Decontamination Effects of Bark Washing with a High-pressure Washer on Peach \([Prunus persica\) (L.) Batsch\] and Japanese Persimmon \([Diospyros kaki\) Thunb.\] Contaminated with Radiocaesium during Dormancy

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The effect of bark washing with a high-pressure washer on deciduous trees contaminated during dormancy by radiocaesium fallout derived from the Fukushima Daiichi Nuclear Power Plant accident was examined using peach \([Prunus persica\) (L.) Batsch\] and Japanese persimmon \([Diospyros kaki\) Thunb.\]. Eighteen-year-old peach trees (‘Akatsuki’) were bark-washed twice with a high-pressure washer on July 5 and 27, 2011. Seven-year-old peach trees (‘Kawanakajima Hakuto’) were bark-washed on January 24, 2012, and thirty-year-old Japanese persimmon trees (‘Hachiya’) were bark-washed on December 21, 2011. For the peach trees, most of the bark was not removed by washing with a high-pressure washer. In contrast, the rough bark of Japanese persimmon was removed completely. No significant differences in the \(^{137}\text{Cs}\) concentration of ‘Akatsuki’ fruit were found between the treatments conducted in the summer of 2011. Upon the bark washing of peach ‘Akatsuki’ trees in summer, the possibility of secondary contamination of leaves via the leachate containing \(^{137}\text{Cs}\) was likely. The \(^{137}\text{Cs}\) concentrations in fruits and leaves of peach ‘Kawanakajima Hakuto’ collected in summer 2012 were decreased significantly by washing treatment conducted in winter 2011–2012. In the year after treatment, the \(^{137}\text{Cs}\) concentrations in fruits and leaves of Japanese persimmon were significantly decreased by the treatment. The effect of the bark washing on decreasing \(^{137}\text{Cs}\) contents in fruits and leaves was greater in Japanese persimmon than in peach. The results for ‘Kawanakajima Hakuto’ and ‘Hachiya’ demonstrated the possibility of additive contamination.

Key Words: contamination, Fukushima Daiichi Nuclear Power Station, high-pressure washing, migration via bark, radioactive fallout.

Introduction

Radioactive fallout from the accident at the Tokyo Electric Power Company Fukushima Daiichi Nuclear Power Plant (FDNPP) was released from March 12 to 14, 2011. This fallout contaminated all fruit production orchards in Fukushima Prefecture. Radioactive deposition was diffused primarily by rainfall and snowfall on March 15, 2011 (Imanaka, 2012).

The budding days for the main deciduous trees in the Fruit Tree Research Centre of Fukushima Agricultural Technology Centre (FTRC), Fukushima City, were March 28, 2011 for peach \([Prunus persica\) (L.) Batsch\] ‘Akatsuki’, March 28 for cherry \([Prunus avium\) (L.) L.] ‘Sato Nishiki’, March 31 for apple \([Malus pumila\) Mill.] ‘Fuji’, and April 8 for Japanese pear \([Pyrus pyrifolia\) (Burm.f.) Nakai] ‘Kosui’. Japanese persimmon \([Diospyros kaki\) Thunb.] and grape \([Vitis\) spp.] sprouted...
later than Japanese pear. Most deciduous fruit trees, except for Japanese apricot \(\textit{Prunus mume}\) (Sieb.) Sieb. et Zucc., had not developed leaves. In the Chernobyl accident (April 26, 1986), radioactive nuclides migrated inward in trees mainly via the leaves because all species of fruit tree had already budded. The current situation is therefore different from this previous accident of a similar type and scale.

\(^{137}\text{Cs}\) levels of 87 and 80 Bq kg\(^{-1}\) FW were detected in peach ‘Akatsuki’ fruit without and after washing collected on May 26, 2011 (Sato and Muramatsu, unpublished data). There were roots of undergrowth running through the topsoil at a high density because sod culture was applied in almost all orchards in Fukushima Prefecture. Therefore, radiocesium contamination by root uptake was likely negligible during the accident year (Sato et al., 2015), the same as in a previous report in relation to the Chernobyl accident (Antonopoulos-Domis et al., 1991). Deposits on the scaffold trunk of peach, cherry, apple, Japanese pear, and grape trees were recognized by measuring the radiation count rate using Geiger-Mueller survey meter at 1 cm distance from the bark surface at the top, side, and bottom sites on the scaffold limb 30 cm from a bifurcation on April 28, 2011 (Sato, 2012). The amount of radiocesium on the frame of the scaffold limb was highest at the top site and lowest at the bottom site (Sato, 2012). In addition, during winter, birch canopies were shown to intercept 20–25% of re-suspended radioactive particles (International Atomic Energy Agency, 2010a). Furthermore, Carini and Bengtsson (2001) introduced in their review an interesting experiment that was conducted by Katana et al. (1988). It showed that \(^{134}\text{Cs}\) migrated into mature apples from the bark of one two-year-old stem kept in contact with \(^{134}\text{Cs}\) solution for 10 weeks. Ono and Tagami (2003) also proved the absorption of \(^{15}\text{N}\) via the trunk bark of 8-year-old ‘Pione’ vines. \(^{15}\text{N}\) was detected at 0.69% in atom excess in the shoot approximately 3 weeks after applying solution of 10% \(^{15}\text{N}\)-urea, which contained 10.5% of \(^{15}\text{N}\) in atom excess, on the trunk after peeling with a high-pressure washer. On the basis of these previous reports and our investigation, we assumed the possibility of inward migration via the bark (Takata et al., 2012).

Decontamination of fruit trees, including peach and Japanese persimmon, was conducted by washing bark with a high-pressure washer in orchards in the northern area of Fukushima Prefecture during winter (November 2011 to March 2012) (Takata, 2013). Washing bark with a high-pressure washer is expected to be an effective method of decontamination of radioactive fallout. Unfortunately, there are no reports on the decontamination of deciduous fruit trees contaminated during dormancy in previous nuclear accidents. Even “Data sheets on countermeasure options in agricultural areas” in the EURANOS Food Handbook (EURANOS, 2009) does not describe bark washing.

In the present study, the effects of bark decontamination treatment by high-pressure washing on radiocesium concentration in fruits and leaves of peach and persimmon trees were investigated in order to examine the possibility of radiocesium migration via bark.

### Materials and Methods

**Effects of bark-washing treatments during the fruit growth period of peach tree in the year of radiocesium exposure on the radiocesium concentration in fruits and leaves**

Six 18-year-old peach ‘Akatsuki’ trees planted in FTRC, approximately 80 km from FDNPP, were used for this experiment. Trees were trained to a modified open-center system with two scaffold limbs. Pruning in summer was conducted before treatment according to the conventional method. Three trees were selected for washing and three did not undergo washing treatments. The radiation count rate was measured using a Geiger-Mueller survey meter (TGS146; Hitachi Aloka, Tokyo, Japan) at the top, side, and bottom (ground-facing) sites on two separate scaffold limbs at 1, 2, and 3 m distance from the bifurcation. Measured values were corrected by subtracting the count rate in the air at 1 m from the ground.

The primary and secondary scaffold limbs were washed with a high-pressure washer equipped a revolution nozzle (MSW1511-S; MARUYAMA Seisakusho Co. Ltd., Tokyo, Japan), at five megapascals (MPa) water pressure using 15 L of water per tree on July 5 and 27, 2011. The leachate after washing was left to drop onto the ground directly. The radiation count rate was measured at the same locations after washing, on July 7 and August 2, 2011. Two to three hundred leaves were collected on August 4, and 10 mature fruits from each tree were collected on August 10, 2011.

**Effect of bark-washing treatment at the dormant stage of peach tree following radiocesium exposure on the radiocesium concentration in fruits and leaves**

Six 7-year-old peach ‘Kawanakajima Hakuto’ trees planted in FTRC were used for this experiment. Trees were trained to a modified open-center system with two scaffold limbs. Pruning in winter was conducted before treatment according to the conventional method. The radiocesium concentration in leaves and mature fruits of six trees was measured on August 29, 2011, before any treatments. Three trees were selected for washing and three were selected for the control group (no treatment). On January 25, 2012, the primary and secondary scaffold limbs were washed using a high-pressure washer equipped with a revolution nozzle (MSW1511-S; MARUYAMA Seisakusho Co. Ltd.), at 5 MPa water pressure using 15 L of water per tree. The leachate after washing was left to drop onto the ground directly. Two to three hundred leaves and 10 to 60 fruits per tree in each of the three washed trees and three control-group
trees were collected four times (on June 19, July 13, August 13, and August 31, 2012).

**Effect of rough-bark-washing treatment on the radiocaesium concentration in fruits and leaves in Japanese persimmon**

For this experiment, six 30-year-old persimmon ‘Hachiya’ trees in an investing orchard in Date City, approximately 60 km from FDNPP, were used. Trees were trained to a modified leader system with several scaffold limbs. Pruning in winter was not performed during two years after the washing treatment. Prior to treatment, bark, fruits, and leaves were collected in the following manner to confirm the radioactive contamination level in the orchard. On November 14, 2011, 50 leaves and 10 fruits per tree were collected from five trees in the investing orchard for routine monitoring measurement. Leaves were sampled only from the non-fruit-bearing middle nodes of the terminal shoot. Old bark was also collected from five trees for monitoring measurement on October 18 and November 14.

Six trees were selected for the decontamination study. These included the five trees for monitoring measurement plus one additional tree. The leader of the main trunk of each washing treatment tree, which had several bearing shoots, was cut to approximately 2.5 m in height before washing. The radiation count rate of the primary and second scaffold limbs and main trunk of each tree was measured using a Geiger-Mueller survey meter (TGS146; Hitachi Aloka) before washing. The measuring position was at the top part on each scaffold limb at 1 and 2 m from the bifurcation and at two places diametrically opposite to each other, 1 m from the ground on the trunk.

The surfaces of scaffold limbs and the trunk were washed with a revolution nozzle-equipped high-pressure washer at 10 MPa water pressure using 100 L of water per tree on December 21, 2011. The leachate after washing was caught using a nonwoven fabric sheet (Oji Kinocloth, Shizuoka, Japan) containing zeolite powder spread on the ground. The count rate of radiation at approximately the same places was also measured after washing on December 28, 2011. In all measurements, measured values were corrected by subtracting the count rate in the air at one meter from the ground.

Sixty leaves and six to 20 fruits per tree in every treatment were collected four times (on July 24, August 23, September 20, and October 22, 2012). In the same way, samples were taken on July 19, August 19, September 18, and October 28, 2013. In all cases, leaves were sampled only from the non-fruit-bearing middle nodes of the terminal shoot. The bark of the three trees in each treatment group was collected on October 28, 2013, to compare the concentration of radiocaesium with measured values in the bark collected on October 18, 2011, prior to washing.

In 2012, three trees decontaminated by washing and cutting back of the leader trunk grown in the same season as the decontamination study samples were added to the radiocaesium measurement of fruits and leaves. Although fruits, leaves, and old bark collected before treatment were combined into a single sample for each organ, fruits and leaves collected in 2012 and 2013 and bark collected in 2013 were prepared for each tree.

An exponential equation expressed as a predictor variable of the temporal changes in the radiocaesium concentration in the leaves and fruits during the year after the nuclear accident (Antonopoulos-Domis et al., 1991) was obtained by the least-squares method adopted with the quasi-Newton method:

$$Y = K \exp(-Dx)$$

where Y is the radiocaesium concentration in the fruits and leaves produced 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 (predictor variable); and D is the decay constant. The effects of temporal decay were evaluated by the decay constant. Furthermore, an equation modeling the decontamination effects was obtained representing the concentration ratio of the washing treatment to that in the control group.

This equation used data from samples collected on November 14, 2011, of six washed trees, including three experimental trees and three unwashed trees in 2012, and three trees of each treatment group in 2013.

**Sample treatments and radiocaesium measurements**

Fruits were shredded using a food processor after peeling and removing peach stones and Japanese persimmon seeds and calyxes. Bark, branches, and moss were cut into pieces with pruning scissors without dehydration treatment. Leaves and pulp were placed in U-8 pots after freeze-drying for 72 hours or more. The radiocaesium concentrations in leaves and pulp were converted to wet weight after freeze-drying using the dry-to-wet ratio. The $^{134}$Cs and $^{137}$Cs concentrations in the samples were analyzed using germanium (Ge) semiconductor detector systems at the Research Center for Electron Photon Science of Tohoku University and at the Faculty of Science of Gakushuin University. The counting time for each sample was about 1 to 24 h, depending on the activity level. Measurements were carried out within 14 days after sampling. Measured activity in each leaf, fruit, bark, and branch was corrected back to the sampling day using a decay compensation formula corresponding to the physical half-life of the measured radioisotope.
Results

Difference between peach trees and Japanese persimmon trees in the effects of high-pressure bark-washing treatment and bark removal

For the peach trees, most of the bark was not removed by washing with a high-pressure washer. In contrast, the rough bark of Japanese persimmon was removed completely by high-pressure washing (Fig. 1).

Effects of bark-washing treatments during fruit growth in the year of exposure on the radiocaesium concentration in fruits and leaves of peach tree

An apparent difference was observed among the top, side, and bottom positions along the circumferential direction on the scaffold limb. The radiation count rates measured on the scaffold trunk were 1.81 kcpm at the sky-facing site, 0.95 kcpm at the side site, and 0.54 kcpm at the bottom site (Table 1). Prior to bark-washing treatment, the radiation count rates on the scaffold limbs were 1.82 and 1.81 kcpm in trees assigned to the washing treatment and control trees, respectively. It significantly decreased to 0.96 kcpm two days after the first washing treatment, and to 0.80 kcpm six days after the second washing treatment; namely, these two washing treatments resulted in 46.6% and 55.9% decreases, respectively, in the radiation count rate (Table 2). No significant difference in the $^{137}$Cs concentration in fruits was observed at harvest between the washed and control trees. However, the interaction between treatments in the measured values in leaves showed a significant difference (Table 3). The value in 2012 was lower in the washed trees than in the control trees at 5% by Tukey’s test.

Effect of the rough-bark-washing treatment on the radiocaesium concentration in Japanese persimmon fruits and leaves

Prior to treatment, the $^{137}$Cs concentrations in leaves and mature fruits of trees assigned to the washing treatment collected on August 29, 2011, were 67.3 and 11.0 Bq·kg$^{-1}$ FW, while the concentrations in leaves and mature fruits of the control trees were 68.5 and 13.3 Bq·kg$^{-1}$ FW, respectively. There was no significant difference between the treatment and control groups by t-test. In 2012, the $^{137}$Cs concentrations in the leaves and fruits in the following developing period after the washing treatment were significantly decreased by the washing treatment, with significant differences between different sampling days (Table 4). On June 19, 2012, when shoots were developing rapidly, the values measured in the control leaves were approximately twofold those in the washing-treatment group. The $^{137}$Cs concentration in the fruits was decreased by half at the harvest time of August 31 from that on June 19. The difference in the $^{137}$Cs concentrations in leaves and fruits between treatments decreased from July 13 to August 31. The $^{137}$Cs concentration in fruits in the washing treatment on June 19 was significantly higher than that on August 31, but no significant difference was found in the control at 5% by Dunnett’s test. The $^{137}$Cs concentration in leaves in the control on June 19 was significantly higher than that on the later sampling days, but no significant difference was found between them after July 13. No significant difference was found between measured values in leaves on every sampling day in the washing treatment at 5% by Tukey’s test.

Effect of the bark-washing treatment of peach tree at the dormant stage following radiation exposure on the radiocaesium concentration in fruits and leaves

Prior to treatment, the $^{137}$Cs concentrations in leaves and mature fruits of trees assigned to the washing treatment collected on August 29, 2011, were 67.3 and 11.0 Bq·kg$^{-1}$ FW, while the concentrations in leaves and mature fruits of the control trees were 68.5 and 13.3 Bq·kg$^{-1}$ FW, respectively. There was no significant difference between the treatment and control groups by t-test. In 2012, the $^{137}$Cs concentrations in the leaves and fruits in the following developing period after the washing treatment were significantly decreased by the washing treatment, with significant differences between different sampling days (Table 4). On June 19, 2012, when shoots were developing rapidly, the values measured in the control leaves were approximately twofold those in the washing-treatment group. The $^{137}$Cs concentration in the fruits was decreased by half at the harvest time of August 31 from that on June 19. The difference in the $^{137}$Cs concentrations in leaves and fruits between treatments decreased from July 13 to August 31. The $^{137}$Cs concentration in fruits in the washing treatment on June 19 was significantly higher than that on August 31, but no significant difference was found in the control at 5% by Dunnett’s test. The $^{137}$Cs concentration in leaves in the control on June 19 was significantly higher than that on the later sampling days, but no significant difference was found between them after July 13. No significant difference was found between measured values in leaves on every sampling day in the washing treatment at 5% by Tukey’s test.

Table 1. Comparison of radiation count rate on bark of ‘Akatsuki’ peach prior to washing treatment by the factor of location.

| Treatment          | Radiation count rate (kcpm)$^a$ |
|--------------------|---------------------------------|
|                    | Top    | Side  | Bottom |
| Bark-washing       | 1.82 ±0.13$^c$ | 0.93 ±0.17 | 0.51 ±0.11 |
| Control (without washing) | 1.81 ±0.08 | 0.96 ±0.18 | 0.57 ±0.01 |
| Average            | 1.81 ±0.08 | 0.95 b  | 0.54 a  |

$^a$ Measured at top, side, and bottom sites along the circumference on the first and second scaffold limbs at 1, 2, and 3 m from a bifurcation with TGS146 (Hitachi Aloka) before bark washing on July 5, 2011. The measured values were corrected by subtracting the count rate in air at 1 m above the ground.

$^c$ Mean ± SD (n = 3).

$^d$ Different letters indicate significant differences at $P < 0.05$ by Tukey’s test (n = 6).

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Fig. 1. (A) Bark of peach prior to washing. (B) Bark of peach after washing with a high-pressure washer at 5 MPa water pressure. (C) Bark of Japanese persimmon prior to washing. (D) Bark of Japanese persimmon after washing with a high-pressure washer at 10 MPa water pressure.
treatment, and 3.19 and 3.91 kcpm, for the control trees, respectively. Measured values on the main trunk of trees assigned to the washing treatment and control trees were 0.58 and 0.35 kcpm, respectively. ANOVA revealed no significant differences in the radiation count rates between the treatments. However, the values measured after treatment were significantly different. For example, measurements from the scaffold limb at 1 and 2 m from the bifurcation were 0.29 and 0.33 kcpm, on the washed trees, and those on the control were 2.94 and 3.81, while the values on the trunk were 0.12 and 0.41 kcpm, respectively (Table 5). On the scaffold limb at 1 and 2 m from the bifurcation, the radiation count rates after the washing treatment decreased by 87.5% and 85.5%, respectively, and by 63.1% on the trunk, compared with the values measured prior to the treatment. However, the radiation count rate on the trunk of the control trees increased by 0.6 kcpm during the seven days between samplings. In 2013, the $^{137}$Cs concentrations in the bark were 30,400 Bq·kg$^{-1}$ FW for the control trees and 22,50 Bq·kg$^{-1}$ FW for the washed trees. The $^{137}$Cs concentrations were 188.0 and 74.8 Bq·kg$^{-1}$ FW in leaves and fruits on November 14, 2011. A significant difference in the $^{137}$Cs concentrations in leaves and fruits was recognized between the washed and control trees in 2012 (Table 6). The $^{137}$Cs concentrations in fruits and leaves decreased according to the number of days after full blossom (Table 6). The $^{137}$Cs concentra-

### Table 2. Decreasing radiation count rate at the top of the scaffold limb in ‘Akatsuki’ peach after bark washing in the year 2011.

| Treatment          | Replicates (Tree) | Distance | Radiation count rate (kcpm) | Decreasing rate (%) |
|--------------------|-------------------|----------|----------------------------|---------------------|
|                    |                   | Before  | After 1st washing | After 2nd washing  |
|                    |                   | July 5, 2011 | July 7, 2011 | Aug. 2, 2011 | Before 1st washing | After 2nd washing |
| Washing            | 3                 | 1 m     | 1.95               | 0.76               | 0.80               | 61.0               | 59.0               |
|                    |                   | 2 m     | 1.70               | 1.11               | 0.65               | 34.7               | 61.8               |
|                    |                   | 3 m     | 1.81               | 1.01               | 0.96               | 44.2               | 47.0               |
|                    |                   | Average | 1.82               | 0.96               | 0.80               | 46.6               | 55.9               |
| No washing (Control) | 3              | 1 m     | 1.88               | 1.88               | 1.89               | 0.0                | 0.5                |
|                    |                   | 2 m     | 1.84               | 1.84               | 1.83               | 0.0                | 0.5                |
|                    |                   | 3 m     | 1.71               | 1.71               | 1.70               | 0.0                | 0.6                |
|                    |                   | Average | 1.81               | 1.81               | 1.81               | 0.0                | 0.2                |

Z\ Measured at the top part on the first and second scaffold trunks at 1, 2, and 3 m from a bifurcation before (July 5) and after the 1st (July 7) and 2nd (July 27) washing. The measured values were corrected by subtracting the count rate in the air at 1 m above the ground. The surfaces of scaffold and secondary scaffold limb were washed using a high-pressure washer at 5 MPa water pressure using 15 L of water per tree twice (July 5 and July 27, 2011).

\ Decreasing rate (D) calculated using the following equation; D = 100(B – A)·B – 1, where B and A indicate the radiation count rate before and after washing treatment.

\ Two-way analysis of variance with replication was conducted for the main effect of the washing treatment and the measured distance.

\*** and NS indicate significance at $P<0.001$ and non-significance, respectively.

\ Not analyzed.

### Table 3. Comparison of temporal changes in the $^{137}$Cs concentration in leaves and fruits of ‘Akatsuki’ peach after bark washing in the year 2011.

| Sampling day | Replicates (Tree) | Fruits | Leaves |
|--------------|-------------------|--------|--------|
|              |                   | Control | Washed | Control | Washed |
| Aug. 10, 2011 | 3                 | 18.3    | 19.6    | 114.1 c | 151.2 c |
| Aug. 10, 2012 | 3                 | 5.6     | 5.5     | 57.2 b  | 32.1 a  |

Z Bark washing treatment with a high-pressure washer was performed on July 5 and July 27, 2011. Decay compensation was calculated back to the sampling day.

\ Two-way analysis of variance with replication was conducted for the main effect of the bark-washing treatment and the sampling day.

\ ***, **, and NS indicate significance at $P<0.001$, 0.01, and non-significance, respectively.

\ Different letters indicate significant differences regardless of treatment at $P<0.05$ by Tukey’s test ($n=3$).
tions in fruits and leaves in the washed trees on July 24 were significantly higher than that on September 20 and October 22. In the control trees, a significant difference was found only in the measured value of fruits between July 24 and October 22 at 5% by Dunnett’s test. In 2013, the same significant effects of the washing treatment on the $^{137}$Cs concentrations in leaves and fruits on October 28 were found, namely, 66.5 and 35.6 Bq·kg$^{-1}$ FW in leaves, and 22.4 and 13.9 Bq·kg$^{-1}$ FW in fruits in control and washed trees, respectively (Sato, 2014).

The least-squares method was used to obtain a significant exponential equation for each year after the deposition of radioactive material. The deduced decay constants of 1.19 in fruits and 1.22 in leaves in the washing treatment group were lower than those in the control group, which were 0.846 for fruits and 0.817 for leaves (Fig. 2). The equation representing decontamination effects was deduced as $1.0 \exp(-0.344)$ for fruits and $1.0 \exp(-0.403)$ for leaves.

### Discussion

It is inferred that radiocaesium migrates inward from the bark surface via plant organs (Momoshima and Bondietti, 1994). Radiocaesium was detected only in the vicinity of the lenticels by analyzing the imaging plates on the bark of ‘Akatsuki’ collected on June 16, 2011 (Takata, 2013). Complementary cells developing around lenticels are considered to play an important role in the gaseous exchange in the inner tissues of the tree. Furthermore, the ray tissue passes through the phloem and wood from periderm to heartwood.

The radiation count rate on the bark was higher at the top site, regardless of the tree or the frame of the limb (Sato, 2012), which indicates that the deposition of...
radiocaesium-contaminated rainfall on the bark surface after the nuclear accident is the migration source into the deciduous trees before sprouting. Actually, the radiocaesium concentration in the bark of grape and Japanese pear showed the same tendency in terms of position for the radiation count rate (Sato, 2012), which indicates that the radiation count rate reflects the level of radiocaesium accumulation. These results are consistent with a previous report stating that birch canopies intercept resuspended radioactive particles during winter (International Atomic Energy Agency, 2010a). The rough bark of the Japanese persimmon was removed efficiently with a high-pressure washer, equipped with a revolution nozzle at 10 MPa water pressure (Fig. 1). The radiocaesium contamination in Japanese persimmon was removed easily by high-pressure washing because of the removal of the rough bark. However, the peach tree does not form rough bark and is wounded by washing at pressure higher than 6 MPa (Fig. 1). Although there was a certain effect by the removal of radiocaesium from the surface of peach canopy by high-pressure washing, there were no significant differences in the $^{137}$Cs concentration between treatments in 18-year-old ‘Akatsuki’ fruits harvested on August 10, 2011 (Table 3). This suggests that the washing treatment has no decontamination effects on the $^{137}$Cs content in fruits. However, the $^{137}$Cs concentration in leaves in the year after treatments differed significantly between the bark-washing and control trees (Table 3). Furthermore, the $^{137}$Cs concentration in peach ‘Kawanakajima Hakuto’ fruits harvested on August 31, 2012, was decreased significantly by the washing treatment in January 24, 2012 (Table 4). These contrasting results suggest that further studies are needed to evaluate the effect of bark washing to control the inward migration of radiocaesium. $^{137}$Cs of $239 \text{ Bq}\cdot\text{kg}^{-1}$ FW was detected in the leaves from a 12-year-old ‘Akatsuki’ tree collected on May 20, 2011 (Sato and Muramatsu, unpublished data). Furthermore, the $^{137}$Cs concentrations in bark at the top part of the scaffold limb, which was collected on June 16, 2011, in one of the trees assigned to the washing treatment in our experiment were

| Sampling day  | Replicates (Tree) | Days after full blossom | $^{137}$Cs concentration (Bq·kg$^{-1}$ FW)$^z$ |  |
|---------------|-------------------|-------------------------|---------------------------------------------|---|
|               |                   |                         | Fruits                                      | Leaves |
|               |                   |                         | Control                                     | Washed  |
|               |                   |                         | Control                                     | Washed  |
| July 24, 2012 | 3                 | 39                      | 57.5 b$^x$                                  | 44.8 b  |
| Aug. 23, 2012 | 3                 | 69                      | 47.2 b                                      | 30.9 b  |
| Sep. 20, 2012 | 3                 | 97                      | 34.6 b                                      | 24.1 a  |
| Oct. 22, 2012 | 3                 | 129                     | 30.1 a                                      | 19.5 a  |

Significance by ANOVA$^y$

- Treatment: ***
- Sampling day: ***
- Interaction: NS

$^z$ Decay compensation was performed back to the sampling day. Washing treatment with a high-pressure washer was performed on Dec. 21, 2011.

$^y$ Two-way analysis of variance with replication was conducted for the main effect of the washing treatment and the sampling day.

$^x$ Different letters indicate significant differences within the same treatment in fruits or leaves at $P<0.05$ by Dunnett’s test ($n=3$).

$^w$ ***, **, and NS indicate significance at $P<0.001$, 0.01, and non-significance, respectively.

**Fig. 2.** Comparison of the exponential equation model of temporal changes in $^{137}$Cs concentration in Japanese persimmon fruits (A) and leaves (B) by bark-washing decontamination. The equation was deduced by the least-squares method, using fruit (A) and leaf (B) data at harvest. Data for 2011 were from one sample combining five trees prior to washing. Data for 2012 were from three control trees and six washed trees. Data for 2013 were from three trees of each treatment in 2013 in Japanese persimmon ‘Hachiya’. * and ** indicate significant difference between treatments at $P<0.05$ and 0.01 by $t$-test.
36800 Bq·kg\(^{-1}\) FW in the surface of the bark and 760 Bq·kg\(^{-1}\) FW in the cortex including secondary phloem with cambium (Sato, 2012). The possibility of root uptake of \(^{137}\)Cs was supposed to be extremely low in consideration of the interaction between the underground roots and the soil profile of \(^{137}\)Cs (Sato, 2014; Sato et al., 2015). It was therefore estimated that a large amount of radiocaesium inward migration via the bark had already occurred before the washing treatment was conducted. Although the findings in ‘Akatsuki’ in the corresponding stage of washing treatments suggested that the decontamination of the bark in July or later was already too late to inhibit radiocaesium migration, the result in the leaves of ‘Akatsuki’ and in both fruits and leaves of ‘Kawanakajima Hakuto’ in 2012 for the next developing season after washing contradicted this hypothesis.

Another reasonable viewpoint, which does not conflict with these results, is that secondary contamination occurred from the treatments. When treatments are conducted, the translocation from leaves to fruits may occur. It is likely that the contaminated liquid that scattered when the bark was washed adhered to the leaves and fruits, and thus became a source of radiocaesium transfer into the fruits. In other words, secondary contamination masked the effect of decreasing the radiocaesium concentration in mature fruits by bark washing. Table 2 shows that 46.6% of radiocaesium was removed by washing. If half of the 36800 Bq·kg\(^{-1}\) FW of \(^{137}\)Cs (Sato, 2012) was scattered by 15 L of water, the influence of secondary contamination to leaves was unlikely to be negligible. Furthermore, the result in ‘Kawanakajima Hakuto’ and the leaves of ‘Akatsuki’ in the next developing year after washing showed that additional contamination by residual radiocaesium on the bark occurred in the year following the accident at FDNPP.

Although the average radiation count rate in the scaffold limbs of the control trees prior to the washing treatment in an examination on Japanese persimmon was 0.91 kcpm higher than that of treated trees (Table 5), no significant difference of radiation count rate for the limb and the trunk between treatments and controls was recognized by ANOVA. There were more than 2.0 kcpm differences in the radiation count rate for the scaffold limb at different measuring sites in the same tree. In fact, the average radiation count rate ± standard deviation at 1 m from a bifurcation for another scaffold limb of the control trees was 2.26 ± 0.19 kcpm. Therefore, the results of ANOVA were acceptable. These results show that the dispersion of the contamination level on the canopy of Japanese persimmon varied widely compared with that of peach (Tables 2 and 5). There are several concave and convex structures on the surface of the rough bark of Japanese persimmon. Substantial radiocaesium remains in the concave part on the surface, but radiocaesium on the convex part can easily flow down during the rainfall. It is also clear that radiocaesium had already migrated inward before washing, because \(^{137}\)Cs ranging from 213 to 724 Bq·kg\(^{-1}\) FW was detected at the wood tissue of branches collected on October 18, 211 (Sato, 2012). A significant decrease in radiocaesium concentration by washing treatment on December 21, 2011, was observed in fruits of Japanese persimmon ‘Hachiya’ collected from the investigated local orchard two years after the nuclear accident (Fig. 2). In 2011, the \(^{137}\)Cs concentrations in the trunk bark were 49600 Bq·kg\(^{-1}\) FW prior to the washing treatment (Sato et al., 2012). In 2013, the \(^{137}\)Cs concentrations in the bark were 30400 Bq·kg\(^{-1}\) FW for the control trees and 2250 Bq·kg\(^{-1}\) FW for the washed trees. Although the \(^{137}\)Cs concentration in the bark of the control trees decreased to 61.3% compared with the measured values of the bark prior to the washing treatment in 2011, it was still 13.5-fold higher than the concentration in the washed trees. A number of fine cracks on the outer bark surface of Japanese persimmon retain raindrops and enable moss to settle. Epiphytic moss had actually been flourishing on the bark in the control-group trees. Moss is known to accumulate radiocaesium, because of its slow growth (International Atomic Energy Agency, 2010b). The increase in radiation count rate of 0.6 kcpm in the trunks of the control trees from December 21 to December 28 demonstrated that additional contamination by downward flow from the upper part of canopy likely occurred (Table 5). The epiphytic moss on the bark of the Japanese persimmon ‘Hachiya’ tree collected on November 14, 2011, contained 217000 Bq·kg\(^{-1}\) FW of \(^{137}\)Cs, which was four times or more higher than the 49600 Bq·kg\(^{-1}\) FW of the trunk bark collected on October 18, 2011 (Sato et al., 2012). Moss is likely to be one of the reservoirs of radiocaesium because radiocaesium attaches to its abundant surface areas and it releases soluble radiocaesium easily after dying. Sato et al. (2012) deduced from an investigation in the same orchard that radiocaesium accumulated on the moss from bark because the moss had a higher (\(^{134}\)Cs + \(^{137}\)Cs) to \(^{110}\)mAg ratio using a \(^{110}\)mAg count of 706 keV than the bark of Japanese persimmon. Since \(^{110}\)mAg, which is one of the radioactive substances released in this nuclear accident, is hardly moved by rainfall, the (\(^{134}\)Cs + \(^{137}\)Cs) to \(^{110}\)mAg ratio could be taken as an indicator of the accumulation of radiocaesium. Furthermore, secondary contamination may occur because of higher \(^{137}\)Cs concentrations in leaves, calyces, and peels prepared by washing, rather than in those prepared without washing (Sato et al., 2012). These results indicate that additional contamination by residual radiocaesium on the bark continued at least in the unwashed persimmon trees two years after initial deposition.

Kikunaga (personal communication, 2013) proposed the following equations to evaluate the effects of bark washing during dormancy on additive radiocaesium
migration:

\[ C_{uw} = C_w + \Delta C \]

where \( C_{uw}, C_w, \) and \( \Delta C \) represent the radiocaesium concentrations in unwashed and washed trees, and additive radiocaesium migration in 2012, respectively. If \( \Delta C = r \times C_{uw}, \) \( r \) is defined as the absorption coefficient via the bark, \( C_w = C_{uw} \times (1 - r) \) and \( (1 - r) = C_w \times C_{uw}^{-1}. \)

From the exponential model equation, when defining \( (1 - r) \) as the washing effect \( E_w, \) the following equation can be obtained:

\[ E_w = (1 - r) = C_w \times C_{uw}^{-1} \]

\[ = K_w \times K_{uw}^{-1} \times \exp\{- (D_w - D_{uw}) \times x\} \]

(1)

where \( K_w \) and \( K_{uw} \) are the radiocaesium concentrations in washed and unwashed trees in the year of the nuclear accident (in our examination, \( K_w = K_{uw} \)), and \( D_w \) and \( D_{uw} \) are decay constants of washed and unwashed trees, respectively. Equation (1) shows that the effects of washing on additive radiocaesium migration can be expressed by an exponential equation. In our study, \( 100(1 - E_w) \) is defined as decreasing effects.

Decreasing effects of the washing decontamination were calculated at 29.1% in fruits and 33.2% in leaves by equation (1), suggesting that decontamination treatments using a high-pressure washer carried out during November 2011 to March 2012 in the production area of deciduous trees were sufficiently effective to prevent additive radiocaesium migration via the bark in the Japanese persimmon. The radiation count rate decreased by 46.6% in peach (Table 2) and 86.5% in Japanese persimmon (Table 5) by one washing treatment. The lower decrease in the radiation count rate in the washed peach tree compared with that in the washed Japanese persimmon was probably because of the removal of the rough bark (Fig. 1). Additional washing treatments are also considered to be effective in older trees, which have considerably rough bark.

Higher \(^{137}\text{Cs}\) concentrations in fruits of ‘Kawanakajima Hakuto’ peach and ‘Hachiya’ persimmon were detected in the earlier stage in the washing treatment trees. The \(^{137}\text{Cs}\) concentrations in leaves in the earlier stage were higher in the control trees of ‘Kawanakajima Hakuto’ peach and in the washed trees of ‘Hachiya’ persimmon. At an early stage, the \(^{137}\text{Cs}\) in fruits may be transported from the storage organ (Momoshima and Bondietti, 1994). Small fruit volume may result in high \(^{137}\text{Cs}\) content at an early stage. On the other hand, the \(^{137}\text{Cs}\) concentration in fruits decreases by the dilution effect of assimilates transported into fruits in the ripening stage (Sato et al., 2015). In addition, the effects of bark washing on decreasing the \(^{137}\text{Cs}\) content in leaves likely affects the \(^{137}\text{Cs}\) content in the matured fruits due to the translocation of \(^{137}\text{Cs}\) from leaves to fruits in the ripening stage (Carini and Bengtsson, 2001). In this regard, the decrease in \(^{137}\text{Cs}\) concentration with days after full blossom in fruits in the washed trees was reasonable. Leaves play an important role in the movement of \(^{137}\text{Cs}\). As soon as the \(^{137}\text{Cs}\) accumulates in the leaf “in which the original absorption took place, it translocates to other sites such as leaves, stems, roots, or is distributed within the entire plant” (Koranda and Robison, 1978). Additive contamination on leaves in peach may occur at an early stage in the control trees of ‘Kawanakajima Hakuto’ peach. Since leaves of Japanese persimmon are still young in the rainy season because of sprouting approximately one month later than peach, the \(^{137}\text{Cs}\) in raindrops or downstream from the tree canopy may be absorbed via leaves easily. Additive contamination in summer may be due to the retention of \(^{137}\text{Cs}\) in leaves during the growing season in the control trees in ‘Hachiya’ persimmon.

Incidentally, in this study, only Japanese persimmon trees assigned to the washing treatment were cut to approximately 2.5 m in height. Because radiocaesium migrated into the trees, tree trunks were considered to be reservoirs for contamination (Takata, 2013; Takata et al., 2012); if more than 2.5 m of the upper part was removed, the contained radiocaesium was also removed, which could cause radiocaesium reduction in the following years. Therefore, the decrease in radiocaesium concentration in fruits and leaves may not have been due to the washing treatment alone. Takata (2013) stated that the radiocaesium concentration in the wood of peach tree decreased dramatically over the fruit growth period, indicating that the wood part is the main reservoir of radiocaesium for re-translocation following the year of exposure. Sato et al. (2014) demonstrated that only 18.5% of all radiocaesium inventories in the wood in a 8-year-old Japanese persimmon tree of 4.45 m in height existed in the upper part (higher than 1.5 m from the ground) in the year of the accident. Furthermore, it is probable that the cut-down treatments would result in increasing the radiocaesium concentration in fruits and leaves in the following year because of decreasing the sink size for re-translocation of the radiocaesium reserves. Therefore, the influence of the cut-down trunk was likely negligible. On the basis of these results in our examination, it was found that the decontamination with high-pressure bark washing was effective for decreasing the radiocaesium concentration in fruits or leaves in both peach and Japanese persimmon trees in the year after the bark washing. Japanese persimmon ‘Hachiya’ is sold as the subsequently developing fruit, namely, ‘Anpogaki’, after air-drying the peeled fruit in winter. Since the \(^{137}\text{Cs}\) concentration of ‘Anpogaki’ is approximately 3.5-fold higher than that of raw fruit, the cost-effectiveness of the bark washing is considered to be higher in Japanese persimmon ‘Hachiya’ than in other deciduous fruit trees for which the fruit is sold fresh.

We conclude that prompt bark washing before sprouting is effective for the decontamination of radio-
caesium deposited on deciduous fruit trees during dormancy and reducing the risk of re-deposition on new plant parts in the year after the nuclear accident.

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