the atmospheric deposition was almost finished by that time. An indirect effect, that is, translocation from above ground parts of tea plants to new shoots, was considered to be the major contamination pathway (Tagami et al., 2012a; Ikka et al., 2018). Export of green tea products decreased due to residual (direct and indirect) radioactivity that was observed in green tea plants in some affected areas in 2011. However, as the main green tea production areas, i.e. the top five prefectures (Shizuoka, Kagoshima, Mie, Miyazaki and Kyoto) are located more than 300 km away from the FDNPP, the measured activity levels in tea leaf products were lower than the limits at that time (500 Bq kg^{-1} dry weight for total radiocaesium ($^{134+137}$Cs) in new shoots of tea plant). These five prefectures produced more than 80% of the total green tea production in Japan in 2010 (MAFF, 2019b). Some green tea is also produced in Ibaraki and Saitama Prefectures, but these prefectures contribute only about 1% to Japan's production of green tea. For Fukushima Prefecture, due to climatic reasons, green tea production is negligible (MAFF, 2019c). It was recognized that the tea production industries were therefore not significantly affected by the nuclear accident. Some tea products with total radiocaesium activities exceeding 500 Bq kg^{-1} were exported and when this was found out, they were

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withdrawn from the market; this occurred for products from some limited areas in 2011. From 2012, the export of green tea has continuously increased (MAFF, 2019a). Nevertheless, for ensuring food safety, green tea is regularly monitored in Japan for residual levels of radiocaesium.

In the previous large radioactive contamination event, the Chernobyl nuclear power plant accident of late April 1986, low levels of radiocaesium in agricultural products including tea plants were detected just after the accident (as compiled by the Nuclear Regulation Authority [NRA], Japan, 2019). Among agricultural products, ^{137}Cs concentrations in tea shoots have the highest possibilities to be affected by direct deposition to the new growth tissues because tea is an evergreen tree. Unfortunately, however, no study was carried out on ^{137}Cs fate in tea plants in Japan at that time. Internationally, results for tea plants were reported in Turkey (Unlü et al., 1995; Topcuoğlu et al., 1997; Zehringer et al., 2018), and the maximum radiocaesium ($^{134+137}$Cs) concentration was ca. 30000 Bq kg^{-1} dry. Unlü et al. (1995) reported that the ^{137}Cs decrease in tea plant leaves might be described by a sum of two exponential functions. From the compilation of the measurements in new shoots of Japanese green tea, we previously applied single exponential function fittings to describe the ^{137}Cs-activity in tea shoots within 3 y after the Chernobyl and FDNPP accidents (Tagami, 2017); however, further analysis in longer time period has not been carried out yet.

Following the deposition of radiocaesium onto tea plant leaves after the Chernobyl and FDNPP accidents, the activity levels were found to decline due to mainly weathering effects and growth dilution. Radiocaesium on the leaves and branches was partially absorbed into the plant body (Unlü et al., 1995; IAEA, 2010; Hirono and Nonaka, 2016; Tagami, 2017). The highest loss rates from a plant surface were observed immediately after the deposition by the FDNPP accident (Hirono and Nonaka, 2016). After the Chernobyl accident, it was observed that radiocaesium was distributed in the whole plant and radiocaesium activities in tea shoots persisted over many years (Unlü et al., 1995; Topcuoğlu et al., 1997). The same phenomenon would be expected after the FDNPP accident. Additionally, radiocaesium can be taken up from soil through the plant's roots. We found for rice paddy fields that radiocaesium mobility in soil was slightly higher in the first year after the accident (Uchida and Tagami, 2018; Tagami et al., 2018), thus the nuclide could be taken up by roots. However, the root uptake process was not the major source for radiocaesium in tea plant because root uptake contributed only 1/8 of the total ^{137}Cs concentration in new shoots in the early summer harvested in 2014 (Ibaraki prefecture, 2014). The translocation of radiocaesium from above ground tissues/storage tissues to new shoots would still be the major pathway.

Following deposition on the tea plant canopy, the activity levels are highest in the first year, then they are one to two orders of magnitude lower in the following years. Estimation of the decreasing rate is important for any management of perennial crops such as fruit trees and tea plants; the rate of decline provides information on when – following deposition of radionuclides – the activity levels in harvested products will be lower than the allowed activity limits of radiocaesium for foods. In Japan, these limits are 100 Bq kg^{-1} of radiocaesium in foods or 10 Bq kg^{-1} in green tea drinks (Ministry of Health, Labour and Welfare, 2012).

In this study, to estimate the decline of radiocaesium in green tea following deposition of radionuclides for a longer time period, we analyzed monitoring data for green tea and derived effective half-lives of ^{137}Cs in green tea following deposition of ^{137}Cs. The values were compared against data obtained after the 1986 Chernobyl accident. For comparison, we measured radiocaesium decline in new leaves of an azalea tree (*Rhododendron* L.) after the FDNPP accident, because that species and tea are in the same order (*Ericales*) of evergreen plants, and they are cultivated to the similar size in Japan. Besides, azalea is usually easily obtained because it is widely used as a garden tree; thus, the species might be useful as a model tree for tea plant.

2. Materials and methods

2.1. Data survey

We carried out a literature survey including peer reviewed journals in English and Japanese languages and reports of Japanese government institutes on environmental radioactivity in Japan following the FDNPP accident. If new shoots of tea plant were sampled several times from one collection site, then the series of data is considered as one data set. Since the ^{137}Cs decrease in the year of the accident is the fastest, data sets collected in 2011 (no additional data after 2012) were included in the analysis. Data continuously sampled at least three to four years from 2011 or 2012 up to 2017 and samplings carried out one or two times a year were also included as data sets. Additionally, data sets were obtained from figures which were published by Japanese government institutes if data were continuously available from 2011 or 2012 for at least three to four years.

Another data source for ^{137}Cs in green tea is food monitoring results. Environmental monitoring programs in Japan near nuclear power plants were started in 1970 according to the archived database of the NRA. The ^{137}Cs-activity levels in tea shoots from 1970 to 2017 are shown in Fig. 1 for harvesting periods in one of the sample collection sites of Shizuoka Prefecture. Deposition from the Chernobyl accident was also detected in green tea in Japan; thus, those data were included in the analysis for effective half-life. Since the same monitoring programs were continued after the FDNPP accident, the data sets were also used to derive effective half-lives after the FDNPP accident. It is noteworthy to see that for observations in the Shizuoka Prefecture site (Fig. 1), the peak level after the Chernobyl accident is similar to the peak level after the FDNPP accident. This sampling site in Shizuoka Prefecture is about 300 km southwest of the FDNPP, and the ^{137}Cs deposition during the FDNPP accident was ca. 3200 Bq m^{-2}, while ca. 200 Bq m^{-2} was observed at the time of Chernobyl accident. Several sampling sites in Shizuoka Prefecture were set for environmental radioactivity monitoring and thus their data sets were separately collated.

2.2. Sampling of azalea leaves

The sample collection site for azalea leaves was in Chiba Prefecture, near the National Institutes for Quantum and Radiological Science and Technology (QST), located about 210 km south from the FDNPP. New and old leaves were collected at several times a year from 2011 to 2016. Immediately after collection, leaf samples were transferred to a laboratory and their fresh weight was obtained, before washing them with

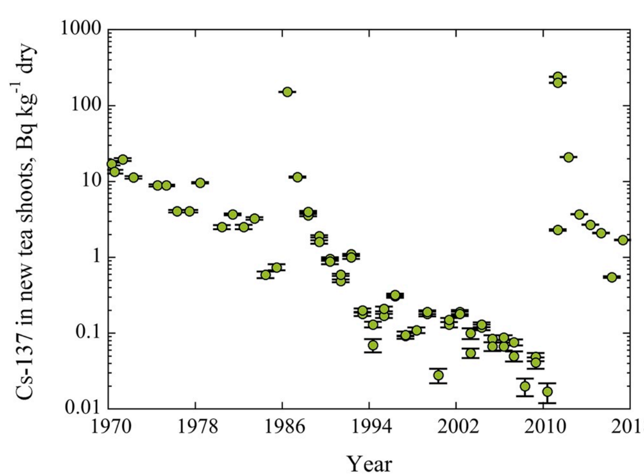

Fig. 1. ^{137}Cs concentrations in new shoots of tea plants in 1970–2017 at tea harvesting periods in one of the observation sites of Shizuoka Prefecture. Bars show standard errors reported in the open source database (NRA, 2019).

tap water to remove dust from the surface; this was done in a washing bowl by changing the water 5 times, and then, finally, the samples were rinsed with reverse osmosis water. All samples were oven-dried to a constant weight at 80 °C in an electric oven for at least 2 d to decrease the sample volume. Then the sample was pulverized and mixed well, before being transferred to a plastic container (100 mL).

Radioactivity concentration in each sample was measured by HPGe detecting system (Seiko EG&G). using 50,000–200,000 s counting interval; the counting time settings depended on the radiocaesium concentration in the samples. The detector was the GEM coaxial type (relative efficiency: 30%). A mixed gamma standard source (MX0333U8, Japan Radioisotope Association) was used for an efficiency correction and two reference standard materials IAEA-156 and 373 were used for an accuracy check. The ^{137}Cs activity was decay corrected to the sampling date, and reported on the dry weight basis.

2.3. Analysis

Often, in a short time after the contamination, the decrease of activity levels in a plant or in an environmental compartment can be described as an exponential function with an effective half-life (T_{eff}). For long observation periods, the decline often consists of several phases. Therefore, sometimes two or even more exponential functions are needed for an appropriate fit of the data. Usually, immediately after the contamination, a component representing a fast loss can be observed, which is followed by a slow loss component.

From Fig. 1 together with data sets from different sampling sites (Supplementary Material Table S1), we found the time-dependence of ^{137}Cs in tea plants from 2011 to 2017 (7 y). Fortunately, for the period immediately after the Chernobyl accident data for seven years from 1986 to 1992 were available. To compare the time-dependence of ^{137}Cs in tea plants after these two accidents, we calculated fast and slow components in the plants according to the sum of two exponential functions which fits appropriately to the ^{137}Cs decreasing trend with time from the year of the accident to the following several or more years:

$$Y(t) = Ae^{-\lambda a\ t} + Be^{-\lambda b\ t} \ldots\ldots\ldots\ldots\ldots\ldots \quad (1)$$

where Y(t) is the activity concentration of ^{137}Cs (Bq kg^{-1} dry or Bq kg^{-1} fresh) in new shoots of tea plants at time t (days after the accident occurred), A and B are constants for fast and slow components, and λ_a and λ_b are loss rates for fast and slow components, respectively. If the activity concentration data were given in fresh mass (Bq kg^{-1} fresh), then we did not convert the value into dry weight basis, but applied the raw data as reported.

The effective half-lives for fast (a short half-life from tea plants, T_{eff_a}, day) and slow (a long half-life from tea plants, T_{eff_b}, day) components are calculated according to:

$$T_{eff} = \ln(2)/\ \lambda \ldots\ldots\ldots\ldots\ldots\ldots \quad (2)$$

The contribution (in percent) of the slow component (C_{slow}) to the total Cs in new shoots of tea plants at the time the accident started (April 26, 1986 for Chernobyl or March 11, 2011 for FDNPP) is calculated according to:

$$C_{slow} = B/\ (A + B) \times 100 \ldots\ldots\ldots\ldots\ldots\ldots \quad (3)$$

For this approximation, the Kaleida Graph software (Synergy Software, version 4.5.3) was used. This software uses the Levenberg-Marquardt algorithm for fitting.

3. Results and discussion

The analysis of the time-dependence of ^{137}Cs in new shoots of green tea plants after the FDNPP accident was done by including five papers in Japanese (Shiraki et al., 2012, 2014; Hirono and Nonaka, 2015; Honda and Miyazaki, 2016; Sekiyama et al., 2016), one paper in English (Hirono and Nonaka, 2016), and five data sets from NRA (2019). For data after the Chernobyl accident, one data set from Turkey (Unlü et al., 1995) and 8 data sets from NRA (2019) were analyzed. All data sets are provided in Supplementary Material Tables S1 and S2. The prefectures where data were measured are shown in Fig. 2. These tea-growing prefectures outside 400 km were only affected to a minor extent by the releases from the FDNPP. Some tea products in 2011 harvested in Chiba, Saitama and Kanagawa Prefectures exceeded 500 Bq kg^{-1} but the concentration decreased rapidly in 2012.

3.1. Effective half-life (T_{eff}) values of ^{137}Cs in new shoots of tea plants after the Chernobyl accident

Information on tea plant management (harvesting and trimming) in Japan (Agronomy Section of the Annual Scientific Conference of Tea, 1977; Kamata et al., 2015) is summarized in Table 1. In the Chernobyl accident, direct deposition occurred in early May 1986 in Japan and thus new shoots were directly contaminated with radionuclides, while in the FDNPP accident, radionuclides deposited on tea plants were partially removed due to spring trimming. Thus, most of the radionuclides that remained on the plants were absorbed in or fixed on old leaves, twigs, and stems. Only a small portion was in the new shoots (for the first harvest) and these radionuclides had translocated from other tissue parts directly contaminated by the FDNPP fallout. Consequently, the contamination conditions for the new shoots of tea plants were different for these two accidents; the direct (surface) contamination in the Chernobyl accident and indirect (internal) contamination in the FDNPP accident. As shown in Fig. 1, although different ^{137}Cs deposition amounts between these accidents were observed in a certain sampling site, the ^{137}Cs concentrations in new shoots of tea plants were similar.

To analyse T_{eff} values for the data sets observed after the Chernobyl accident, from 1986 to 1992, suitability of eq. (1) was considered using the correlation coefficient (R) between ^{137}Cs activity concentrations in new shoots of tea plants and time after the accident. Fig. 3 shows an example from the data sets; a single-exponential function fitting resulted in R = 0.82 and a two-exponential function fitting (eq. (1)) resulted in R = 0.99. Seven other data sets similarly showed better R values with eq. (1) than with single-exponential function fitting. We therefore concluded that the two-exponential function fitting was appropriate for long-term monitoring data sets including the first year after the direct deposition to tea plants.

The calculated values for fast and slow T_{eff} are shown in Table 2. The resulting values for fast and slow T_{eff} from all data sets were log-normally distributed, respectively, so the geometric mean (GM) value could be calculated together with geometric standard deviation (GSD). Arithmetic mean (AM) and standard deviation of T_{eff} values were also calculated. The GM value of slow component percentage was 4%. Thus, on average, more than 95% of deposited ^{137}Cs on new shoots of tea plants was the fast component and ^{137}Cs in this component decreased rapidly with T_{eff} of about 2 months. The GMs of the fast and slow components of the activity decline were 66 d and 900 d, respectively. Pröhl et al. (2006) reported that the ecological half-life observed after the Chernobyl accident in certain fruits would be more than 3 y when the year 1987 was excluded. The period of the ecological half-life was relevant to T_{eff_b} in this study; when we calculated ecological half-life considering physical half-life of 30.2 y for ^{137}Cs, the GM was 2.7 y, and that agreed reasonably well with the result by Pröhl et al. (2006). The agreement of these results for tea leaves and certain fruits implied that some tree species in a temperate climate would have a similar effective half-life of radiocaesium.

3.2. Effective half-life (T_{eff}) values of ^{137}Cs in tea and azalea leaves after the FDNPP accident

The literature survey resulted in: (A) four data sets collected in 2011 (Shiraki et al., 2012; Hirono and Nonaka, 2015), (B) ten data sets with

Fig. 2. Sampling sites in Japan: 1986–1992 data were from Shizuoka, Kyoto, Shimane and Kagoshima Prefectures, and 2011–2017 data were from Chiba, Saitama, Kanagawa, and Shizuoka Prefectures.

Table 1
Timings of tea plant management practices in Japan and radionuclide depositions from the Chernobyl and FDNPP accidents.

	Feb			Mar			Apr			May			Jun			Jul			Aug			Sep			Oct			Nov		
Treatment			S	S	S	S			1	1	1	1			2	2	2		3	3					A	A	A	A		
FDNPP					●	●	●																							
Chernobyl										●																				

S: Spring trimming, A: Autumn trimming, 1, 2 and 3: Green tea harvest seasons, 1, 2 and 3rd.
●: Radionuclide deposition timing

sampling started from 2011 to 2013, 2014 or 2017 (Shiraki et al., 2014; Hirono and Nonaka, 2016; Sekiyama et al., 2016; NRA, 2019) and (C) one data set collected in 2012–2014 (Honda and Miyazaki, 2016).

According to the Chernobyl fallout results, there is a day before which the fast component was the major part of ^{137}Cs content in new shoot of tea plants, and then the slow component became the major part. For example, the red point in Fig. 3 at day X is the day of the major component change. If samplings of a data set finished before that day, or samplings started after that day, two-exponential function decline would not be appropriate because the slow component or the fast component was missing, respectively.

^{137}Cs concentrations in both components are the same at day X; therefore, we obtain the following equation.

$$Ae^{-\lambda a\ X} = Be^{-\lambda b\ X} \text{.................} \quad (4)$$

From eq. (4), day X is calculated as follows.

$$X=(\ln A - \ln B)/(\lambda_a-\lambda_b) \text{.................} \quad (5)$$

For the data sets listed in Table 2, calculated day X ranged from 114 to 633 d (specific results not shown), and the data were distributed normally. Thus AM value of 371 d was calculated for day X; this result

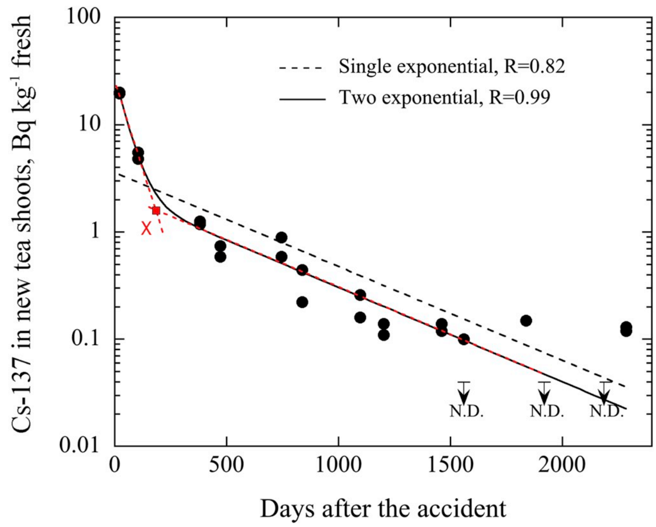

Fig. 3. Single-exponential function and two-exponential function fittings for ^{137}Cs concentrations in new shoots of tea plants observed in one of the observation sites of Shizuoka Prefecture after the Chernobyl accident. N.D. showed not detected. At day X (red point), the same amount of ^{137}Cs was contributed from fast and slow components. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

implied that it would not be appropriate to use eq. (1) for a sampling data set which was finished within 1 y (data sets A) or a data set with sampling started from the next year after the accident (data set C). Thus single-exponential function fittings were used for data sets A and C and two-exponential function fittings were used for data sets B.

The T_{eff} values calculated for the data collected after the FDNPP accident are summarized in Table 3 according to the conditions above. Shiraki et al. (2014) applied a single-exponential function using data from 2011 to 2013. However, in the present paper, we re-evaluated those data using a two-exponential function fit. Honda and Miyazaki (2016) reported data sampling that started in 2012, the second year after the accident; therefore, the fast component representing a fast T_{eff_a}, would not be identified in their results. Thus, a single-exponential function fitting was used which resulted in a high correlation coefficient of R = 0.98 for the slow component.

Similar to the data collected after Chernobyl accident, the T_{eff_a} and T_{eff_b} values from all data sets were log-normally distributed, so GM values were calculated, and they are given with AM and SD values in Table 3. The T_{eff} GM values of fast and slow components were 50 d and 416 d, respectively. The GM value of the slow component percentage (4.4%) was similar to that observed after the Chernobyl accident.

For comparison, ^{137}Cs concentration in new and old azalea leaves were plotted against time in Fig. 4, and the fitting results using equation (1) for fast and slow components are shown in Table 4. Except for T_{eff_a} value in new leaves of azalea, T_{eff_a} value in old leaves and T_{eff_b} values in new and old leaves were similar to the T_{eff_a} and T_{eff_b} values observed for tea leaves. T_{eff_a} in new azalea leaves was slightly longer than those for tea plants, however, the difference was only 1.5 times the maximum T_{eff_a} value of new shoots of tea plants. Thus, as expected from the similar characteristics of these tree species, ^{137}Cs concentrations in new leaves decreased in a similar manner. Radiocaesium concentration data from azalea would be a proxy of tea plants. About one year after the accident and thereafter, ^{137}Cs concentrations in new leaves were slightly higher than those in old leaves (Fig. 4). This result indicates that ^{137}Cs was actively concentrated in new leaves. As well, the longer T_{eff_b} for new leaves than for old leaves implied a change in the major transfer pathways from "internal transfer of radiocaesium to new leaves" to "soil-to-plant root uptake". Our previous study on herbaceous plants found long-half-lives, and the main radiocaesium transfer pathway was root uptake (Tagami and Uchida, 2017); this process should also be true for woody plants.

3.3. Comparisons of effective half-lives in tea plants after two nuclear accidents

Because the contamination pathways of new shoots of tea plants in the first year were different – i.e., direct contamination of the shoots in 1986, and contamination of the tea plants and translocation of radiocaesium to the shoots which had not emerged much at the time of deposition in 2011 (indirect contamination), comparison of T_{eff} data would be of interest. Logarithms of the T_{eff} data were used for this analysis and the results showed that the T_{eff} values of the fast and slow components for these two accidents did not show any significant difference in the analysis of variance (ANOVA) test.

We carried out a further analysis by separating fast component T_{eff} values (T_{eff_a}) using single-exponential function fittings (data sets A), and two-exponential function fittings for the FDNPP (data sets B) and the Chernobyl (data sets 1986–1992), the results are shown in Table 5. Although we expected T_{eff_a} would be faster using a single-exponential function because the decrease would be fast at an early stage of the accident, the ANOVA test did not show significant differences among these three groups after the T_{eff} data were changed to logarithms. The cases using the two-exponential function fitting described the fast component decrease fraction appropriately. The main reason is because

Table 2
Effective half-life (T_{eff}) values of fast and slow components for new shoots of tea plants observed in Japn after the Chernobyl accident.

Prefecture/City or Town	Sampling years	λ_a	T_{eff_a}, d	λ_b	T_{eff_b}, d	Slow component (C_{slow}), %	Ref.
(Turkey)[b]	1986–1992		125		1110	5.5	Unlü et al. (1995)
Shizuoka/Hamaoka	1986–1992	0.028	25	0.0017	420	4.7	This study, data from NRA, 2019
Shizuoka/Omaezaki	1986–1992	0.0064	109	0.0004	1940	2.6	This study, data from NRA, 2019[8]
Shizuoka/Sagara	1986–1992	0.020	35	0.0020	340	8.1	This study, data from NRA, 2019
Shizuoka/Shuzenji	1986–1992	0.010	70	0.0016	440	4.3	This study, data from NRA, 2019
Shizuoka/Iwata	1986–1992	0.0073	95	0.0020	360	8.3	This study, data from NRA, 2019
Kyoto/Kaya	1986–1992	0.0080	86	0.00004	15900	0.6	This study, data from NRA, 2019
Shimane/Kashima	1986–1992	0.0081	86	0.0017	410	4.9	This study, data from NRA, 2019
Kagoshima/Sendai	1986–1992	0.017	40	0.0005	1270	3.9	This study, data from NRA, 2019
	N		9		9	9	
	AM		75		2500	6.4	
	SD		35		5100	6.7	
	GM		66		900	4.0	
	GSD		1.8		3.5	2.2	
	Min		25		340	0.6	
	Max		125		15900	8.3	

N: Number of data; AM: Arithmetic mean; SD: Standard deviation; GM: Geometric mean; GSD: Geometric standard deviation. [a]Samples were collected in Turkey
[b] The samples were collected in Turkey.

Table 3
Calculated effective half-life (T_{eff}) values of fast and slow components for new tea shoots observed after the FDNPP accident.

Prefecture/City, town or village	Sampling year	λ_a	T_{eff_a}, d	λ_b	T_{eff_b}, d	Slow component (C_{slow}), %	Data source
Kanagawa/Odawara	2011	0.015	47				Shiraki et al. (2012)
Kanagawa/Minami-ashigara	2011	0.015	45				Shiraki et al. (2012)
Kanagawa/Kiyokawa	2011	0.015	46				Shiraki et al. (2012)
Kanagawa/Sagamihara	2011–2013	0.011	60	0.0015	460	28.1	Shiraki et al. (2014)
Shizuoka/Shimada	2011	0.020	35				Hirono and Nonaka (2015)
Shizuoka/Shimada	2011–2013	0.026	26	0.0030	230	5.3	Hirono and Nonaka (2016)
Shizuoka/Omaezaki (H)[a]	2011–2017	0.012	59	0.0024	290	13.0	NRA (2019)
Shizuoka/Omaezaki	2011–2017	0.0066	105	0.0011	640	4.3	NRA (2019)
Shizuoka/Makinohara[b]	2011–2017	0.0072	96	0.0014	500	4.9	NRA (2019)
Shizuoka/Izu[c]	2011–2017	0.0068	102	0.0005	1540	1.1	NRA (2019)
Shizuoka/Iwata	2011–2017	0.014	51	0.0017	400	4.2	NRA (2019)
Saitama/Iruma	2012–2014			0.0026	260		Honda and Miyazaki (2016)
Chiba/(Site-A)	2011–2014	0.027	26	0.0031	220	1.6	Sekiyama et al. (2016)
Chiba/(Site-C)	2011–2014	0.018	38	0.0010	700	1.5	Sekiyama et al. (2016)
Chiba/(Site-D)	2011–2014	0.017	41	0.0027	250	5.8	Sekiyama et al. (2016)
	N		14		11	10	
	AM		55		500	7.0	
	SD		27		380	8.2	
	GM		50		416	4.4	
	GSD		1.6		1.8	2.7	
	Min		26		220	1.1	
	Max		105		1540	28	

N: Number of data; AM: Arithmetic mean; SD: Standard deviation; GM: Geometric mean; GSD: Geometric standard deviation.
[a] Previously the town name was Hamaoka.
[b] Previously the town name was Sagara.
[c] Previously the town name was Shuzenji.

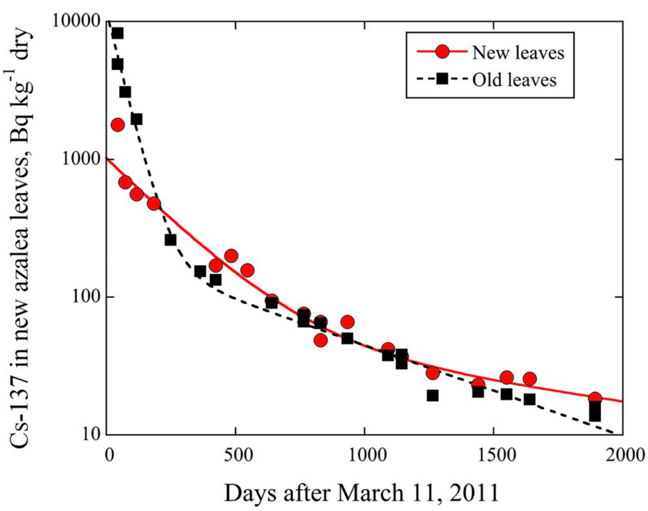

Fig. 4. Two-exponential function fitting result for ^{137}Cs concentrations in new and old leaves of azalea collected in Chiba Prefecture after the FDNPP accident.

Table 4
T_{eff} values of fast and slow components for new and old leaves of azalea collected in Chiba Prefecture observed after the FDNPP accident.

Tissue	λ_a	T_{eff_a}, d	λ_b	T_{eff_b}, d	Slow component, %
New leaves	0.0044	157	0.0006	1110	5.9
Old leaves	0.018	40	0.0015	460	1.7

percentages of ^{137}Cs in fast component were much higher than those in slow component.

Following both accidents, about 95% of the total radiocaesium in the new shoots of tea plants decreased with half-lives of 25–125 d and only ca. 5% was attributed to the slow component. The initial, fast component in new shoots of tea plants is related to direct contamination and/or fast translocation from old tissues of a tree; hence, the decrease in the activity levels should be due to weathering and growth dilution. This is relevant for both FDNPP and Chernobyl accidents; because they occurred in the spring, and thus similar fast T_{eff} values as well as fast components were observed. The long-term activity in green leaves is due to uptake of radiocaesium from soil, which is much lower than the uptake via the foliage (IAEA, 2010). The long-term decline is due to decreasing bioavailability of radiocaesium for root uptake, which is caused by the continuously increasing sorption of caesium to soil solid phase (Tagami and Uchida, 1996).

Table 5
T_{eff} values of the fast component calculated using single- and two exponential functions.

Year and type of fitting	N	GM, d	GSD	Min, d	Max, d
1986–1992, T_{eff_a}, Two-exponential	9	65.9	1.8	24.8	125
2011, T_{eff}, Single-exponential (data sets A)	4	41.0	1.3	29	47
2011–2013, 2011-14 or 2011–2017, T_{eff_a}, Two-exponential (data sets B)	10	53.6	1.7	26	105

N: Number of data; GM: Geometric mean; GSD: Geometric standard deviation.

Regarding green tea drinks, the direct or indirect contamination situation needs to be considered because the radiocaesium extractability from the new shoots of tea plants should be different. Direct contamination means radiocaesium attached to the surface of the new shoots while indirect contamination means internal contamination. Therefore, in the former case, more radiocaesium would be extracted than that in the latter case, if we were to adopt vegetable data (IAEA, 2010; Radioactive Waste Management Funding and Research Center, 2013). When a typical brewing method was applied, that is, a water to tea leaf product weight ratio of about 40:1, we previously found that radiocaesium extractability from commercially sold tea leaf produced in 2011 was ca. 50–70% with 90 °C water (Tagami et al., 2012b). Shiraki et al. (2013c) reported similar results using tea samples collected in 2012, and Cook et al. (2016) also found the similar extractability in Japanese tea products purchased in Canada. Unfortunately, no green tea extraction data were collected just after the Chernobyl accident and we could not compare the extractability between tea products obtained under different contamination conditions.

Radiocaesium ($^{134+137}$Cs) standard limit in green tea drinks in Japan is 10 Bq kg^{-1} (Ministry of Health, Labour and Welfare, 2012). It should be noted that types of tea leaf products, time of brewing, change of water:leaf ratio, and water temperature would affect the extractability (Tagami et al., 2012; Oya et al., 2015; Zehringer et al., 2018). However, considering the amounts of radiocaesium deposition in green tea production areas after the FDNPP accident, and T_{eff} values mentioned above, radiocaesium concentration in new shoots of tea plants would not increase. Therefore, green tea drinks would not exceed the standard limit in the future, even if 100% of the radiocaesium was extracted.

3.4. Relationship for radiocaesium between new shoots and deposition for tea plants

It would be informative to compare concentration ratios (CRs) of radiocaesium between new shoots at the time of first harvest (Bq kg^{-1} dry) and total deposited amounts (Bq m^{-2}) in 1986 and 2011, by which the effect of direct contamination would be elucidated. Using deposition data from the NRA (2019), we estimated deposition densities from Chernobyl and FDNPP accidents separately by summing deposition in May to June 1986 for the former and March to June 2011 for the latter at the nearest deposition sampling site for each tea plant T_{eff} measurement site. Using equation (1), we could calculate radiocaesium concentration in new shoots of tea plants for the first harvest. If the data available were for fresh weight, then the average value of dry/wet ratios, 0.263 (Shiraki et a., 2013a) was applied to convert the ^{137}Cs data to a dry-weight basis. The activities in new shoots of tea plants were calculated for May 10, 1986 for the Chernobyl accident (15 days after the accident occurred), and for May 10, 2011 for the FDNPP accident (60 days after the accident occurred).

Comparison between calculated concentration ratios observed after the Chernobyl and FDNPP accidents are shown in Fig. 5; the detailed data are available in Supporting Information Table S3. The ANOVA test results indicated that these data were statistically different ($p < 0.01$). The GMs of the CR values for Chernobyl and FDNPP fallouts were 0.60 (range: 0.26–1.44) m^2 kg^{-1} dry and 0.19 (range: 0.06–0.38) m^2 kg^{-1} dry, respectively. As expected, the direct deposition to the new shoots would affect the radiocaesium concentration. However, it should be noted that for the FDNPP accident, the translocation of caesium from directly contaminated tissues (old leaves, twigs and stems) as well as growth dilution affected concentrations of the new shoots.

The translocation of radiocaesium absorbed by old leaves and twigs would be the major source of radiocaesium in new tissues. However, this process did not continue for a long time. On February 2, 2012, Shiraki et al. (2013b) compared radiocaesium levels between washed and unwashed tea plants and found no difference in ^{137}Cs concentrations of the new shoots in 2012. Thus, radiocaesium uptake by above ground plant parts was obviously no longer relevant 11 months after the accident. Probably within a short time period after deposition on the tea plants, radiocaesium deposited on the new shoots of tea plants was taken up by the whole plant.

Thus, after both the Chernobyl and FDNPP accidents, Cs stored in the tea plants was rapidly translocated to new shoots for the first year or the first two years but then, for the slow Cs component, the initial contamination route of the plant became less important for long-term considerations. It should be noted that the long-term activities in green tea is the result of both, the residual activity from the initial deposition on a plant and the radiocaesium taken up from soil. This means that the decline of uptake from soil is also reflected in the effective half-life. As both processes occur simultaneously, their actual contributions cannot be quantified.

4. Conclusions

Radiocaesium decline in Japanese green tea following deposition of radionuclides were calculated using the two-exponential function and

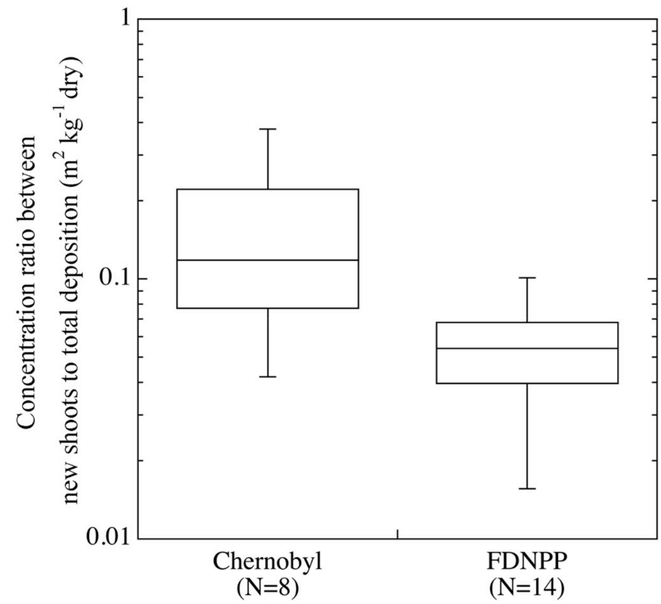

Fig. 5. A comparison of radiocaesium CRs between first new shoots to total deposition.

effective half-lives of fast and slow components were reported. Data from monitoring studies performed in Japan after the accidents in the Chernobyl and the FDNPP were used and compared. The contamination conditions for the new shoots of tea plants were different for these two accidents; for the FDNPP accident, translocation of radiocaesium deposited onto old leaves and twigs to the new growth was the main pathway, while direct deposition on the new shoots was the major source after the Chernobyl accident and it was lower than that in 2011.

For fast T_{eff} and slow T_{eff}, the ANOVA test did not show significant differences between these two accidents. Thus, it was clear that ^{137}Cs declines in new shoots of tea plants after these accidents in a similar manner. Detailed measurements were carried out using ^{137}Cs levels of an azalea tree after the FDNPP accident and from the similar T_{eff} values, we concluded that azalea can be a proxy of tea plant. These T_{eff} observation results implied that radiocaesium concentration would not increase in tea leaf products in the future.

Further analysis was carried out by calculating concentration ratios of ^{137}Cs between new shoots at the time of first harvest (Bq kg^{-1} dry) and total deposited amounts (Bq m^{-2}) in 1986 and 2011 were different; thus, the effect of direct contamination was elucidated. However, further studies are needed to clarify why T_{eff} did not differ between the Chernobyl and FDNPP accident.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2019.106109.

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