Evaluation of the deposition of ^{137}Cs in Japanese persimmon trees and yuzu trees from rainfall by collecting raindrops with sphagnum pads

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Abstract

The radiocaesium emitted due to the Tokyo Electric Power Fukushima Daiichi Nuclear Power Plant accident, initially migrated into fruit trees via the above-ground part of the trees. The behavior of the intercepted ^{137}Cs was evaluated for 6 persimmon and 22 yuzu trees. Radiocaesium can be deposited as dry or wet sediment. To measure the amount of ^{137}Cs in stemflow and raindrops, a collection pad comprised of sphagnum was used. This was attached to trees in a persimmon orchard and several yuzu orchards over 3 years, starting in 2016. Additionally, the effect of bark-washing on persimmon was examined by measuring the amount of ^{137}Cs trapped by the sphagnum pads on the leaf, calyx and main trunk following rainfall. The amount of ^{137}Cs mm^{-1} of precipitation retained by leaves was highest in 2016 in both types of fruit tree and decreased with time. The highest ^{137}Cs interception was detected on the same individual trees in 2016 and again in 2017. In the yuzu orchard in 2018, a higher concentration of ^{137}Cs in fruit than that in previous year were detected in the tree with more ^{137}Cs collected on the leaves than that in the previous year. This indicates the increase in the concentration of ^{137}Cs in fruit also depended on ^{137}Cs contamination on the leaves. These findings demonstrated that it is possible to identify trees which intercepted higher levels of ^{137}Cs during rainfall using sphagnum pads. Therefore those trees are most likely to have more contaminated leaves or fruits.

Keywords: calyx, leaves, raindrop, stemflow, Fukushima Daiichi nuclear power plant accident

INTRODUCTION

The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, caused by the Great Eastern Japanese earthquake occurred on March 11, 2011. A large quantity of radioactive material was released into the environment from March 12 to 14, 2011 (International Atomic Energy Agency, 2015). The radioactive deposition occurred primarily via rainfall and snowfall. The highest radioactive levels were recorded on March 15, 2011 (Chino et al., 2011). The Fukushima Prefecture is one of the major deciduous fruit production areas in Japan (Ministry of Agriculture, Forestry and Fisheries, 2018). Fallout and contamination occurred over a wide range of different fruit tree orchards in the Fukushima Prefecture. Therefore, a focus was on researching the environmental pathway for radioactive particles to enter fruit in these orchards. Both ^{137}Cs and ^{134}Cs were deposited with ^{137}Cs constituting the major concern for orchards due to its long physical half-life (30.1 years), so only this radionuclide will be considered here. The ^{137}Cs activity concentration in most deciduous fruit decreased with time, and by 2020 was below the detection limit for most fruits, with the exception of Ampo-gaki persimmon and yuzu (*Citrus junos* Siebold ex Tanaka). Ampo-gaki is a local specialty in the northern region of the prefecture and produced by drying Japanese persimmon (*Diospyros kaki* Thumb.) which increases the ^{137}Cs concentration compared with the fresh fruit. Yuzu fruit is also a local specialty in Fukushima

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city, and used mainly as a flavoring material in various processed foods. In the initial period after the FDNPP accident, the interception of ^{137}Cs by the leaves and fruit of the evergreen yuzu trees resulted in 10-folds increase of ^{137}Cs concentrations compared to other deciduous fruit (Hamada et al., 2012; Sato, 2020). The concentration of ^{137}Cs in fruit for persimmon and yuzu trees decreased within a few months to values that were below the provisional activity limits of 100 Bq kg^{-1} FW, applied to general foods from April, 2012. However, a small number of fruit still exceeded the limits.

One aspect considered, was the potential for secondary contamination. Migration of intercepted ^{137}Cs due to rainfall onto the fruit tree canopy. This may come from the surrounding environment, such as the forest surrounding the orchards (Sato et al., 2015). As the FDNPP accident occurred during the dormancy period for deciduous orchards in Fukushima, ^{137}Cs was deposited directly onto the bark of the fruit trees and directly onto the topsoil. ^{137}Cs was also transferred to the soil via throughflow (wet leaves shed water onto the ground surface) and water flowing over bark, termed “stemflow” (Schimmack et al., 1993; Kato et al., 2012; Loffredo et al., 2014). The dynamics of throughfall and stemflow in forests has been widely reported (e.g., Loustau et al., 1992; Steinbuck, 2002; Mattaji et al., 2012). However, there have been few studies of these processes in fruit tree orchards.

There was some concern that ^{137}Cs may be transferred from the fruit tree surfaces exposed during the dormant period to leaves and fruits due to rainfall during the growing season. In response, a research study was initiated to specify and quantity the routes for ^{137}Cs contamination in fruit orchards. Initial studies showed that sphagnum pads effectively collected ^{137}Cs in stemflow and raindrops on fruit trees (Sato et al., 2017a, b). In this study, we investigated the amount of ^{137}Cs in the stemflow from the canopy and in raindrops transferred to the calyx or leaves during rainfall.

MATERIALS AND METHODS

Study sites and trees

A Japanese persimmon orchard in the Date city (37°47’43.1”N; 140°34’14.3”E) and a yuzu orchard in Fukushjma city (37°46’25.5”N; 140°28’24.8”E), approximately 60 and 65 km northwest of the FDNPP were selected for investigation in 2011 to 2019. Six Japanese persimmon ‘Hachiya’ trees (Figure 1A) and 22 yuzu trees (Figure 1B) were used in this study. The yuzu orchard was located in a mountainous area, so the yuzu trees were planted on a steep sloping site, and some trees were directly below a road. Three of the persimmon trees were washed (“washed tree”, WT) with a high-pressure washer on December 21, 2011 (Sato et al., 2015). The other trees, including 22 yuzu trees, were not washed (“unwashed tree”, UWT). Five cm of topsoil in the yuzu orchard was removed as part of a decontamination procedure from March to August in 2015.

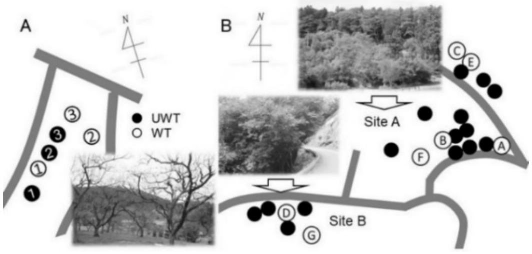

Figure 1. Diagrametic representation of the survey orchards and trees. Japanese persimmon orchard. WT means “washed tree”. These were washed with a high-pressure washer and UWT means “unwashed tree” (A). Yuzu orchards. Six trees at A to F were used to measure the amounts of deposited ^{137}Cs onto the leaves of the canopy and surrounding forest during rainfall from 2016 to 2018 (B). Twenty-two trees including trees for A to F were used for the study to collect ^{137}Cs in stemflow through the trunk during the spring growth flush in 2019.

Preparing the ^{137}Cs collection pads

Two size of the sphagnum collection pads were made by encapsulating approximately 2.5 g (for calyx and leaf) and 6 g (for trunk) of sphagnum (Besuguro-Supagumosu, New Zealand, NZ) into a tea pack (polyester polyethylene composite fiber). Sphagnum from NZ was used because it was expected to contain a negligible amount of ^{137}Cs from past nuclear accidents and weapons testing (Sato et al., 2017a).

Studies in the Japanese persimmon orchard

The different aspects of this study are outline in Table 1. To quantify concentrations of ^{137}Cs in the stemflow due to bark washing, sphagnum pads were tied onto the trunk of UWT and WT in 2015. The sphagnum pads were covered with polyethylene film to shield the pads from throughfall (Figure 2). In 2016, four locations in each of 3 UWTs were selected for a study that aimed to quantify the relationship between the amounts of deposited ^{137}Cs on the calyx and the concentration of ^{137}Cs in fruit due transfer by raindrops from the canopy. Sphagnum pads were attached onto the calyx of the fruit. Another fruit on the same branch was also bagged as a control. The concentration of ^{137}Cs from bagged mature fruit and non-bagged fruit with attached sphagnum pads on the same branch were compared. The deposited ^{137}Cs on the leaf of the UWT and WT were compared in both 2017 and 2018. To monitor the annual contamination status in the study trees, the bark of the main trunk of UWT and WT were initially sampled to measure the concentration of ^{137}Cs on October 18, 2011 prior to bark washing. Subsequent, measurements were taken on October 28, 2013, June 4, 2015, June 2, 2017 and April 9, 2018.

Figure 2. Images of the methods used to collect ^{137}Cs in stemflow and raindrop using a sphagnum pad. To collect the ^{137}Cs in stemflow, sphagnum pads were attached onto the trunk (A) of UWT and WT in the Japanese persimmon orchard and of UWT in the yuzu orchard. To collect the deposited ^{137}Cs, sphagnum pads were attached onto the calyx (B). To study the effect of deposited ^{137}Cs on the calyx and the concentration of ^{137}Cs in mature fruit, a fruit on a lateral branch was selected and a sphagnum pad attached and bagged, control treatment (C). To collect the intercepted ^{137}Cs by leaves, sphagnum pads were attached onto leaves of the persimmon and the yuzu trees (D).

Studies in the yuzu orchard

An outline of the study is also shown in Table 1. Sphagnum pads were attached onto leaves at the marginal section of the canopy of the yuzu trees. This was to confirm the amounts of deposited ^{137}Cs on the leaf during rainfall (Figure 2). Four trees for A to D shown in Figure 1 were surveyed for a period of three years from 2016 to 2018. The concentration of ^{137}Cs in mature fruit was measured by collecting five fruits from each tree to explore the relationship between the amount of ^{137}Cs deposited on leaves and the concentration of ^{137}Cs in the fruit. In 2019, sphagnum pads were attached onto the trunk of 22 selected trees to trap the ^{137}Cs in stemflow during the spring flush of rapid growth (Figure 2). Leaves of the spring shoots were collected on June 26 to measure the ^{137}Cs concentration. The ratio of the concentration of ^{137}Cs in leaves of the spring high growth period, June 26, 2019 was compared to that on June 13, 2018. A regression analysis was carried out between the amounts of the ^{137}Cs in stemflow and the annual ratio of the concentration of ^{137}Cs in leaves of the spring shoot.

Table 1. Study outline for collection of ^{137}Cs in raindrops and stemflow using a sphagnum pad from two experimental orchards.

Orchard	Study year	No. of trees	Collection period	Location of attached pad	No. of pads tree^{-1}	Sampling days of fruit or leaf
Japanese persimmon	2015	3UWT&3WT	Jul. 8-23	Trunk	1	Oct. 19 (fruit)
	2016	3 UWT	Jun. 15-Oct. 19	Calyx	4	
	2017	3UWT&3WT	Jun. 15-Sep. 20	Leaf	3(WT), 5(UWT)	
	2018	3UWT&3WT	Jun. 27-Oct. 24	Leaf	3(WT), 4(UWT)	
Yuzu	2015	13 UWT	-	-	-	Nov. 6 (fruit)
	2016	4 UWT	Jun. 25-Oct. 28	Leaf	6	Nov. 14 (fruit)
	2017	6 UWT	Jun. 12-Oct. 18	Leaf	4	Oct. 25 (fruit)
	2018	5 UWT	Jun. 13-Oct. 25	Leaf	4	Jun. 13 (leaf), Oct. 25 (fruit)
	2019	22 UWT	Jun. 14-Jun. 26	Trunk	2	Jun. 26 (leaf)

Sample treatments and radiocaesium measurements

The sphagnum samples attached to the trunk of the Japanese persimmon and the yuzu trees were removed from the tea pack and placed in a 100 mL polypropylene container (U-8 pots). Whereas the sphagnum pads and tea packs attached onto the calyx and leaves were directly measured for ^{137}Cs. Bark samples were collected from five trees prior to washing on October 18, 2011. These were combined into one sample for ^{137}Cs concentration measurement. Bark samples from each tree were collected after 2013. The Japanese persimmon fruit were shredded using a food processer, after peeling, removing seeds and calyxes. Yuzu fruits were separated into the pericarp (flavedo with albedo) and pulp with seeds. Leaves and fruit were placed in U-8 pots after freeze-drying for at least 72 h. The concentration of ^{137}Cs in all samples measured using a germanium detector (GEM40-76 germanium detector, Seiko EG&G ORTEC, Tokyo, Japan) at the Fukushima University. Gamma-ray emission at energies of 662 keV measured for 3,600 to 80,000 s. The concentration of ^{137}Cs in bark was converted to a DW basis using a dry-to-wet ratio after drying for one day at 105°C. After freeze-drying, the concentration of ^{137}Cs in the fruit of yuzu trees was converted to a wet weight basis using a dry-to-wet ratio. The means of the measured values from the pericarp and pulp with seeds was assumed to represent the concentration of ^{137}Cs in yuzu fruit.

Calculation of the effective half-life of the concentration of ^{137}Cs in bark of Japanese persimmon and of the retention of ^{137}Cs during rainfall

A single negative exponential model expressed as a predictor variable of the temporal changes in the concentration of ^{137}Cs in the bark during the first year after the nuclear accident (Antonopoulos-Domis et al., 1991). This was obtained by the least-squares method adopted for the quasi-Newton method:

$$y = K\exp(-\lambda x) \tag{1}$$

$$T_{eff} = \ln 2\ \lambda^{-1} \tag{2}$$

where y is the concentration of ^{137}Cs in the barks collected in the xth year after the nuclear accident; K is the concentration in the year of the nuclear accident; x is the number of years after the nuclear accident; and λ is the decay constant; T_{eff} is the effective half-life.

The retention of ^{137}Cs due to rainfall was represented as the following.

$$R = 1000A\ P^{-1}\ (\text{mBq g}^{-1}\ \text{mm}^{-1}) \tag{3}$$

where R is the retention of ^{137}Cs, A is the amount of ^{137}Cs intercepted per 1 g of sphagnum (Bq g^{-1}) and P is the amount of precipitation during the collection period at each orchard (mm).

RESULTS AND DISCUSSION

Studies in the Japanese persimmon orchard

The concentration of ^{137}Cs in the bark of both the UWT and WT decreased with time. The concentration of ^{137}Cs in the bark of WT was significantly lower than that of UWT by approximately one order of magnitude for each year of sampling (Figure 3A). The measured values of UWT and WT were fitted to a single negative exponential model. However, the concentrations of ^{137}Cs in the bark for both treatments in the last year of measurement (2018) were lower than the values predicted by the relevant model. The T_{eff} values for the UWT and WT were 2.1 and 3.2 years. The amount of ^{137}Cs g^{-1} DW sphagnum (mean ± SD) encapsulated in the tea pack collected from stemflow on the main trunk of UWT and WT was 1.08±0.15 and 0.37±0.12 Bq g^{-1}, respectively. There was significantly lower amounts of ^{137}Cs from the trees that had been bark-washed (at p=0.0003 by t-test). Similarly, the retention of ^{137}Cs on leaves via raindrop significantly lower in bark-washed trees (Table 2). These results indicate that the concentration of ^{137}Cs in stemflow and raindrops in the canopy reflected the contamination status of the bark during six years after FDNPP accident, especially if there is a 10 times difference in pollution levels. The amount of ^{137}Cs in the sphagnum pad attached onto the calyx in UWT was 0.04-1.6 Bq g^{-1} DW per 1g of sphagnum. The higher the value of ^{137}Cs intercepted within one tree, the larger was the range of variation, as shown by tree UWT1. Figure 3C shows the concentration of ^{137}Cs in the fruit which was bagged, non-bagged or equipped with sphagnum pad on the same lateral branch, expressed as Bq kg^{-1} DW. The amounts of ^{137}Cs (Bq g^{-1} DW) of sphagnum pads attached on the calyx in four locations of the branch are reported on the *x*-axis. There was no significant correlation in the concentration of ^{137}Cs in fruit due to bagging or to the amount of intercepted ^{137}Cs by sphagnum pads, whereas there were significant differences in the concentration of ^{137}Cs in fruit within a branch, which varied by up to three folds.

Table 2. The retention of ^{137}Cs on sphagnum pad attached onto calyx or leaves of Japanese persimmon. Arithmetic mean and standard deviation of the values per year and ANOVA results are reported.

Bark washing	Replicate tree label (Figure 1)	^{137}Cs retained (mBq g^{-1} mm^{-1})[a]		
		Year[b]		
		2016 (Calyx)	2017 (Leaves)	2018 (Leaves)
UWT	1	0.79	0.60	0.08
	2	0.27	0.16	0.17
	3	0.67	0.18	0.12
		0.58±0.27	0.31±0.25	0.12±0.05
WT	1		0.10	0.08
	2		0.09	0.04
	3		0.19	0.02
			0.13±0.06	0.05±0.03
p value by ANOVA[c]	Year		0.013*	
	Bark washing		0.014*	

[a] Accumulated precipitation (mm) during the collection period in 2016, 2017 and 2018 were 680, 449 and 408.

[b] Sphagnum pads were attached on calyx in 2016 and on leaves in 2017 and 2018.

[c] The measured values in 2017 and 2018 by logarithmic transformation. Significance: *p<0.05.

Based on the effective half-life of ^{137}Cs, there was a considerable reduction in the concentration of ^{137}Cs in the bark due to bark washing for up to the first three years after the accident. High pressure bark washing conducted six months after the FDNPP accident on Japanese persimmon trees reduced the concentrations of ^{137}Cs in fruit by about 30% (Sato et al., 2015). Previous studies have indicated that there may be secondary contamination of fruit from the bark. Sekizawa et al. (2016a, b) and Sato (2020) demonstrated that ^{137}Cs migrated to fruit via the calyx. In this study, the bagged fruits showed higher concentrations than that of the non-bagged fruit on the same branch. This is in contrast to the prior studies. The source of ^{137}Cs migration was unlikely to be on the calyx. Which is consistent with the view of Sekizawa et al. (2016b) that stemflow and throughflow was unlikely to influence the concentration of ^{137}Cs in the fruit via the calyx. An alternative potentially important pathway was suggested by Sato (2020) who demonstrated that the transfer rate of ^{137}Cs via leaves at the young fruit stage or the fruit growing stage was 10.2±3.3 or 16.4±5.0% in the Japanese persimmon 'Hachiya'. This previous study indicated the transfer rate of ^{137}Cs via leaves depended on the distance between fruit and the contaminated leaf. If the deposition onto leaves is assumed to be similar to that on the calyx, differences in the orders of two magnitudes were found by this study. In addition, contamination of the leaves may contribute to the differences in the concentrations of ^{137}Cs in fruit for the year 2016 (Figure 3).

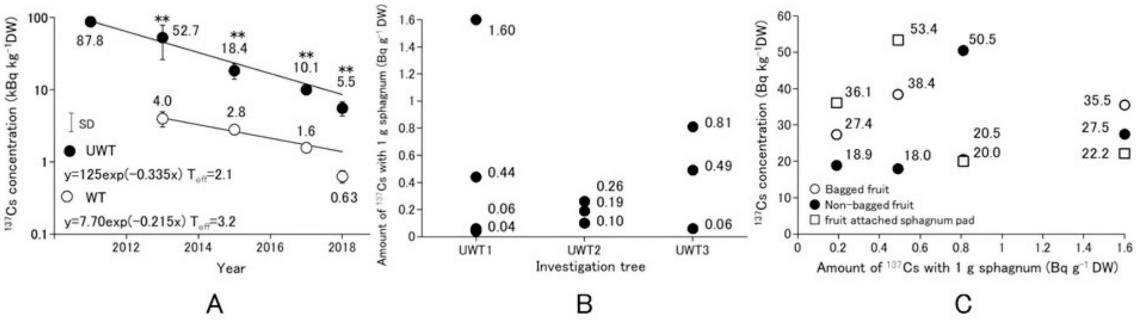

Figure 3. Comparison of change with time in the concentration of ^{137}Cs in bark of Japanese persimmon between bark-washed tree and unwashed trees (A); the amount of ^{137}Cs intercepted by sphagnum pad attached on the calyx in the three UWT in 2016 (B) and the concentration of ^{137}Cs in the bagged, and non-bagged fruit and fruit with a sphagnum pad (non-bagged) on the same lateral branch (C). One sphagnum pad in each of UWT2 and UWT3 dropped in (B). Significance: **$p<0.01$ by t-test in (A).

Studies in the yuzu orchard

Over the two years from 2016, the amount of ^{137}Cs accumulated in the sphagnum pad attached to yuzu leaves decreased by one-third. However, there were large differences between individual trees (Figure 4A). Figure 4B, C show changes with time in the retention of ^{137}Cs by the sphagnum pads and the concentration of ^{137}Cs in fruit in four trees for A to D over 3 years. There was no significant difference in the retention of ^{137}Cs among trees in 2016 or 2017. This was due of large differences in the amount of intercepted ^{137}Cs within one tree which is consistent with the observations on Japanese persimmon. Furthermore, the retention of ^{137}Cs in tree D was significantly higher than any of other two trees ($p<0.05$ by Tukey's test) sampled. From 2017 to 2018, an increase in the retention of ^{137}Cs was observed (Figure 4B). The concentration of ^{137}Cs in fruit of tree A and C decreased each year, whereas that of tree B increased markedly in the two years after 2015 and then decreased in 2018. The roots of B tree were exposed at the time of soil removal and subsequent heavy rain may enhanced the absorption of ^{137}Cs. Interestingly, the concentration of ^{137}Cs in fruit of tree D (Figure 4C) had the same trend with year as that of the retention of ^{137}Cs (Figure 4B). Furthermore, there was a significant positive correlation between the retention of ^{137}Cs on leaves in 2019 and the annual ratio of the concentration of ^{137}Cs in leaves of the spring shoot in 2019-2018 (Figure 5). In the yuzu orchard, four trees in site B, planted just below the road had higher ^{137}Cs retention (Figure 5).

The yuzu orchard area is located on a steep slope surrounded by the forest consisting of evergreen conifers, evergreen broad-leaved trees and deciduous broad-leaved trees.

Sphagnum pads were attached onto the leaves on the outer margin of the yuzu tree canopy, enabling collection of raindrops diffused from the surrounding environment. The highest retention of ^{137}Cs was detected in tree C in 2016 (Figure 4B) probably because it was directly under evergreen conifers. However, Figure 4A, B show the amount of ^{137}Cs deposition from the environment was reduced due to weathering (Kato et al., 2012 and 2017). Conversely, a similar time trend was found for the retention of ^{137}Cs. In 2018, the concentration of ^{137}Cs in fruit increased in tree D. This was caused by secondary contamination due to raindrop diffused from the environment. Shiraishi (1973) indicated ^{137}Cs migrates to pulp directly via the contaminated surface pericarp in Satsuma mandarin (*Citrus unshiu* Marc.). Sato (unpublished) has demonstrated that the transfer rate of ^{137}Cs via spring leaves in yuzu was a third higher than that of the deciduous trees. From these findings and the result of the study in 2019 (Figure 5), it seems that yuzu may be subjected to secondary contamination of leaves or fruits via raindrops/rainfall.

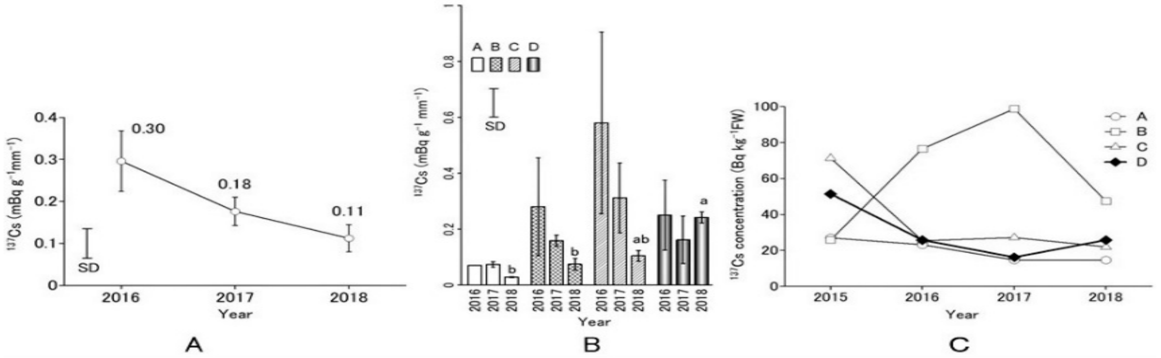

Figure 4. Changes in the retention of ^{137}Cs on the sphagnum pad attached on yuzu leaves over time (A, B), the concentration of ^{137}Cs in fruit (C). Different letters in graph B indicate a significant difference at $p<0.05$ by Tukey's test using the measured value in 2018.

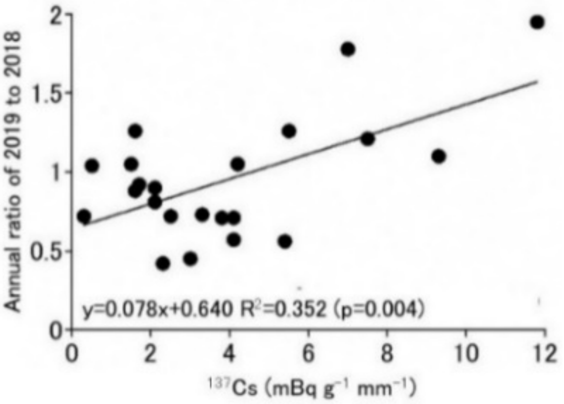

Figure 5. Relationship between the retention of ^{137}Cs in 2019 and the annual ratio of ^{137}Cs concentration in leaves for 2019 to 2018.

CONCLUSIONS

The following conclusions can be drawn from this study:

- The amount of ^{137}Cs in stemflow and raindrops in the canopy of Japanese persimmon reflected the contamination status of the bark for at least 6 years after the FDNPP accident.
- ^{137}Cs migration into fruit via calyx in Japanese persimmon was negligible. The main pathway of ^{137}Cs in stemflow and raindrop into fruit is more likely to be via leaves.

- The amount of collected ^{137} Cs per 1 mm of precipitation was higher in 2016 (the first year of measurement) in both Japanese persimmon and yuzu orchards and thereafter decreased with time.
- It was confirmed that secondary contamination of leaves or fruits in yuzu was due to radiocaesium in raindrops.
- Yuzu seems to be more susceptible to the secondary contamination of leaves or fruits than Japanese persimmon.
- Sphagnum pads were a useful tool to quantitatively evaluate secondary contamination from the canopy and the environment.

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