Radiocesium concentrations in wild mushrooms after the accident at the Fukushima Daiichi Nuclear Power Station: Follow-up study in Kawauchi village

Makiko Orita¹, Kanami Nakashima¹, Yasuyuki Taira¹, Toshiki Fukuda¹, Yoshiko Fukushima¹,³, Takashi Kudo², Yuko Endo³, Shunichi Yamashita³ & Noboru Takamura¹

Since the accident at the Chernobyl Nuclear Power Plant, it has become well known that radiocesium tends to concentrate in wild mushrooms. During the recovery process after the accident at the Fukushima Daiichi Nuclear Power Station (FDNPS), it is important to perform follow-up measurements of the activity concentrations of radiocesium in mushrooms. We evaluated the activity concentrations of the detected artificial radionuclides (radiocesium) in wild mushrooms collected from Kawauchi village, which is within 30 km of the FDNPS, in 2015, four years after the accident. We found that the radiocesium was determined in 147 of 159 mushroom samples (92.4%). Based on the average mushroom consumption of Japanese citizens (6.28 kg per year), we calculated committed effective doses ranging from <0.001 to 0.6 mSv. Although committed effective doses are relatively limited, even if residents have consumed mushrooms several times, continuous monitoring of the radiocesium in mushrooms in Fukushima is needed for sustained recovery from the nuclear disaster.
artificial radionuclides (radioesium) in wild mushrooms collected in Kawauchi village (Fig. 1), located within 30 km of the FDNPS, and we found radioesium activity concentrations exceeding 100 Bq/kg (the current regulatory limit for radioesium for general foods) in 125 of 154 mushrooms (81.2%) collected in 2013, two years after the accident15. During the recovery process following the accident at the FDNPS, it is important to perform follow-up measurements of the radioesium in mushrooms, to monitor the dynamics of radioesium in the environment, and to minimize the internal radiation exposure of residents of Fukushima through the consumption of contaminated foods. In this study, we evaluated the radioesium in wild mushrooms in Kawauchi village collected in 2014 and 2015, three and four years after the accident.

**Results**

The activity concentrations of radioesium [the sum of individual activity concentrations (134Cs + 137Cs)] in mushrooms collected in 2015 are summarized in Fig. 2. The activity concentration of radioesium was detected in 147 of 159 mushroom samples (92.5%). Among them, less than 99 Bq/kg of radioesium was detected in 24 mushroom samples (15.1%), 100–999 Bq/kg was detected in 80 mushroom samples (50.3%), and more than 1,000 Bq/kg was detected in 43 mushroom samples (27.0%). On the other hand, radioesium was not detected in 12 mushroom samples (7.5%).

The activity concentrations of radioesium [the individual activities of 134Cs and 137Cs and the sum of individual activity concentrations (134Cs + 137Cs)] in each species of mushroom samples collected in 2015 are summarized in Table 1. Radioesium concentrations was detected in 68 of 68 samples (100%) of *Sarcodon aspratus*, 17 of 17 samples (100%) of *Hygrophorus russula*, and 9 of 10 samples (90%) of *Albatrellus confluens*. The maximum activity concentration was 2,200 Bq/kg of 134Cs in *Hygrophorus russula* and 5,600 Bq/kg of 137Cs in *Sarcodon*.
In the activity concentrations of 137Cs (half-life = 30.1 years) between 2014 and 2015. Although the residential houses have been extensively decontaminated since the accident, the forests of Fukushima Prefecture have not been decontaminated yet. As our results suggest that it takes time to observe a decrease in the radioactivity in wild mushrooms, careful discussion will be needed among stakeholders to determine the necessity of decontaminating the forests in Fukushima.

On the other hand, we calculated committed effective doses ranging from <0.001 to 0.6 mSv based on the average annual intake of mushrooms by Japanese citizens. Previously, we calculated effective doses ranging from 0.1–1.60 mSv in 2013.26. These results suggest that internal radiation exposure due to the consumption of wild mushrooms remained relatively limited in Kawauchi village. We have evaluated the activity concentrations of 134Cs Median (minimum–maximum) (Bq/kg) Detection limit of 134Cs (median) (Bq/kg) Detection limit of 137Cs (median) (Bq/kg) Detection limit of 137Cs (median) (Bq/kg) Detection limit of 137Cs (median) (Bq/kg)

**Table 1.** Concentrations of radiocesium in wild mushroom samples collected in 2015. *n: the number of mushroom samples. *n.e.: the median activity concentration of artificial radionuclides was not quantified. *n.d.: the activity concentration of artificial radionuclides could not be determined.

**Saprophytic**

| Species                  | n | 137Cs Median (minimum–maximum) (Bq/kg) | Detection limit of 134Cs (median) (Bq/kg) | 137Cs Median (minimum–maximum) (Bq/kg) | 137Cs Median (minimum–maximum) (Bq/kg) |
|--------------------------|---|--------------------------------------|----------------------------------------|--------------------------------------|----------------------------------------|
| Armillaria mellea        | 7 | 10 (10–30)                           | 10                                     | 70 (<7–820)                          | 7                                      |
| Lyophyllum decastes      | 5 | 10 (11–30)                           | 11                                     | n.e. (<10–140)                       | 10                                     |
| Armillaria tabescens     | 3 | 20 (<8–50)                           | 8                                      | 30 (<7–30)                           | 7                                      |
| Pholota squarrosa        | 2 | n.d.                                 | 11                                     | 30 (30–40)                           | 9                                      |
| Pleurotus ostreatus      | 2 | 10 (<10–20)                          | 10                                     | 40 (20–60)                           | 8                                      |
| Lentinula edodes         | 1 | 170                                  | 12                                     | 650 (0.0)                            | 9                                      |
| Grifola frondosa         | 1 | n.d.                                 | 13                                     | n. d.                                | 13                                     |

**Symbiotic**

| Species                  | n | 137Cs Median (minimum–maximum) (Bq/kg) | Detection limit of 134Cs (median) (Bq/kg) | 137Cs Median (minimum–maximum) (Bq/kg) | 137Cs Median (minimum–maximum) (Bq/kg) |
|--------------------------|---|--------------------------------------|----------------------------------------|--------------------------------------|----------------------------------------|
| Grifola frondosa         | 1 | n.d.                                 | 13                                     | n. d.                                | 13                                     |

**Discussion**

Recently, we evaluated the radioactivity in wild mushroom samples collected in Kawauchi village and found that radioactivity exceeding 100 Bq/kg was detected in 125 of 154 mushroom samples (81.2%) collected in 2013, two years after the FDNPS accident. In our current study, radioactivity concentrations were detected in 123 of the 159 mushroom samples (77.4%) collected in 2015, four years after the accident. These results suggest that the portion of mushroom samples with radioactivity concentrations above 100 Bq/kg did not dramatically change in Fukushima over two years. In accordance with the shorter half-life of 134Cs, the activity concentrations of 134Cs (half-life = 2.1 years) in 2015 were significantly lower than those found in 2014, whereas there was no difference in the activity concentrations of 137Cs (half-life = 30.1 years) between 2014 and 2015. Although the residential houses have been extensively decontaminated since the accident, the forests of Fukushima Prefecture have not been decontaminated yet. As our results suggest that it takes time to observe a decrease in the radioactivity in wild mushrooms, careful discussion will be needed among stakeholders to determine the necessity of decontaminating the forests in Fukushima.

On the other hand, we calculated committed effective doses ranging from <0.001 to 0.6 mSv based on the average annual intake of mushrooms by Japanese citizens. Previously, we calculated effective doses ranging from 0.1–1.60 mSv in 2013. These results suggest that internal radiation exposure due to the consumption of wild mushrooms remained relatively limited in Kawauchi village. We have evaluated the activity concentrations of...
radiocesium in local foods produced in the village in 2013 and 2014. We showed that the number of samples exceeding the regulatory radiocesium limit (100 Bq/kg for general foods) was five of 4,080 sampled vegetables (0.1%), 652 of 1,986 (32.8%) sampled edible wild plants and fungi, and eight of 647 (1.2%) sampled fruits. In addition, the internal radiation doses resulting from ingesting these foods ranged from 24.4 to 42.7 μSv for males and from 21.7 to 43.4 μSv for females; this confirms that the internal radiation doses are acceptably low compared to the public dose limit of 1 mSv/year. Although residents who returned to the village may have a higher chance of consuming locally produced foods, it does not increase the meaningful of internal doses.

Kawauchi village is the first local authority to return to its hometown following the evacuation after the accident. Currently, almost 68% of residents returned to the village and restarted their lives. Before the accident, the village was famous for its wild mushrooms, including *Sarcodon aspratus* and *Tricholoma matsutake*. Because the collection and consumption of wild mushrooms is a part of the culture of this village, residents are keenly interested in the radiocesium in the wild mushrooms. In 2013, we began collaborating with residents to prepare

![Figure 3. Comparison of concentrations of radiocesium of *Sarcodon aspratus* collected in 2014 and 2015.](image)

![Figure 4. Sampling spots color coded by the concentration of radiocesium in mushrooms collected at each location in 2015. The bottom and top ends of the box and the bar inside the box correspond to the 25th, 75th, and 50th sample percentiles, respectively. The circles with black dots represent extreme values.](image)
a “mushroom map” that includes information about radionuclide concentrations in the mushrooms collected in the village\(^1\). During the recovery phase from the nuclear disaster, the engagement of stakeholders, including residents, local authorities, and scientists, is important to deciding the future direction of the community. The International Commission on Radiation Protection (ICRP) emphasizes that stakeholder engagement is key to the development and implementation of radiological protection strategies for most existing exposure situations, and as experience in stakeholder engagement has grown, it has become possible to use many of the lessons learned as a basis for the development of best practices among the radiation protection community\(^19\). We believe our collaboration in Kawauchi village will contribute to the development of such practices.

Our study has several limitations. We could not evaluate the relationship between radionuclide activity concentrations in mushroom samples and the concentrations in soil due to insufficient soil samples. Further comprehensive studies are necessary to evaluate the activity concentrations of radionuclides in mushroom samples in Fukushima after the accident. Additional analytical uncertainties arise because the committed effective doses from dietary intake of mushrooms cannot measure day-to-day variations in individuals. Further, we did not evaluate the potential loss of radionuclides upon cooking in mushrooms. The influence of eating habits, including cooking methods, must be considered. In this study, we evaluated internal doses from the ingestion of mushrooms, but we did not evaluate the external doses received from being in contaminated forest areas while collecting mushrooms. Further comprehensive analyses with detailed reports on all areas around FDNPP are needed.

In conclusion, we evaluated the activity concentrations of radionuclides in wild mushrooms in Kawauchi village collected in 2014 and 2015, three and four years after the accident, and we confirmed that radionuclides were still detected in most samples. We explained our current results to the residents in the village. Although committed effective doses are relatively limited, we believe that continuous monitoring of the activity concentrations of radionuclides in mushrooms and risk communication with residents in Fukushima is needed for sustained recovery from the nuclear disaster.

**Materials and Methods**

**Sampling of mushrooms.** All wild mushrooms were collected in Kawauchi Village (the public office, N37°20′, E140°48′), Fukushima Prefecture, Japan. Kawauchi village is located approximately 20 to 30 km southwest of the FDNPP and was partially included in the Evacuation Order Area (within a 20-km radius of the FDNPP). During the initial phase of the accident at the FDNPP, almost all residents were evacuated from the village. Saito et al. reported soil deposition density maps of gamma rays that were constructed on the basis of results from large-scale soil sampling in June 2011; these maps showed levels of artificial radionuclides of 100–300 kBq/m\(^2\) for \(^{134}\)Cs and 100–300 kBq/m\(^2\) for \(^{137}\)Cs in Kawauchi village\(^20\). On 31 January 2012, the head of Kawauchi village declared that residents who resided at least 20 km away from the FDNPP could return to their homes based on the declaration by the Japanese Prime Minister that the FDNPP reactors had achieved a state of “cold shutdown.”
Mushrooms have been sampled every year during mushroom season (summer–autumn) since 2013. We asked residents of the village to collect mushrooms and to show the location of each mushroom. We collected 81 Sarcodon aspratus mushroom samples from September to November of 2014, and 159 mushroom samples of 23 species from September to November of 2015. Among the 159 mushroom samples collected in 2015, 68 (43.0%) were Sarcodon aspratus, 17 (10.7%) were Hygrophorus russula, and 10 (6.3%) were Albatrellus confluens (Table 1). The variety of collected mushrooms is quite wide, but some of these are not well known or easy to find, so they were not included in the sampling as they are less likely to be collected for ingestion. All mushroom samples collected were classified according to type into their typical categories of saprophytic or symbiotic, as shown in Table 1.

After collection, all samples were washed by water to remove the soil, and broken into smaller pieces using a mixer machine. The samples, approximately 41 g each wet weight (median), were enclosed in 100-mL plastic containers made of polypropylene (inner diameter, 50 mm; height, 14–42 mm) for radionuclide measurements. All samples were measured fresh and analyzed with a high-purity germanium detector (ORTEC®, GMX30–70, Ortec International Inc., Oak Ridge, TN, USA) coupled with a multi-channel analyzer (MCA7600, Seiko EG&G Co., Ltd., Chiba, Japan) for 3,600 s. The measuring time was set to detect the objective radionuclide, and the gamma-ray peaks used for the measurements were 604.66 keV for $^{134}\text{Cs}$ (2.1 y) and 661.64 keV for $^{137}\text{Cs}$ (30 y). Decay corrections were made based on the sampling date, and detector efficiency calibration was performed for different measurement geometries using mixed-activity standard volume sources (Japan Radioisotope Association, Tokyo, Japan). Activity concentrations of radiocesium were automatically adjusted based on the date of collection, and the data were defined as the activity concentrations at the collection date. The relative efficiency was 31%, and energy resolution of the spectrometer was 1.85 keV for $^{60}\text{Co}$. The detection limit was 11.5 Bq/kg for $^{134}\text{Cs}$ and 9.2 Bq/kg for $^{137}\text{Cs}$ (median), and counting errors were ±7.3 Bq/kg for $^{134}\text{Cs}$ and ±14.1 Bq/kg for $^{137}\text{Cs}$, respectively. Sample collection, processing, and analysis were executed in accordance with standard methods of radioactivity measurement authorized by the Ministry of Education, Culture, Sports, Science, and Technology, Japan21. All measurements were performed at Nagasaki University (Nagasaki, Japan). The sum of $^{134}\text{Cs}$ and $^{137}\text{Cs}$ concentrations was indicated as “activity concentration of radiocesium” in order to aid comparison with the radioactivity measurement of mushrooms collected in 2015. Probability values less than 0.05 were considered statistically significant. All statistical analysis was performed using SPSS statistics 22.0 (SPSS Japan, Tokyo, Japan).

Committed effective doses. The committed effective dose from $^{137}\text{Cs}$ and $^{134}\text{Cs}$ due to mushroom consumption was calculated using the following formula:

$$H_{\text{int}} = C \cdot D_{\text{int}} \cdot e$$

where $C$ is the activity concentration of the detected artificial radionuclide (radiocesium) (Bq/kg), $D_{\text{int}}$ is the dose conversion coefficient for adult intake (age 20 and older, $1.9 \times 10^{-2}$ mSv/Bq for $^{134}\text{Cs}$ and $1.3 \times 10^{-2}$ mSv/Bq for $^{137}\text{Cs}$), and $e$ is the estimated value of annual intake from the latest statistical data issued by the Ministry of Health, Labour, and Welfare, Japan in 201521,22. From this report, annual intakes of mushrooms were estimated at 6.28 kg/year in Japanese citizens, which is based on mean values of adults (>20 y). We assumed that annual intake could be attributed to each species.

Statistical analysis. Values of activity concentrations of radiocesium are presented as median, minimum and maximum. The number of samples of each type of mushroom are listed in Table 1. Analysis of variance (ANOVA) was performed to compare radiocesium activity concentrations in mushroom samples collected in 2014 with radiocesium activity concentrations in mushrooms collected in 2015. Probability values less than 0.05 were considered statistically significant. All statistical analysis was performed using SPSS statistics 22.0 (SPSS Japan, Tokyo, Japan).

References

1. Developments since the 2013 UNSCEAR reports on the levels and effects of radiation exposure due to the nuclear accident following the Great East-Japan Earthquake and Tsunami. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). http://www.unscear.org/docs/publications/2016/UNSCEAR_WP_2016.pdf (accessed on 2 December 2016).
2. International Atomic Energy Agency (IAEA). The Fukushima Daiichi accident. http://www-pub.iaea.org/books/IAEABooks/10962/The-Fukushima-Daiichi-Accident (accessed on 2 December 2016).
3. Nagatoki, S. et al. Measurements of individual radiation doses in residents living around the Fukushima Nuclear Power Plant. Radiat Res. 180, 439–447 (2013).
4. Nagatoki, S. & Takamura, N. A review of the Fukushima nuclear reactor accident: radiation effects on the thyroid and strategies for prevention. Curr Opin Endocrinol Diabetes Obes 21, 84–93 (2014).
5. Nagatoki, S. & Takamura, N. Radioactive Doses - Predicted and Actual - and Likely Health Effects. Clin Oncol (R Coll Radiol) 28, 245–254 (2016).
6. Measurements of internal radiation by WBC (from June 2011 to October 2016) [in Japanese]. Available at http://www.pref.fukushima.lg.jp/site/portal/ps-wbc-kensa-kekka.html (accessed on 2 December 2016).
7. Orita, M. et al. Internal radiation exposure dose in Iwaki city, Fukushima prefecture after the accident at Fukushima Dai-ichi Nuclear Power Plant. PLoS One. 9, e114407 (2014).
8. Skuterud, L. et al. Contribution of fungi to radiocesium intake by rural populations in Russia. Sci. Total Environ. 193, 237–242 (1997).
9. Jesko, T. et al. Age-dependent dynamics of cesium radionuclide content in inhabitants of the Bryansk region, Russia: A seven-year study. Radiat Prot Dosim. 89, 179–182 (2000).
10. Gullien, J. & Baere, A. Radioactivity in mushrooms: A health hazard? Food Chem. 154, 14–25 (2014).
11. Hille, R. et al. Current development of the human and environmental contamination in the Bryansk-Gomel Spot after the Chernobyl accident. *Rad Environ Biophys.* 39, 99–109 (2000).
12. Hoshi, M. et al. Radiocesium in children residing in the western districts of the Bryansk Oblast from 1991–1996. *Health Physics.* 79, 182–186 (2000).
13. Travnikova, I. G. et al. Contribution of different foodstuffs to the internal exposure of rural inhabitants in Russia after the Chernobyl accident. *Radiat Prot Dosim.* 93, 331–339 (2001).
14. Sekitani, Y. et al. Evaluation of $^{137}$Cs body burden in inhabitants of Bryansk Oblast, Russian Federation, where a high incidence of thyroid cancer was observed after the accident at the Chernobyl nuclear power plant. *Radiat Prot Dosim.* 141, 36–42 (2010).
15. Nakashima, K. et al. Radiocesium concentrations in wild mushrooms collected in Kawauchi Village after the accident at the Fukushima Daiichi Nuclear Power Plant. *Peerj.* 3, e1427 (2015).
16. Orita, M. et al. Concentrations of radiocesium in local foods collected in Kawauchi Village after the accident at the Fukushima Daiichi Nuclear Power Station. *Sci Rep.* 6, 28470 (2016).
17. Taira, Y. et al. Evaluation of environmental contamination and estimated exposure doses after residents return home in Kawauchi Village, Fukushima Prefecture. *Environ Sci Technol.* 48, 4556–4563 (2014).
18. Orita, M. et al. Determinants of the return to hometowns after the accident at Fukushima Dai-ichi nuclear power plant: a case study for the village of Kawauchi. *Radiat Prot Dosimetry.* 156, 383–385 (2013).
19. Lochard, I. et al. International Commission on Radiological Protection. ICRP Publication 111 - Application of the Commission's recommendations to the protection of people living in long-term contaminated areas after a nuclear accident or a radiation emergency. *Ann ICRP.* 39, 1–4, 7–62 (2009).
20. Saito, K. et al. Detailed deposition density maps constructed by large-scale soil sampling for gamma-ray emitting radioactive nuclides from the Fukushima Dai-ichi Nuclear Power Plant accident. *Journal of Environmental Radioactivity.* 139, 308–319 (2015).
21. Ministry of Education, Culture, Sports, Science, and Technology, Japan. Environmental radioactivity and radiation in Japan (Available: http://www.kankyo-hoshano.go.jp/en/index.html).
22. International Commission on Radiological Protection (ICRP). 1995. Age-dependent doses to members of the public from intake of radionuclides: Part 5. Compilation of ingestion and inhalation dose coefficients. *Ann ICRP.* 26, 1–91 (1996).
23. Ministry of Health, Labour and Welfare (MHLW). National Health and Nutrition Survey (2012).

**Acknowledgements**
We appreciate the residents of Kawauchi village who kindly cooperated to collect mushrooms.

**Author Contributions**
Conceived and designed the experiments: M.O., K.N., N.T. Performed the experiments: M.O., K.N., T.F., Y.F. Analyzed the data: M.O., K.N., T.K., Y.E. Contributed reagents/materials/analysis tools: M.O., N.T. Wrote the paper: M.O., T.Y., T.K., S.Y. and N.T. Makiko Orita and Kanami Nakashima are co-first authors

**Additional Information**
**Competing Interests:** The authors declare that they have no competing interests.

**Publisher's note:** Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

**Open Access** This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2017