

Radioactive cesium contamination of edible wild plants after the accident at the Fukushima Daiichi Nuclear Power Plant

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1. Introduction

Japanese citizens eat a wide variety of edible wild plants (Ikeda, 1984). Its traits are strong in snowy district. Eastern Japan including Fukushima Prefecture is one of the areas with heavy snow regions. Radioactive contamination of edible wild plants has been reported at sites in eastern Japan following the 2011 accident at the Fukushima Daiichi Nuclear Power Plant. Because the spatial and temporal contamination trends remain unclear, only preliminary guidelines for contamination of edible wild plants were available (Kiyono and Akama, 2013) to protect peoples who live in Japan. Efforts were therefore required to develop such guidelines based on scientific data. In 2012, the Special Forest Products Office of Japan's Forestry Agency compiled the test results for radioactive cesium in farm products and edible wild plants (http://www.maff.go.jp/j/kanbo/joho/saigai/s_chosa/index.html). High levels of radioactive cesium contamination were found in *Eleutherococcus sciadophylloides* (*koshiabura*) in the spring of 2012, with a maximum dose of 2,900 Bq kg⁻¹ (fresh weight basis; "FW" henceforth) and 11 of 14 samples had a dose greater than 1,000 Bq kg⁻¹ FW on 20 September 2012. The results of a test in the spring of 2013 are still being compiled, but doses of 11,900 Bq kg⁻¹ FW were reported for some samples of *Eleutherococcus sciadophylloides* (Fukushima Prefecture, 2013). Hence, the status of radioactive contamination is inadequately documented and may change.

Under the "New Standard Limits for Radionuclides in Foods" guidelines established by the Department of Food Safety, Pharmaceutical and Food Safety Bureau, Ministry of Health, Labour and Welfare (http://www.mhlw.go.jp/english/topics/2011eq/dl/new_standard.pdf) in April 2012, 100 Bq kg⁻¹ FW (low materials) is the threshold for unacceptably high contamination in general foods. Consumption of edible wild plants with a dose of 100 Bq kg⁻¹ FW or higher is controlled. Some producers and vendors voluntarily controlled contamination in their shipments by setting standards lower than those established by the government. On the other hand, personal (non-commercial) harvesting of

edible wild plants has not been controlled.

Kiyono and Akama (2013) developed draft guidelines to minimize the risk of collecting edible wild plants that are highly contaminated with radioactive cesium based on their sampling of 30 edible portions in 15 species of edible wild plants collected in May 2012 in Kawauchi and Otama Villages in Fukushima Prefecture. Because radioactive contamination of edible wild plants may change as radioactive cesium moves through an ecosystem, these guidelines should be frequently updated to reflect these movements.

To provide more data on these movements, we collected 82 samples of 20 species of edible wild plants in forests in Kawauchi and Otama Villages and Koriyama City in Fukushima Prefecture, in the spring of 2013, measured current levels of radioactive ¹³⁴Cs and ¹³⁷Cs contamination, and investigated the factors that affected the doses from these isotopes in edible wild plants. We also analyzed the temporal changes in contamination between the spring of 2012 and the spring of 2013. Based on the results, we have revised our previous draft guidelines (Kiyono and Akama, 2013) to further reduce the risk of collecting highly contaminated edible wild plants.

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2. Study site and methods

The study site has a temperate climate. The mean annual rainfall was 1,465 mm and the mean annual temperature

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was 10.3°C in Kawauchi Village and 1,163 mm and 12.1°C in Koriyama City in 1981–2010 (Japan Meteorological Agency, 2013). For Otama Village, no data were available. However, Otama Village may have a similar climatic condition to Koriyama City. The land-use types can be broadly grouped into three classes: (1) natural forest, in which deciduous broadleaf trees such as *Quercus* spp. and *Castanea crenata* are dominant; (2) planted forest of *Cryptomeria japonica* and *Pinus densiflora*, and other conifers; (3) agricultural land (e.g., irrigated rice fields, upland crop fields) and pasture land.

Part of the edible wild plants is grown on agricultural land. They were excluded in our study. We collected a total of 82 samples of the edible portions of 19 edible wild plants and 1 medicinal herb from 28 April to 14 May 2013 along the forest roads and main paths in and around the permanent sample plots set by FFPRI for monitoring radioactive contaminations of forest in Kawauchi Village (Otsupe, 37°16'32.1"N to 37°17'17.9", 140°48'15.8"E to 140°47'48.2"E, 590–730 m asl; and Togenomori, 37°22'52.02"N, 140°42'56.69"E, 693 m asl), Otama Village (Tamai-Maegatake, 37°34'4.0"N to 37°34'27.42"N, 140°18'20.8"E to 140°18'15.82"E, 730–830 m asl), and Koriyama City (Tadano, 37°23'02.8"N to 37°22'48.7"N, 140°14'3.4"E to 140°14'46.0"E, 365–368 m asl), about 25 km south-west, 65 km east, and 75 km east of the Fukushima Daiichi Nuclear Power Plant respectively. Surface geology (Geological Survey of Japan, AIST, 2012) of the sites where we collected the plant samples was classified as granites in Kawauchi Village, volcanic detritus in Otama Village, and sedimentary rocks in Koriyama City. The plant species (collected organs) were *Fallopia japonica* (*itadori* in Japanese) (buds, leaves, and stems), *Schizophragma hydrangeoides* (*iwagarami*) (buds, leaves, and current year branches and stems), *Aralia cordata* (*udo*) (buds, leaves, and stems), *Coptis japonica* (*ouren*, a medicinal herb) (buds, leaves, and inflorescence), *Erythronium japonicum* (*katakuri*) (leaves and inflorescence), *Matteuccia struthiopteris* (*kusasotetsu*) (fronds), *Eleutherococcus sciadophylloides* (buds, leaves, and current year branches and stems), *Aster microcephalus* var. *ovatus* (*nokongiku*) (buds, leaves, and stems), *Osmunda japonica* (*zenmai*) (fronds without pinnae), *Aralia elata* (*taranoki*) (buds, leaves, and current year branches and stems), *Hydrangea petiolaris* (*tsuruajisai*) (buds, leaves, and current year branches and stems), *Sambucus racemosa* subsp. *sieboldiana* (*niwatoko*) (buds, leaves, inflorescence, and current year branches and stems), *Helwingia japonica* (*hanai-kada*) (buds, leaves, inflorescence, and current year branches and stems), *Kalopanax septemlobus* (*harigiri*) (buds, leaves, and current year branches and stems), *Petasites japonicus* (*fuki*) (leaves), *Syneilesis palmata* (*yaburegasa*) (leaves), *Osmundastrum cinnamomeum* var. *fokiense* (*yamadorizenmai*) (fronds without pinnae), *Artemisia indica* var. *maximowiczii* (*yomogi*) (buds, leaves, and stem), *Clethra barbinervis* (*ryobu*) (buds, leaves, and current year branches and stems), and *Pteridium aquilinum* (*warabi*) (fronds). The air dose at a height of 1 m was measured at

every site where we collected the plant samples using a scintillation survey meter (ALOKA γ -RAY SCIENT. SURVEY METER TCS-172B, Hitachi Aloka Medical, Ltd., Tokyo). These rates ranged from 0.52 to 4.11 $\mu\text{Sv h}^{-1}$ in Kawauchi Village, from 0.23 to 0.31 $\mu\text{Sv h}^{-1}$ in Otama Village, and from 0.61 to 0.85 $\mu\text{Sv h}^{-1}$ in Koriyama City.

Of the 82 samples, 20 were taken from the same individuals or at the same location as in our May 2012 survey (Kiyono and Akama, 2013) in Kawauchi and Otama Villages. We checked the depth of the root system of *Erythronium japonicum*, the least contaminated species in our previous study (Kiyono and Akama, 2013), at a site in Otama Village. All samples of the edible portions were washed with tap water and then rinsed with distilled water before analysis. After draining the water and blotting the samples dry with paper towels to remove surface moisture, they were weighed to determine the FW, oven-dried at 75°C for 72 h or longer, and then weighed. Samples were then cut with a household blender into small pieces to pass through a 3 mm screen. The processed samples were then packed into 100-mL polystyrene containers and subjected to gamma-ray spectrometry measurements. Between 2 May and 4 June 2013, the samples were measured with an HPGe coaxial detector system (GEM20-70, DS-P600 Gamma Studio, Seiko EG & G, Tokyo, Japan) at FFPRI for at least 1,800 s. Detection limit for measurements of radioactive elements was set with the three sigma rule in the DS-P600 Gamma Studio and resulted in the median value of around 80 Bq kg⁻¹ DW (dry weight basis; "DW" henceforth) in our plant samples. Gamma-ray peaks of 604 and 662 keV were used for measurements of ¹³⁴Cs and ¹³⁷Cs, respectively. The measurement system was calibrated using a standard gamma-ray source (MX033MR, Japan Radioisotope Association, Tokyo, Japan). The coincidence-sum effect of gamma-rays from ¹³⁴Cs was corrected. We did not correct for decay because the time between sampling and measurement was sufficiently short that negligible decay would have occurred.

To investigate the factors responsible for differences in the doses of radioactive cesium among the samples, we performed multiple regression analysis by means of stepwise forward regression with $F_{\text{in}} = F_{\text{out}} = 2.0$. Because ¹³⁴Cs levels are related to ¹³⁷Cs levels, as we discuss later, we only used the ¹³⁷Cs levels in the multiple regression analysis and some other analyses to avoid the effects of covariance. When the dose was less than the detection limit, we used the detection limit as the measured dose. We used the 2008 version of the Ekuseru-Toukei software (Social Survey Research Information Co., Ltd., Tokyo, Japan) for calculations. We selected the following seven explanatory variables: (1) the air dose at a height of 1 m at the sample site (ranged from 0.23 to 3.8 $\mu\text{Sv h}^{-1}$); (2) the height class of the individuals that were sampled: trees or woody lianas (unless otherwise noted, "trees" hereafter), 3.5 to 20 m in height; shrubs, woody vegetation less than 3 m in height; herbs, nonwoody vegetation; dummy values of 2 for trees and 1 for shrubs and 0 for herbs were used (0–2); (3) whether the habitat was under a tree crown or at an open site; dummy values were 2 for sites

under a tree crown (including at the forest edge), 1 for partial cover, and 0 for open sites (0–2); (4) plant species with holdfasts (an epiphyte, such as a liana, with holdfasts or suckers to attach to other vegetation) or not; dummy values of 1 for plants with holdfasts and 0 for other plants were used (0–1); (5) growing at a lowlying site that collects surface water and groundwater or not; dummy values of 2 for a lowlying site, 1 for midslope, and 0 for a site where water may not accumulate were used (0–2); (6) the water content of the edible portion (0.71–0.92); and (7) the length of the edible portion (2–60 cm). These variables were selected from the perspective of radioactive cesium sources in the environment. The air dose at a height of 1 m was highly proportional ($R^2 = 0.8854$, $n = 390$, $p < 0.0001$) to total radioactive cesium activity of organic layer and soil (0–5 cm) (Bq m^{-2}) in 2011 (Forestry Agency, 2012). Although the air dose has decreased by decay etc., the ranks of air dose and total radioactive cesium activity of organic layer and soil (0–5 cm) at the monitoring points have rarely changed in 2011–2012 in the study site (Kaneko, personal communication). For these reasons, we substituted the air dose for the total radiocesium activity of organic layer and soil (0–5 cm).

The partial correlation coefficient between some of the explanatory variables was relatively high. For example, plants with holdfasts are apt to be found ($r = 0.3014$, $p = 0.0002$) at a lowlying site. Similarly, a lowlying site was often found under tree shade ($r = 0.3042$, $p = 0.0004$). The mean length of the edible portion was relatively short in species with holdfasts ($r = -0.3590$, $p = 0.0008$) and was

positively correlated with the water content ($r = 0.3784$, $p < 0.0001$). The latter indicates that herbs generally had a long edible portion and a higher water content than trees and shrubs. However, we did not combine or eliminate any variables to eliminate the effects of covariance because our data may not be enough to analyze these relationships in detail.

3. Results and discussion

3.1 Radioactive cesium contamination of edible wild plants

Doses of radioactive cesium in the oven-dried samples ranged from 88 to 27,000 Bq kg^{-1} DW for ^{137}Cs and from 164 to 41,600 Bq kg^{-1} DW for $^{134}\text{Cs} + ^{137}\text{Cs}$. The latter was equivalent to a range from 22 to 4,870 Bq kg^{-1} FW. Of the 82 samples, 20 had doses less than 100 Bq kg^{-1} FW. The most contaminated samples were *Osmundastrum cinnamomeum* var. *fokiense* on a FW basis and *Hydrangea petiolaris* on a DW basis.

We found significant correlations between the air dose and the dose of radioactive cesium in edible wild plants in the spring of 2012 (Kiyono and Akama, 2013) and other researchers obtained similar results. For example, Forestry Agency (2013a, b) published these relationships for male flowers, the wood, bark, and leaves of *Cryptomeria japonica*. Hasegawa *et al.* (<http://dx.doi.org/10.1016/j.jenvrad.2013.06.006>) found this relationship for earthworms. Of the 20 species that we surveyed in the spring of 2013, five species have seven or more number of samples each. Their relationships between the air dose (at a height of 1 m) and ^{137}Cs

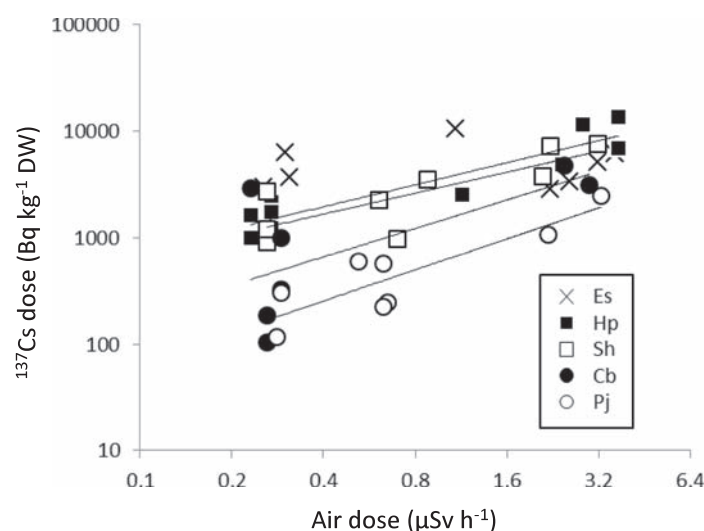


Fig. 1. Air doses (at a height of 1 m) and the doses of ^{137}Cs in five edible wild plant species.

Es, *Eleutherococcus sciadophylloides*; Hp, *Hydrangea petiolaris*; Sh, *Schizophragma hydrangeoides*; Cb, *Clethra barbinervis*; Pj, *Petasites japonicus*. The following statistically not significant or significant relationship were found between the dose per unit dry weight (y) and the air dose (x): For *Eleutherococcus sciadophylloides*: ($R^2 = 0.0095$, $p = 0.8188$), for *Hydrangea petiolaris*: $y = 3636 x^{0.6858}$ ($R^2 = 0.8621$, $p = 0.0001$), for *Schizophragma hydrangeoides*: $y = 3010 x^{0.6511}$ ($R^2 = 0.6489$, $p = 0.0088$), for *Clethra barbinervis*: $y = 1479 x^{0.8870}$ ($R^2 = 0.4450$, $p = 0.1016$), for *Petasites japonicus*: $y = 614.3 x^{0.9662}$ ($R^2 = 0.7607$, $p = 0.0047$).

in the oven-dried samples differed among species (Fig. 1).

Hydrangea petiolaris and *Schizophragma hydrangeoides* were more contaminated than *Petasites japonicus* at all air doses. *Eleutherococcus sciadophylloides* had the highest dose for values of the air dose below around $1.2 \mu\text{Sv h}^{-1}$, but was unique in that it showed no significant increase in ^{137}Cs with increasing air dose, nor was it significantly correlated with topography, height layer, tree shade, water content, or length of the edible portion. *Hydrangea petiolaris* and *Schizophragma hydrangeoides* are lianas that develop holdfasts on the stem or bark of other plants but derive water and nutrients from the humus and soil. They might have derived some of their radioactive cesium from the contaminated bark and humus through their holdfasts (Kiyono and Akama, 2013). On the other hand, the least contaminated edible wild plant was *Erythronium japonicum* with doses ranging from undetectable ($\leq 23 \text{ Bq kg}^{-1} \text{ DW}$) to $88 \text{ Bq kg}^{-1} \text{ DW}$ at Otama and $133 \text{ Bq kg}^{-1} \text{ DW}$ at Koriyama. The roots of this species reached a depth of 15 to 20 cm in the soil, and the soil at these depths may not have become severely contaminated by the spring of 2013. A press release published by Forestry Agency (2013b) suggested that 64 to 92% of the radiocesium was distributed in the top 5 cm of forest soils in the summer of 2012. *Erythronium japonicum* is a spring ephemeral, and its aboveground organs only appear in April and May at the study sites, so the species can take up radiocesium through its aboveground organs (Mitsui *et al.*, 1955) for 2 months, which is a far shorter period than other plants. These factors may have reduced uptake of radioactive cesium by this species.

Similar doses of ^{134}Cs and ^{137}Cs were emitted into the natural environment by the Fukushima accident. If no other similar accident has occurred and the ^{137}Cs in natural ecosystems derived from historical nuclear tests and the Chernobyl nuclear accident are neglected, then the ratio of ^{134}Cs to ^{137}Cs will decrease at a certain predictable rate because the half-life is around 2 years for ^{134}Cs and around 30 years for ^{137}Cs . The following serves an example of this mechanism. The samples of edible wild plants in the present study had ratios of ^{134}Cs to ^{137}Cs that ranged from 0.41 to 0.64 and averaged 0.53. The ratio decreased significantly ($p = 0.00004$) from the values reported in May 2012, which ranged from 0.51 to 0.84 and averaged 0.67 (Kiyono and Akama, 2013).

The water contents of the edible portion of the wild plants ranged between 0.71 and 0.92 by weight, and averaged 0.86. On average, the tree species had significantly lower ($p = 0.0012$) water contents (0.85 ± 0.04 , mean \pm SD) than the herb species (0.88 ± 0.03).

3.2 Factors related to radioactive cesium contamination of edible wild plants

Multiple regression analysis for all species except *Eleutherococcus sciadophylloides* showed that the air dose was significantly positively correlated with the ^{137}Cs dose ($R^2 = 0.651$, $p < 0.0001$). However, the ^{137}Cs dose was significantly higher for plants with holdfasts ($p = 0.0003$) or those that grew at sites with catchment topography ($p =$

0.0182). These findings are similar to those in our previous study (Kiyono and Akama, 2013) in the spring of 2012, around 14 months after the accident. The final multiple-regression equation retained three statistically significant parameters:

$$^{137}\text{Cs} = 1175 + 2223 \ln(AD) + 2545 HF + 926 LL \text{ (adj. } R^2 = 0.651, p < 0.0001, n = 75) \text{ (1)}$$

where ^{137}Cs is the dose of ^{137}Cs ($\text{Bq kg}^{-1} \text{ DW}$) in plants; AD is the air dose ($\mu\text{Sv h}^{-1}$); $HF = 2$ when the plant has holdfasts but 0 when there are no holdfasts; and $LL = 2$ when the plant grew at a lowlying site that collects surface water and groundwater; 1 for midslope, and 0 for a site where water may not accumulate.

Every parameter had a positive coefficient. Thus, the ^{137}Cs dose in the edible wild plants increased with increasing air dose, in plants with holdfasts, and in plants growing in a lowlying site.

3.3 Temporal changes in the doses of radioactive cesium in edible wild plants

We compared the 20 samples taken from the same places in 2012 and 2013. There was no significant difference ($p = 0.071$) in the ^{137}Cs dose between years (Fig. 2), whereas the air dose at these 20 sites decreased significantly ($p = 0.0026$) in 2013 (Fig. 3). The total amount of radioactive cesium in the forest ecosystem has decreased over time at close to the natural radioactive decay rate. The changes in the doses per unit DW differ greatly among plant parts. For instance, radioactive cesium doses in leaves and branches have decreased, but the dose in the soil in the summer of 2012 had increased to between 2 and 3 times its level in the summer of 2011 (Forestry Agency, 2013b). Some radioactive cesium in the leaves and branches may have been leached by the rain and transported into the surface soil. When the radioactive cesium enters the ecosystem, edible wild plants, and particularly shrub and herb species with

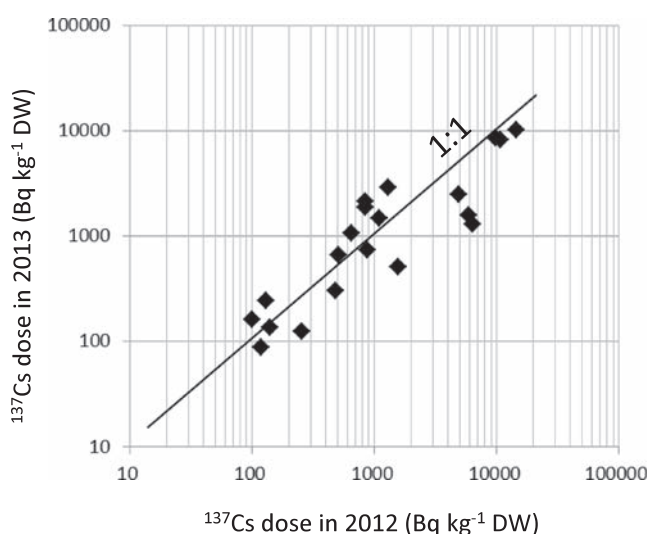


Fig. 2. Relationships between the ^{137}Cs dose in edible wild plants for the 20 sample locations that were the same in 2012 and 2013.

The 1:1 line indicates equal doses in 2012 and 2013.

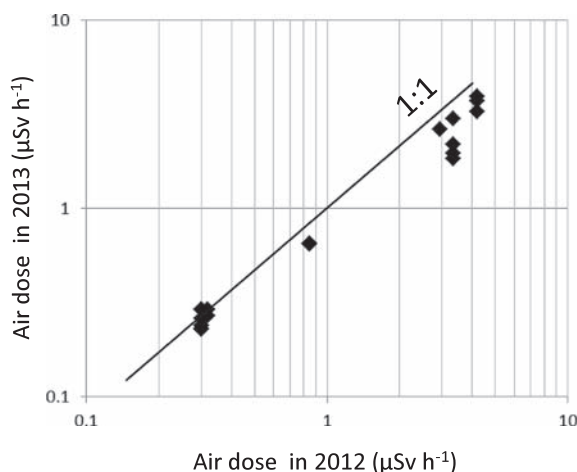


Fig. 3. Relationships between the air dose (at a height of 1 m) in the two years at the 20 sample locations that were the same in 2012 and 2013.

The 1:1 line indicates equal rates in 2012 and 2013.

roots developing in the shallow soil, may take up the radioactive cesium via their roots. However, the mechanisms responsible for the increase of radioactive cesium doses in edible wild plants is not clear, thus future trends cannot currently be predicted.

3.4 Reducing the risk of consuming edible wild plants that are highly contaminated with radioactive cesium

We applied the “New Standard Limits for Radionuclides in Foods” published by the Department of Food Safety, Pharmaceutical and Food Safety Bureau, Ministry of Health Labour and Welfare to radioactive cesium in food on a FW basis; this guide specifies a danger threshold at 100 Bq kg⁻¹

FW. Finally, we investigated the relationship between the total dose of ¹³⁴Cs and ¹³⁷Cs in edible wild plants per unit FW and the environmental and plant factors investigated in this study.

The sum of the ¹³⁴Cs and ¹³⁷Cs doses was significantly positively correlated with the air dose (Fig. 4, $R^2 = 0.2913$, $p < 0.0001$, $n = 82$). Even though the air dose was the same for all plants at a given site, the radioactive cesium concentration differed by up to 100 times. Edible wild plants with relatively high concentrations were *Eleutherococcus sciadophylloides*; two plants with holdfasts, *Hydrangea petiolaris* and *Schizophragma hydrangeoides*; and plants that grew at a lowlying site. Thus, these plants should be avoided to decline dose levels greatly. However, even if we avoid such high-risk plants, the radioactive cesium concentration varied by up to 10 times for a species at the same air dose. Our results provide insufficient data to explain this large difference. All samples exceeded 100 Bq kg⁻¹ FW when the air dose was greater than 0.8 μSv h⁻¹ and for air doses ranging from 0.2 to 0.8 μSv h⁻¹, about one-third of the samples exceeded the threshold of 100 Bq kg⁻¹ FW even after eliminating the high-risk species. Considering the movement of these isotopes through the ecosystem and the rate of decay of these radioactive materials, the relationship between the air dose and radioactive contamination of plants that is shown in Fig. 4 may vary over time. However, thus far, the relationships in Fig. 4 are similar to those that we reported 1 year ago (Kiyono and Akama, 2013).

Based on the findings of the present study and the draft guidelines we developed in our previous study (Kiyono and Akama, 2013), we have proposed a revised set of guidelines when collecting edible wild plants to minimize the risk of consuming edible wild plants that might be highly contami-

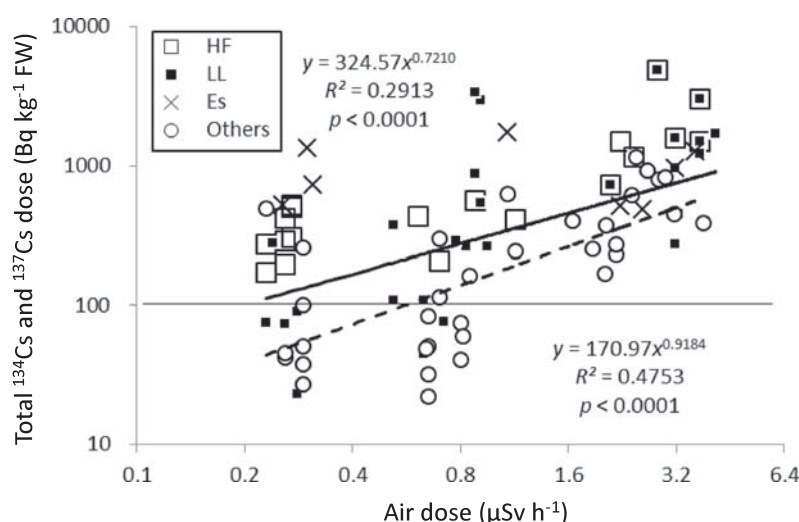


Fig. 4. Relationships between the air dose and the sum of ¹³⁴Cs and ¹³⁷Cs in edible wild plants in the spring of 2013.

Es, *Eleutherococcus sciadophylloides*; HF, Plants with holdfasts; LL, Plants growing at a lowlying site. Others, the other samples. The solid line represents the regression line for data from all samples (including *E. sciadophylloides*), and the broken line represents the regression results only for the “Others”. The horizontal line at 100 Bq kg⁻¹ FW represents the threshold for unacceptably high contamination.

nated with radioactive cesium (Appendix 1).

4. Conclusions

Our survey revealed that the degree of radioactive cesium contamination of edible wild plants increased with increasing air dose. Species with holdfasts and plants growing at a lowlying site are at particularly high risk of dangerous contamination. However, the radioactive cesium contamination at a given air dose varied by as much as 100 times among species. At sites with an air dose higher than $0.8 \mu\text{Sv h}^{-1}$, no edible wild plants had dose levels less than $100 \text{ Bq kg}^{-1} \text{ FW}$. These findings were similar to the results in our previous study (Kiyono and Akama, 2013) using samples taken in the spring of 2012. We will continue our research to clarify the mechanisms underlying changes in cesium contamination and the trends for this contamination so we can continue to revise our guidelines for the safe use of edible wild plants in areas of forest and rangeland affected by the Fukushima accident.

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Appendix 1: Draft guidelines when collecting edible wild plants to reduce the risk of consuming edible wild plants contaminated with radioactive cesium

These draft guidelines represent our best advice based on the current limited data available on cesium contamination after the Fukushima accident. Although it is safest to avoid consuming plants from the regions affected by the accident, following these guidelines will reduce your risk if you choose to consume such plants.

- (1) Edible wild plants are more highly contaminated at sites where the air dose (Japan Atomic Energy Agency, <http://ramap.jmc.or.jp/map/eng/>) is high. Do not collect edible wild plants at sites where the air dose was $0.8 \mu\text{Sv h}^{-1}$ or higher in the spring of 2013.
- (2) Edible wild plants with holdfasts (e.g., some lianas) and plants growing at a lowlying site (where surface water and groundwater flow, such as valley bottoms and small hollows) are more highly contaminated for a given air dose.
- (3) Do not consume *Eleutherococcus sciadophylloides* growing at contaminated sites. This species is likely to be highly contaminated at sites with air doses of $0.2 \mu\text{Sv h}^{-1}$ or higher, and other conditions that may increase the degree of contamination remain unclear.
- (4) The relationship between the air dose and the radioactive cesium concentration in plants will change over time as a result of radioactive decay and movement of radioactive cesium through the ecosystem. Please use the most recent information that is available to guide your decision.