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Estimation of Radioactive Cesium Concentration in Persimmon Fruits of Different Branches of Individual Trees

Sachie HORII[1]*, Haruhito SEKIZAWA[2], Mayumi HACHINOHE[3] and Shinnosuke KUSABA[1]

[1] Institute of Fruit Tree and Tea Science, National Agriculture and Food Research Organization, Tsukuba, Japan
[2] Agriculture, Forestry and Fishery Department, Fukushima Prefectural Government, Fukushima, Japan
[3] Institute of Food Research, National Agriculture and Food Research Organization, Tsukuba, Japan

Abstract
This study investigated the variations in ^{137}Cs concentrations in the fruits and leaves of persimmon trees, which had been affected by radioactive cesium for several years. The trees were divided into lateral or main branches, and ^{137}Cs concentrations of fruits and leaves in the branches were measured. The ^{137}Cs concentration differed among the fruits of the branches of the same tree, and the difference in ^{137}Cs concentration in the fruits of each branch was confirmed for multiple years. A difference in ^{137}Cs concentration of approximately ten times was found between the fruits of each branch on the same tree. In addition, a correlation was found between ^{137}Cs concentrations in fruits and leaves. These results suggest that a difference might exist in the ^{137}Cs concentration of each branch of the tree at the time of fallout in 2011, and it was considered that the difference might persist.

Discipline: Horticulture
Additional key words: “Ampo-gaki,” Cs-137, dried fruit, radioactive fallout

Introduction

In 2011, radioactive material was released during the accident at the Fukushima Daiichi Nuclear Plant, operated by the Tokyo Electric Power Company. Radioactive materials, such as cesium, fell to fruit-producing areas in the Fukushima Prefecture. Deciduous fruit trees, except the Japanese apricot, were present before germination, and radioactive materials accumulated on the bark of the main stem and skeletal branches (Takata 2013). Moreover, radioactive cesium was detected in the endodermis of peach trees on June 16, 2011. Large amount of radioactive cesium has migrated from the bark to the inner tissues of the tree within 60 days of contamination (Abe et al. 2012, Sato 2018, Takata et al. 2020).

Since many radioactive substances were found in the bark, whether bark removal and cleaning would reduce the concentration of radioactive cesium in fruits was investigated. Although the removal rate differs due to varying peeling properties of barks of different tree species, the radiation dose rate on the tree surface decreased upon bark removal and cleaning. As a result of this survey, bark removal and cleaning with a high-pressure washing machine were performed in the orchards (Agriculture, Forestry and Fishery Department of Fukushima Prefecture 2012). The decontamination effect by the tree washing treatment persisted until 2018, that is, after 7 years of the treatment (Sato 2018).

The cesium concentration in semi-dried persimmons (“Ampo-gaki”), which is a specialty product of Fukushima Prefecture, increases due to the reduction in water content. In some cases, the radioactive cesium concentration in the product exceeded the Japanese standard value for general food (^{134}Cs+^{137}Cs, 100 Bq/kg). Therefore, production was discontinued in Date City of Fukushima Prefecture and the surrounding area for 2 years after the accident. As the concentration of radioactive cesium in raw fruit has been decreasing since

*Corresponding author: horiis404@affrc.go.jp
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2013, processing of "Ampo-gaki" has resumed in a limited area. This area is called the "processing model area," which refers to an area where the radioactive cesium concentration of young and ripe fruits was low, and more than 80% of the fields had a radioactive cesium concentration of 10 Bq/kg or less. Screening level (SL) (^{134}Cs+^{137}Cs, 50 Bq/kg) is a quick method used to identify products that are below the Japanese standard value during pre-shipment inspection. All processed "Ampo-gaki" were subjected to nondestructive screening inspection for radioactive substances, and only products with radioactive cesium below the Japanese standard value were distributed. The percentage of products that exceeded the SL has been low since the beginning of processing in 2013 and was 0.003% in 2020 (Fukushima Prefecture Ampo-gaki Production Area Promotion Association, 2021).

A survey of trees that produced fruits with excess SL revealed that very few trees with higher ^{137}Cs concentrations in fruits were cultivated at the edge of the orchard (Sekizawa et al. 2019). In addition, ^{137}Cs concentrations in the leaves and fruits have been reported to be highly correlated (Horii et al. 2019). Therefore, trees with high ^{137}Cs leaf concentrations were cut and new trees were replanted in fields where the products exceeded SL. As a result, the number of products that exceeded the SL decreased annually, but products that exceeded the SL are still detected rarely. Although the difference in ^{137}Cs concentration in fruits in the field has been clarified (Sekizawa et al. 2019), the difference in ^{137}Cs concentration in fruits of the same tree has not been investigated. Horii et al. (2022) reported that the concentration of ^{137}Cs in fruits and leaves varied depending on their position (e.g., branches which they are locating) even within an individual tree. However, this paper reported the only result of a single tree, and thus more detailed investigation with using multiple trees was required for obtaining a more concrete conclusion. Therefore, this study investigated the differences of ^{137}Cs concentration in fruits from each branch (either main or lateral one) with using multiple trees, and then examined the variation in ^{137}Cs concentration in the fruits within each tree. The results were then compared among multiple trees, in order to clarify the difference between trees.

Materials and methods

Four trees (A-D) were surveyed in three orchards in Date City, Fukushima Prefecture, Japan. Trees A and B were cultivated in the same orchard. The air dose rates (measured at 1 m above the ground in March 2018) around trees A and B were 0.48 μSv/h and that around trees C and D were 0.12 and 0.54 μSv/h, respectively. Trees A and B were surveyed for 2 years (2018-2019), and trees C and D were surveyed for 3 years (2017-2019). The lateral branches of trees A, B, and C were segmented based on the branch circumference (≥ 16 cm) (Photo 1). The circumference of the thickest branch was 20 cm. The branches were numbered in a counterclockwise direction from the east of the tree. Tree D was divided into four main branches. In 2011, all trees were washed using a high-pressure washing machine in all three fields. Four to five fruits were sampled from each lateral branch of trees A, B and C. Three to five fruits were sampled from each main branch of tree D during the harvest season. The harvest was at late October in 2017 and 2018 and at late September in 2019 (in 2019, fruits were harvested earlier than usual because the trees were scheduled to be cut in October). The harvested fruits were washed with tap water. The fruits were peeled, and the calyxes and seeds were removed. Thereafter, the pulp of each fruit was crushed using a mixer and filled in a U8 container (RIG, Yokote, Akita, Japan). The ^{137}Cs concentration in the fruits was measured using a germanium semiconductor detector (GC2520-7500SL and GC4020-7500SL) (CANBERRA, Meriden, CT, USA). In addition, the leaves of each branch were randomly collected in September 2019 to clarify the difference in ^{137}Cs concentration in the leaves of different branches. More than 100 leaves were sampled from each branch that were without insect feeding damage or lesions. The leaves were then washed and dried in a dryer at 70°C until no change in weight was observed. They were crushed using a mixer and filled in a U8 container. ^{137}Cs concentration was measured using a germanium semiconductor detector. Statistical analysis was performed using the R software (R Core Team, 2019).

Results

1. Change in ^{137}Cs concentration of fruits

Tree A consisted of 13 lateral branches. In 2018, the fruits of branch number (No.) 4 had a higher ^{137}Cs concentration than that of the fruits of other branches, except branch No. 13 (Fig. 1). The average ^{137}Cs concentration of fruits of branches 1, 2, 4, 6, 7, 8, 12, and 13 was significantly lower in 2019 than that in 2018.

Tree B consisted of ten lateral branches. The ^{137}Cs concentration in the fruits of lateral branches Nos. 5 and 6 was lower than that in the fruits of other branches, except branch No. 3 (Fig. 2). In 2019, the ^{137}Cs concentration of fruits of branch No. 2 was the highest (33.5 Bq/kg) and 9.7 times higher than the lowest ^{137}Cs concentration of fruits of branch No. 6. The ^{137}Cs

Photo 1. Photograph of the persimmon tree C
The numbers in the photograph represent the number of branch.

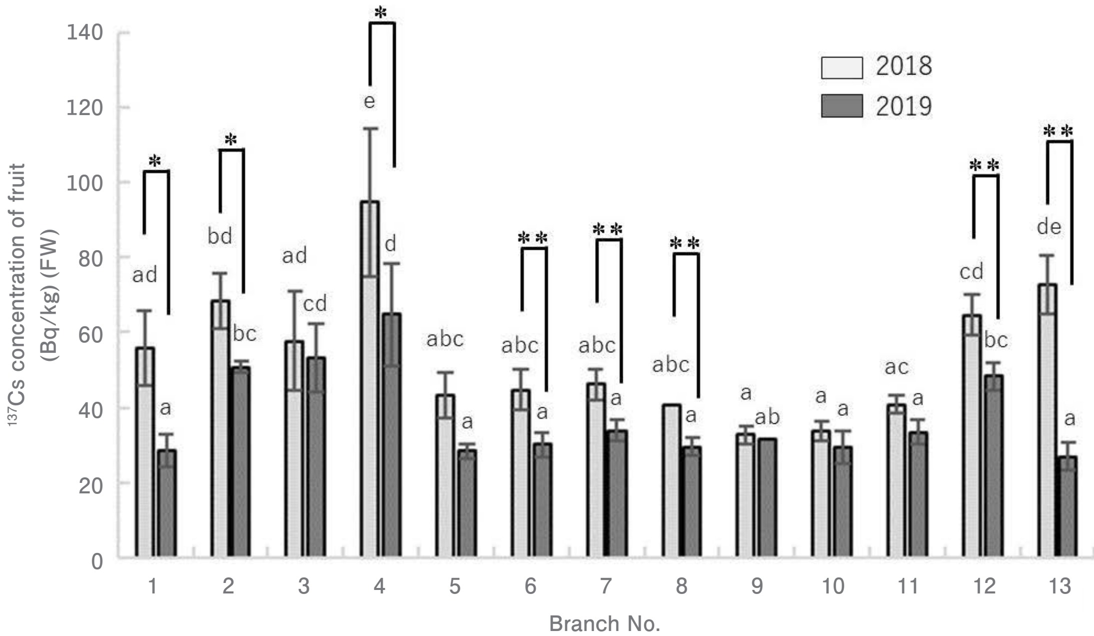

Fig. 1. ^{137}Cs concentration of fruits on different branches of Tree A
Vertical bars indicate the standard deviation (n = 4-5).
Bars with no standard deviation show the average value of ^{137}Cs concentrations in the two fruits.
FW, fresh weight
Different letters indicate significant differences between the average ^{137}Cs concentration in each branch in the same year based on the Tukey–Kramer test at $P < 0.05$.
*($P < 0.05$) and **($P < 0.01$) show significant differences between 2018 and 2019 based on the *t*-test.

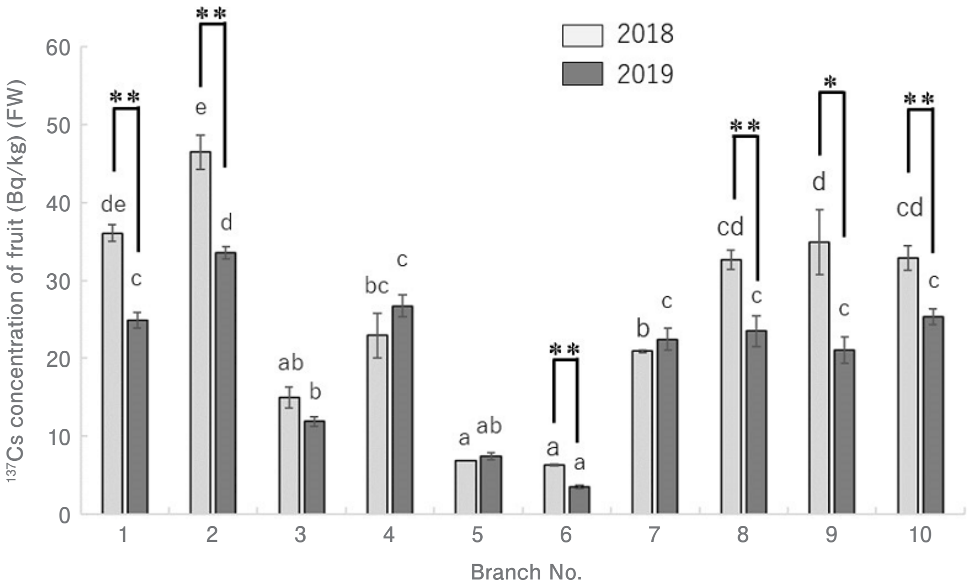

Fig. 2. ^{137}Cs concentration of fruits on different branches of Tree B

Vertical bars indicate the standard deviation (n = 4-5).

Bars with no standard deviation show the average value of ^{137}Cs concentrations in the two fruits.

FW, fresh weight

Different letters indicate significant differences between the average ^{137}Cs concentration in each branch in the same year based on the Tukey–Kramer test at $P < 0.05$.

*($P < 0.05$) and **($P < 0.01$) show significant differences between 2018 and 2019 based on the *t*-test.

concentrations in the fruits of branches 1, 2, 6, 8, 9, and 10 were significantly lower in 2019 than in 2018.

Tree C was segmented into 12 lateral branches. Within 3 years, the ^{137}Cs concentration in the fruits of branches 8 and 9 tended to be lower than that in the fruits of other branches (Fig. 3). In 2019, the ^{137}Cs concentration of fruits of branch No. 3 (38.8 Bq/kg) was 10.2 times higher than that of the fruits of branch No. 8 (3.8 Bq/kg). In addition, the ^{137}Cs concentration in the fruits of branch Nos. 2, 4, and 6 was significantly lower in 2019 than in 2017. Branch No. 8 had a high ^{137}Cs concentration in 2018, which was significantly different from the ^{137}Cs concentrations observed in 2017 and 2019.

In case of tree D, the average value of the ^{137}Cs concentration of fruits of each main branch (i.e., the average ^{137}Cs concentration of the whole tree) was 7.0 Bq/kg, 4.8 Bq/kg, and 3.3 Bq/kg in 2017, 2018, and 2019, respectively (Fig. 4). The values of fruits of each branch ranged from 3.3 to 11.0 Bq/kg, 2.4 to 8.5 Bq/kg, and 2.2 to 4.9 Bq/kg in 2017, 2018, and 2019, respectively. In 3 years (2017-2019), the ^{137}Cs concentration in the fruits of branch No. 4 remained higher than that in the fruits of branch No. 2. The ^{137}Cs concentration in the fruits of branches No. 2, 3, and 4 was significantly lower in 2019 than in 2017. Furthermore, the ^{137}Cs concentration in the fruits decreased significantly every year in branch No. 3.

2. Relationship between ^{137}Cs concentrations of leaves and fruits

Although the number of samples was limited, a high correlation was found between ^{137}Cs concentrations in the fruits and leaves of trees A, B, and C (Fig. 5). Among the four trees, the correlation coefficient of tree B was the highest (r = 0.98, $P < 0.01$). The correlation coefficients for trees A and C were 0.89 ($P < 0.01$) and 0.92 ($P < 0.01$), respectively. However, no correlation was found in tree D (r = 0.47, $P = 0.53$).

Discussion

Comparing the ^{137}Cs concentration of the fruits from all branches of tree B, the ^{137}Cs concentration of the fruits of branches 5 and 6 was lower than that of the fruits of other branches. Similarly, branches 8 and 9 of tree C had fruits with lower ^{137}Cs concentrations than the other branches. This phenomenon was observed for several years. In the 2019 survey, in tree B, ^{137}Cs concentration per branch was 9.7 times higher in the fruit with the highest average ^{137}Cs concentration than in the fruit with

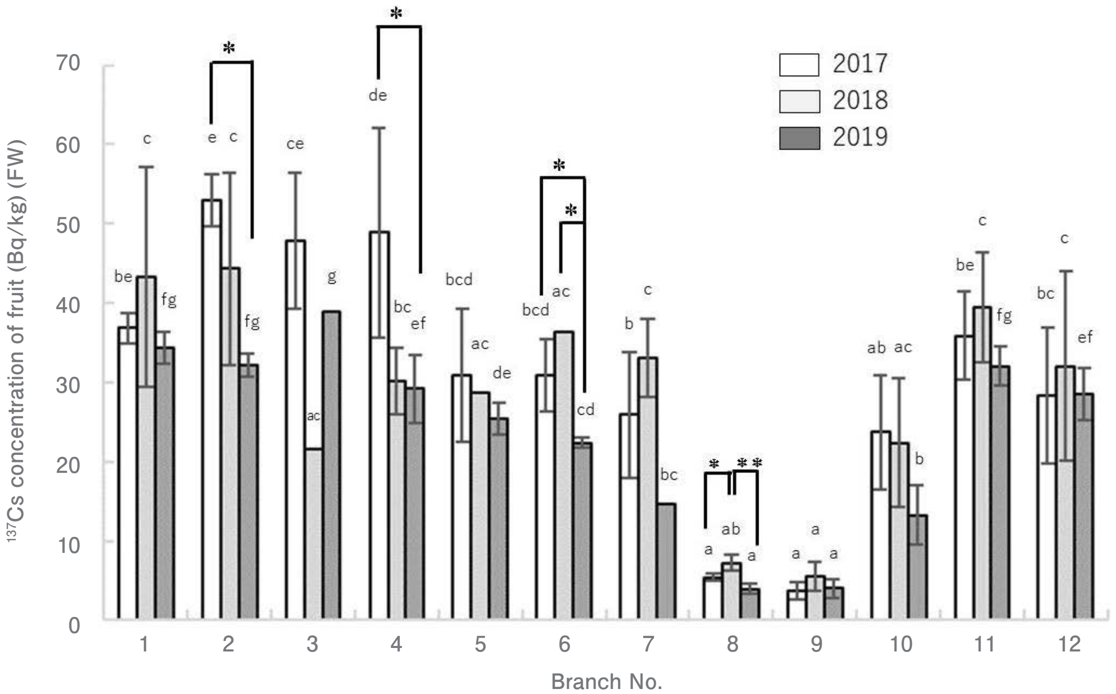

Fig. 3. ^{137}Cs concentration of fruits on different branches of Tree C
Numbers in the graph correspond to the branch numbers in Photo 1.
Vertical bars indicate the standard deviation (n = 4-5).
FW, fresh weight
Bars with no standard deviation show the average value of ^{137}Cs concentrations in the two fruits.
Different letters indicate significant differences between the average ^{137}Cs concentration in each branch in the same year based on the Tukey–Kramer test at $P < 0.05$.
*($P < 0.05$) and **($P < 0.01$) show significant differences between 2017, 2018, and 2019 based on the Tukey–Kramer test.

the lowest concentration. Similarly, in tree C, the average ^{137}Cs concentration difference between the branches with the highest and lowest fruit ^{137}Cs concentrations was 10.3 times. These results indicate that ^{137}Cs concentrations in fruits may differ in different branches of the same tree. When calculated the average ^{137}Cs concentrations in fruits over each branch and compared their highest and the lowest concentrations within each tree, the values showed large fluctuations depending on trees. In addition, the variation of ^{137}Cs concentration in fruits within branches also differed depending on the trees. In a survey of trees that received ^{137}Cs directly, the concentration of radioactive cesium in the current year's branches (new organs) was more than 5.2 times lower in persimmons (Sato et al. 2014) and approximately 10 times lower in peaches (Yuda et al. 2014) and blueberries (Kusaba et al. 2015) than in second year branches. After the fallout of ^{137}Cs, its concentration decreased significantly upon new organ formation. The ^{137}Cs concentration in a newly formed organ is affected by the ^{137}Cs concentration at the site where the organ was differentiated. Therefore, it was considered that ^{137}Cs concentration differed according to branch division. In addition, although no data are shown, no relationship was observed between the number of fruits on the branch and the concentration of ^{137}Cs in this study.

In this study, the branches were divided by their circumference (≥ 16 cm), and all branches were 8 years old or more (in 2017). It is thought that ^{137}Cs fell directly onto the branches in 2011. Although the branches received a drop of ^{137}Cs at the same time as the surrounding branches, some branches had lower ^{137}Cs concentrations in fruits and leaves compared with other branches. Possible reasons for the different ^{137}Cs concentrations in the branches are discussed below. First, it is possible that different amounts of ^{137}Cs dropped on different branches initially. The weights of the lateral branches of tree B (data not shown) were compared to assess the amount of ^{137}Cs that fell on the branches. Heavy branches were assumed to have occupied a lot of space, and a large amount of ^{137}Cs had fallen on them. Therefore, we hypothesized that heavier branches would receive more ^{137}Cs. Branch No. 9 of Tree B was the heaviest (8.1 kg). However, the ^{137}Cs concentration of the fruits of branch

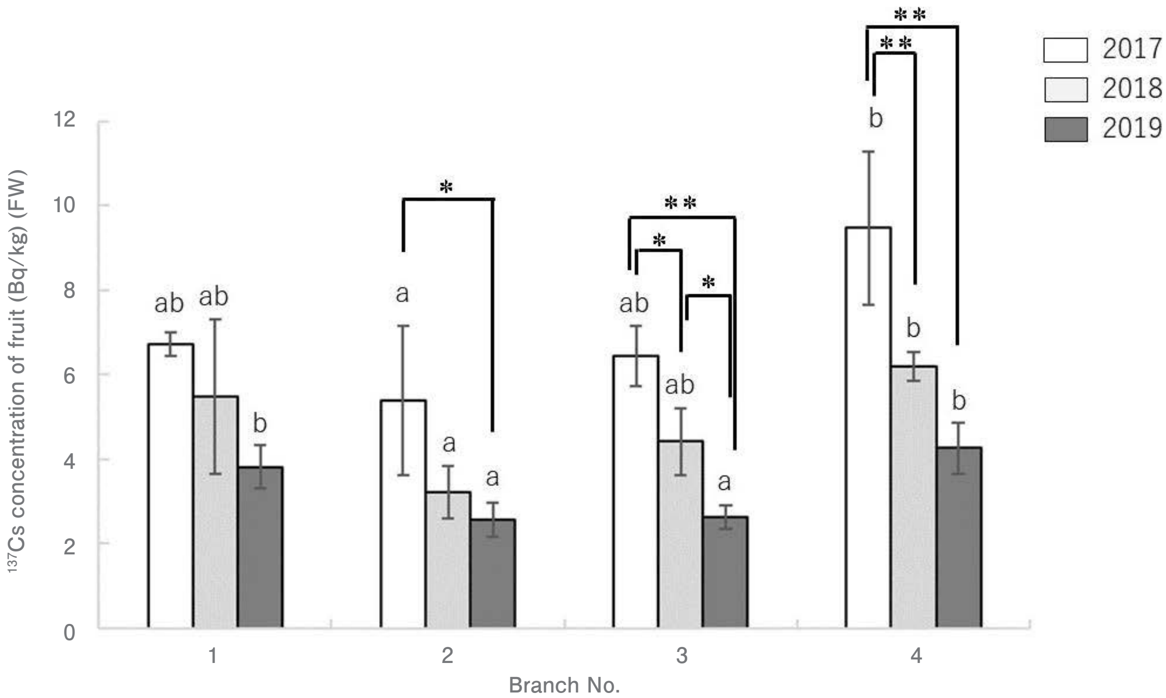

Fig. 4. ^{137}Cs concentration of fruits on different branches of Tree D

Vertical bars indicate the standard deviation (n = 3-5).

FW, fresh weight

Different letters indicate significant differences between the average ^{137}Cs concentration in each branch in same year based on the Tukey–Kramer test at $P < 0.05$.

*($P < 0.05$) and **($P < 0.01$) show significant differences between 2017, 2018, and 2019 based on the Tukey–Kramer test.

No. 9 was 21.0 Bq/kg in 2019, which was the average concentration in each branch in tree B. The relationship between branch weight and ^{137}Cs concentration was weak, even after considering the results of branches other than No. 9 in tree B. Therefore, it is considered that there was a difference in the concentration of the radioactive fallout itself or that the fallout of ^{137}Cs was physically hindered by obstacles. Additionally, the amount of ^{137}Cs on the bark may have differed due to uneven tree washing. It has also been reported that moss grows easily on the bark of persimmon trees, and ^{137}Cs concentration in epiphytic bark is higher than that in non-epiphytic parts (Sato et al. 2017). Because ^{137}Cs is also absorbed through the bark, it is possible that the ^{137}Cs concentration in the fruits might have increased due to epiphytic moss.

Another possibility is that the different absorption rates of ^{137}Cs in different branches, from the surface to the inner tissues, resulted in varying concentrations of ^{137}Cs in the branches. This has also been investigated in other tree species. In cedar, ^{133}Cs on the bark migrates to the xylem through the parenchyma (Aoki et al. 2017). In addition, radioactive cesium applied to the stems and trees of peaches has been detected in its fruits and leaves (Sato et al. 2014). Similarly, radioactive substances are thought to have been absorbed from persimmon bark. Moreover, because the absorption rate varies according to the branch, it is possible that the branch with a high ^{137}Cs concentration absorbed a large amount of ^{137}Cs from the bark. Differences in the distribution of cesium in wood depending on tree species have been reported (Ohashi et al. 2014). It has been shown that the distribution of cesium in wood is related to moisture content (Kuroda et al. 2013, Okada et al. 2012) and concentration of alkali metals (Momoshima et al. 1995, Okada et al.2011), such as potassium (Iizuka et al. 2018). These factors are thought to be related to absorption from the surface of the branches. In addition, cesium distribution has been shown to differ depending on the organ in rice (Kondo et al. 2015, Ishikawa et al. 2018). Although rice is an annual plant and its distribution mechanism is different from that of fruit trees, it is possible that the distribution of cesium from the underground parts to the aboveground parts differs depending on the branch or organ in the tree.

The following are the reasons for the varying ^{137}Cs concentrations in fruits and leaves among branches: (1) the amount of ^{137}Cs that fell on the branch was different, (2) the absorption rate of ^{137}Cs from the surface to the inner tissues was different, and (3) the transfer rate of

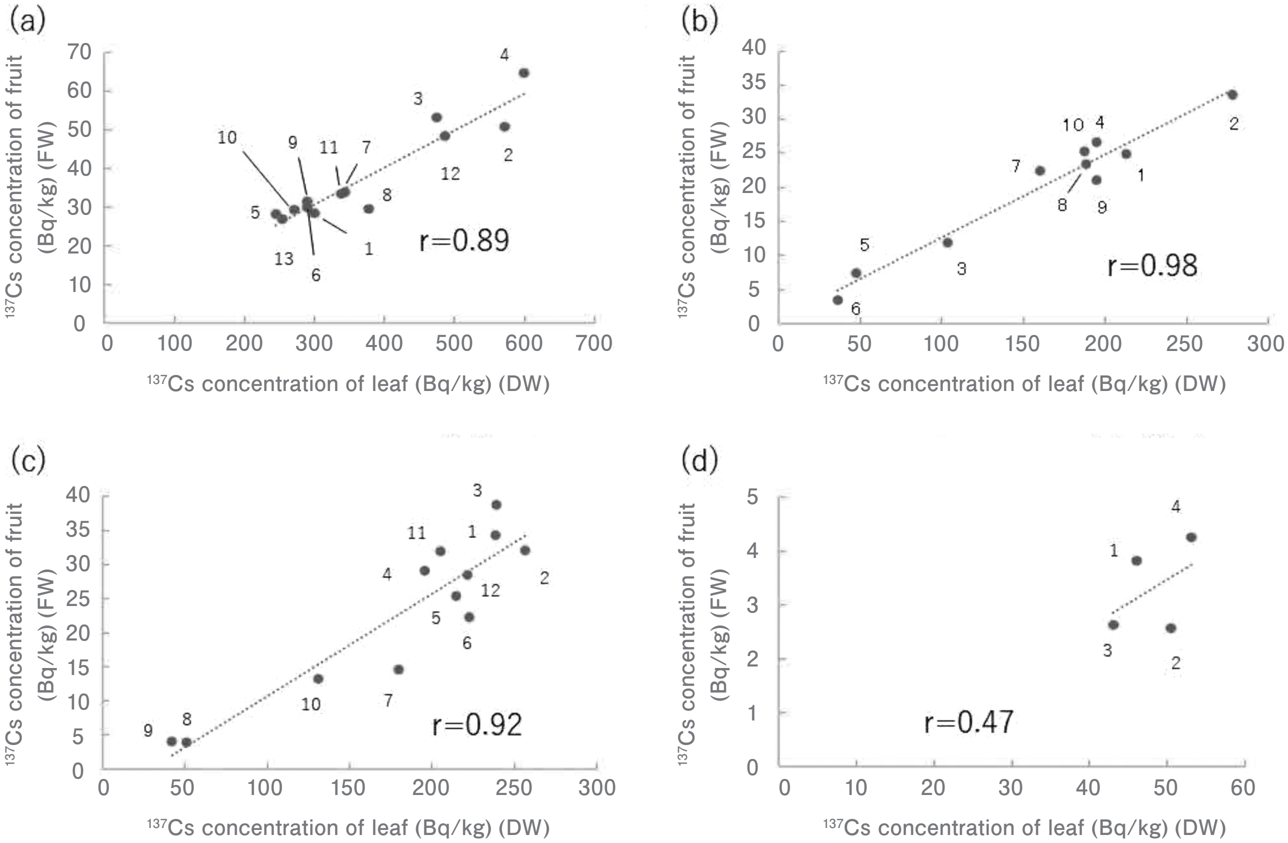

Fig. 5. Relationship between ^{137}Cs concentration of fruits and leaves

FW, fresh weight; DW, dry weight

a) Relationship between ^{137}Cs concentration of fruits and leaves in Tree A ($P < 0.01$)

b) Relationship between ^{137}Cs concentration of fruits and leaves in Tree B ($P < 0.01$)

c) Relationship between ^{137}Cs concentration of fruits and leaves in Tree C ($P < 0.01$)

d) Relationship between ^{137}Cs concentration of fruits and leaves in Tree D ($P = 0.53$)

The numbers in the graph correspond to the branch numbers in Figures 1-4.

^{137}Cs from the root to the branch was different. These multiple factors are responsible for the varying ^{137}Cs concentrations in the fruits and leaves of the different branches. Additionally, the measurement of ^{137}Cs concentration in branches seems to be important for resolving the factors that affect the variation in ^{137}Cs concentration. Hence, a more detailed investigation is required to clarify the involvement of these factors.

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