Chapter 12
Radiocesium Dynamics in Wild Mushrooms During the First Five Years After the Fukushima Accident

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Abstract  Dynamics of radiocesium in wild mushrooms, especially in mycorrhizal fungi, in forest ecosystems were investigated for 5 years after the Fukushima nuclear accident, in relation to substrates such as litter, soil and wood debris. Some mushroom species contained a high level of radiocesium in the first or second year, and then the radiocesium content decreased. Changes in radiocesium activities were ambiguous for many other mushrooms. Radiocesium accumulation with time was not common contrary to expectations. Reduction of radiocesium activities in litter and increase in mushrooms and soils, i.e. transfer of radiocesium from litter to mushrooms and soils, was recognized in the first and second year, but it was not obvious in subsequent years. Radiocesium accumulated in several mushroom species, especially in mycorrhizal fungi, while radiocesium in the other mushrooms did not exceed those in the neighboring forest litter. Similar differences in radiocesium level among mushroom species were observed in relation to $^{40}\text{K}$ levels, though $^{137}\text{Cs}/^{40}\text{K}$ ratio in mushrooms was lower than in O horizon, but at the same level of the A horizon in general. These facts suggested differences in the mechanisms of cesium accumulation. Residual $^{137}\text{Cs}$ due to nuclear weapons tests or the Chernobyl accident still remained in mushrooms and soils. From the ratio of the past residual $^{137}\text{Cs}$, it was suggested that the residual $^{137}\text{Cs}$ was tightly retained in the material cycles of forest mushroom ecosystem, whereas $^{137}\text{Cs}$ emitted from the Fukushima accident was still fluid.

Keywords  Chernobyl nuclear accident · Nuclear weapons tests · Radioactive fallout · Radiocesium · The University of Tokyo Forests · Transfer factor · Wild mushrooms

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Abbreviations

Cs cesium
DW dry weight
FW fresh weight
K potassium
NPP nuclear power plant
NWT nuclear weapons test
TF transfer factor
UTF the University of Tokyo Forest

12.1 Introduction

Radioactive material released from the Fukushima Daiichi nuclear power plant (F1-NPP) accident spread over a wide area of East Japan. Wild mushrooms often contain a high level of radiocesium in even lower contaminated areas. The University of Tokyo has seven research forests located in East Japan, 250–660 km from F1-NPP, where radiocesium contamination is low (0.02–0.12 μSv/hr. of air dose rate 1 year after the accident). These forests are used for many activities including research, education, forest management and recreation.

Radiocesium contaminated wild and cultivated mushrooms is a major concern for consumers of these forest products. Fungi, including mushrooms, are also one of the major and important components of the forest ecosystem. Radioactive contamination of mushrooms should be considered not only from the viewpoint of food but also from the viewpoint of its effects on plants and animals through its circulation in the forest ecosystem. Therefore, we have surveyed radiocesium contamination in relation to mushrooms in the University of Tokyo Forests.

Mushrooms have been reported to accumulate radiocesium (Byrne 1988; Kammerer et al. 1987; Mascanzoni 1987; Muramatsu et al. 1991; Sugiyama et al. 1990, 1994). The transfer factors (TF) for radiocesium in mushrooms were reported to be 2.6–21 in culture tests (Ban-nai et al. 1994). However, the radiocesium concentration ratio in mushrooms relative to the soil was rather low and the ratio was often <1 in a field study (Heinrich 1992). Symbiotic mycorrhizal mushrooms tend to have higher TF of $^{137}$Cs than the saprobic fungi in general, though different mushroom species also have widely varying degrees of radiocesium activity (Heinrich 1992; Sugiyama et al. 1993).

Another feature of fungi is the considerable proportion of $^{137}$Cs in forest soil is retained by the fungal mycelia, and fungi are considered to prevent the elimination of radiocesium from ecosystems (Brückmann and Wolters 1994; Guillitte et al. 1994; Vinichuk and Johanson 2003; Vinichuk et al. 2005). Thus, fungal activity is likely to contribute substantially to the long-term retention of radiocesium in the organic layers of forest soil by recycling and retaining radiocesium between fungal mycelia and soil (Muramatsu and Yoshida 1997; Steiner et al. 2002; Yoshida and Muramatsu 1994, 1996). In fact, some examples of long-term $^{137}$Cs radioactivity persistence in mushrooms in forests and transfer to animals have been reported,
whereas that in plants had short ecological half-lives (Fielitz et al. 2009; Kiefer et al. 1996; Zibold et al. 2001).

In previous reports (Yamada 2013; Yamada et al. 2013), radiocesium contamination of wild mushrooms in the University of Tokyo Forests half a year after the Fukushima accident has been summarized. We found rapid uptake of radiocesium in one species of mushroom after the Fukushima accident and residual contamination from atmospheric nuclear weapons tests (NWT) or the Chernobyl accident. In the current study, the dynamics of radiocesium were surveyed over a 5 year period in wild mushrooms and their substrates (litter, soil or wood debris) in relatively low-contaminated forest areas, and features of the dynamics of mushroom contamination were elucidated, paying attention to accumulation and retention of radiocesium in mushroom related forest ecosystems. The raw data of our surveys were presented in Yamada et al. (2018).

### 12.2 Research Sites and Sampling

Mushrooms appeared in the autumn of 2011–2015 (Table 12.1) and their presumptive substrates, i.e., the O horizon (organic litter layer, called A₀ horizon in Japan), the A horizon (mineral layer and accumulated organic matter), and the C/O horizon

| Research forest | Lifestyle | Species                          | Japanese name                      |
|-----------------|-----------|----------------------------------|------------------------------------|
| UTHF (Hokkaido) | M         | Lyophyllum connatum              | Oshiroishimeji                     |
|                 | M         | Suillus grevillea                | Hanaiguchi                         |
| UTCF (Chichibu) | M         | Russula emetica                  | Dokubenitake                       |
|                 | M         | Tricholoma saponaceum            | Mineshimeji                        |
|                 | S         | Bondarzewia berkeleyi            | Oomiyamatonbimai                   |
|                 | S         | Hericium erinaceum               | Yamabushitake                      |
|                 | S         | Sarcomyxa edulis (synonym Panellus serotinus) | Mukitake |
|                 | S         | Trametes versicolor              | Kawaratake                         |
| FIWSC (Fuji)    | M         | Amanita caesareoides             | Tamagotake                         |
|                 | M         | Chroogomphus rutilus             | Kugitake                           |
|                 | M         | Lactarius hatsudake              | Hatsutake                          |
|                 | M         | Lactarius laeticolor             | Akamomitake                        |
|                 | M         | Lyophyllum shimeji               | Honshimeji                         |
|                 | M         | Suillus grevillea                | Hanaiguchi                         |
|                 | M         | Suillus luteus                   | Numeriiguchi                       |
|                 | M         | Suillus viscidus                 | Shironomeriiguchi                  |
|                 | S         | Armillaria mellea                | Naratake                           |
|                 | S         | Pholiota lubrica                 | Chanametsumutake                   |
|                 | S         | Hypholoma sublateritum           | Kuritake                           |
|                 | S         | Lentinula edodes                 | Shiitake                           |
|                 | S         | Pholiota microspora              | Nameko                             |
|                 | S         | Pleurotus ostreatus              | Hiratake                           |
| UTCBF (Chiba)   | M         | Catathelasma imperial            | Oomomitake                         |

*M mycorrhizal fungi, *S* saprobic fungi
(mineral layer with a small quantity of organic matter, which is little affected by pedogenic processes (Soil Survey Staff 2014)) of the soil, or mushroom logs were collected from six (in 2011), 5 (in 2012) and 4 (between 2013 and 2015) research forests shown in Fig. 12.1. Figures 12.2 and 12.3 show examples of samples, the appearance of the environment where samples were collected and sample preparation for radioactivity measurement. The concentrations of $^{134}$Cs, $^{137}$Cs and $^{40}$K were determined using a germanium semiconductor detector (GEM-type, ORTEC, SEIKO EG&G, Tokyo, Japan). Distribution of radiocesium deposition and $\gamma$-ray air dose rate in 2011 was presented in a previous report (Yamada 2013).
Gamma ray air dose rate (μSv/h) 1 m above ground level was measured with a dose rate meter (TC100S, Techno AP Co. Ltd., Japan) using a CsI (Tl) scintillation detector. Although considerable variation in dose rate was observed among UTFs due to environmental variation such as geological features, trends of changes and levels in dose rate were similar within each UTF. Similar levels of pre-Fukushima contamination from nuclear weapons tests and the Chernobyl accident were estimated from 137Cs/134Cs ratio in soils in Chiba (UTCBF) and Fuji (FIWSC). Initial air dose rate in Fuji, however, was somewhat lower than that in Chiba, probably due to geological features. Dose rate slightly decreased in Chiba with time, whereas the decrease was not clear in Fuji. However, in 2015, dose rate in Fuji was similar to the dose rate recorded in Chiba. The original dose rate before the Fukushima accident was thought to be low in Fuji and Chiba. Although contamination due to the Fukushima accident did not reach Hokkaido, the dose rate was higher in Hokkaido (UTHF) than that in Fuji and Chiba. One year after Fukushima accident, the dose rate in Chichibu (UTCF) was higher than that in other UTFs, and was over 0.1 μSv/h, especially in high mountain areas, then gradually reduced by about half by 2015. Dose rate may decrease further in Chichibu, as the dose rate due to the Fukushima accident was estimated at approximately 50 nGy/h (0.05 μSv/h equivalent dose rate of radiocesium) in Chichibu (Minato 2011), whereas the dose rate in other UTFs appeared to become almost stable by 2015.
12.4 Dynamics of Radiocesium in Each of the University of Tokyo Forests (Fig. 12.5)

12.4.1 Litter and Soil Layer

Hokkaido (UTHF): We believe no Fukushima-derived contamination reached Hokkaido because $^{134}$Cs was not detected. $^{137}$Cs was often below the detection limit, and its concentration in the A horizon was similar to the concentration in the O horizon, indicating the contamination in Hokkaido was old from the viewpoint of transfer to the soil. Similarly, $^{137}$Cs was regularly detected in mushrooms even at low levels, but $^{134}$Cs was not detected. It indicated that radiocesium in mushrooms was from the pre-Fukushima fallout.
Chichibu (UTCF): Radiocesium levels in the O horizon were high (200–4400 Bq/kg DW) half a year after the accident, and then decreased relatively rapidly. At the same time, the A horizon also contained $^{134}$Cs (20–120 Bq/kg DW), indicating a rapid transfer to the A horizon because $^{134}$Cs derived from past emissions had already decayed. Subsequent transfer to the A horizon was recognized for example in Tricholoma saponaceum-collected site, however, transfer was generally small. It was possible that radiocesium was mobilizing to lower regions such as valleys or
colluvial slope because of steep slopes, with the exception of a few sites (e.g., the flat land where \( T. saponaceum \) was collected).

Chiba (UTCBF): Radiocesium decreased in the O horizon with time. A certain proportion of radiocesium appeared to transfer into the A horizon even by 2012, however, no clear subsequent transfer was recognized; It might reach a stable condition because of local environmental factors.

Fuji (FIWSC): A unique feature of FIWSC is that a C/O horizon of volcanic Scoria exists instead of an A horizon. FIWSC is covered with Scoria which is a volcanic immature soil ejected from Mt. Fuji. The transfer of radiocesium from the O horizon to the C/O horizon was low (see below). A large proportion of mycorrhizal mycelia may exist in the surface litter layer, and this resulted in mushrooms accumulating a larger amount of radiocesium. Outside of Fuji, heavily contaminated mushrooms have been repeatedly reported around Mt. Fuji despite being a low-contaminated area. A considerable proportion of the contamination was thought to be derived from nuclear weapons testing and the Chernobyl accident.

**12.4.2 Mushrooms**

*Russula emetica* in Chichibu had a high level of radiocesium. Soil analyzed from the *R. emetica*-collection site was also highly contaminated compared with other sites in Chichibu; fallout from the radioactive plume appeared to have deposited here by chance. The dose rate of this highly contaminated site, however, was lower than that of the surrounding sites. The level of \( ^{137} \text{Cs} \) in *Pholiota lubrica*, collected in Fuji, which absorbed quite a high level of radiocesium in the first year of the accident, gradually decreased in one site but remained at the initial level for 4 years in another site. Dynamics of \( ^{137} \text{Cs} \) in the O horizon might reflect the difference because mycelia of *P. lubrica* was spread widely in the O horizon. Six months after the accident, Fukushima-derived radiocesium concentration in mushrooms was lower than that of soils except for *P. lubrica*. Some mycorrhizal mushrooms such as *Suillus grevillea*, *S. viscidus*, *Amanita caesareoides*, *Lyophyllum shimeji* and *Lactarius laeticolor* in Fuji contained less \( ^{134} \text{Cs} \) compared with \( ^{137} \text{Cs} \). It was concluded that the past contamination remained (See Sect. 12.8).

*Trametes versicolor* in Chichibu had a low radiocesium concentration in 2011; the majority of the radiocesium seemed to be derived from the Fukushima accident judging from the proportion of \( ^{134} \text{Cs} \). The radiocesium content in *T. versicolor* was high between 2012–2014, indicating the accumulation in mycelia, but decreased in 2015. In other saprobic mushrooms, a high concentration of radiocesium was detected in *Sarcomyxa edulis* (synonym *Panellus serotinus*) in the first year of the accident, then the content decreased in 2013 and 2014 to the same level found in *T. versicolor*. Litter and soils of the sites, where both mushrooms were collected, were contaminated with radiocesium. Several saprobic mushrooms were collected and surveyed in Fuji; In *Lentinula edodes*, *Pleurotus ostreatus*, *Armillaria mellea* and *Pholiota microspora*, radiocesium level was much higher compared with bark and
wood as substrates, except for bark in 2012. Radiocesium concentration was low in *L. edodes* and *P. ostreatus* but accumulated in *A. mellea*. Saprobes are thought to absorb radiocesium in proportion to the contamination level of the substrate. However, absorption seemed low compared with some mycorrhizal fungi. High radiocesium content in *A. mellea* might be due to the wide distribution of its mycelia in litter and soil, like *P. lubrica* and several mycorrhizal fungi.

## 12.5 Dynamics of Radiocesium in the Same Sampling Sites (Figs. 12.6 and 12.7)

Over a four year period (2011–2015), the decrease in radiocesium concentration by physical decay was 0.912 and 0.262 for $^{137}$Cs and $^{134}$Cs, respectively (calculated from the half-life of both isotopes). $^{137}$Cs content of the O horizon gradually decreased with time more than the rate of physical decay in general, whereas the changes of $^{137}$Cs level were ambiguous in several sites of Chiba and Fuji. $^{137}$Cs was shown to migrate very slowly into the A horizon in Belarus soils after the Chernobyl accident (Kammerer et al. 1994; Pietrzak-Flis et al. 1996; Rühm et al. 1998). In all sites visited in the current study, obvious transfer of radiocesium from the O horizon as well as reduction of radiocesium was not observed in the A or C/O horizon. In the case of mushrooms, *Pholiota lubrica* in Fuji (1 site) and *Catathelasma imperiale* in Chiba (1 site) showed a constant reduction in radiocesium level. The radiocesium concentration in European mushrooms increased for a few years after the Chernobyl accident (Borio et al. 1991; Smith and Beresford 2005); one case of *P. lubrica* and *Suillus grevillea* in Fuji showed a similar pattern, with an increase in radiocesium once during 2011–2012 and a decrease after 2012. In other sites or other mushroom

![Fig. 12.6 Changes in $^{137}$Cs concentration in mushrooms and soils from same sampling sites](image)

No data: either no mushrooms were collected on the site or the radiocesium concentration was below the detection limit.
species, a reduction of radiocesium was not obvious. Because radiocesium activity at each soil depth changes with time, radiocesium activity in different fungal species at different mycelial depths are also expected to vary with time (Rühm et al. 1998; Yoshida and Muramatsu 1994). The variation observed among sites of our field study may be due to geographic and pedological conditions.

The scatter diagram (Fig. 12.7) also showed a decrease of $^{137}$Cs with time in general. The decrease was conspicuous especially in O horizon. $^{137}$Cs concentration in A horizon was low at an early stage of post-accident and no obvious increase or reduction was observed. These results suggested a part of $^{137}$Cs migrated from the O horizon to the A horizon, but a large proportion remained in the O horizon. In the case of mushrooms, considerable variations of the changes in $^{137}$Cs level were observed between species.

**12.6 The Relationship Between Radiocesium Contamination of Mycorrhizal Mushrooms and Soils (Fig. 12.8)**

Mushroom/O or A (C/O) horizon ratio of $^{137}$Cs was compared in mycorrhizal fungi, and found to be high in Fuji and low in Chiba. Additional data is necessary on the same mushroom species, for example between Chichibu and Fuji, to reveal what
environmental factors caused such differences. The mushroom/O or A (C/O) horizon ratio of >1 was common in Chichibu and Fuji on a dry weight basis. The ratio on a fresh weight basis was approximately equal to one with a wide range. These findings corresponded with the results of a field study in Europe (Heinrich 1992). Heavily contaminated *R. emetica* detected in Chichibu did not have a high ratio compared with other mycorrhizal fungi. High contamination could have been due to heavy soil contamination rather than a biological feature of this species. *P. lubrica* in Fuji showed a clear reduction over time for both mushroom/O horizon and mushroom/C/O horizon ratios. In other sites or other mushroom species, reduction in the mushroom/O or A (C/O) horizon ratios were not obvious. A wide ratio range was observed even within the same species, and no obvious radio cesium accumulation was observed in mushrooms over time.

Fig. 12.8 Mushroom/soils ratio of $^{137}$Cs concentration
Mushroom/O horizon ratio (a, c) or mushroom/A (C/O) horizon ratio (b, d) of $^{137}$Cs concentration on dry weight basis (a, b) or on fresh weight basis (c, d)
No data: either no mushrooms were collected on the site or the radio cesium concentration was below the detection limit
12.7 Possible Mechanism Determining Radiocesium Content – The Relationship Between $^{137}\text{Cs}$ and $^{40}\text{K}$
(Figs. 12.9 and 12.10)

Mushrooms generally had a lower ratio of $^{137}\text{Cs}$ to $^{40}\text{K}$ than the O horizon, but a similar ratio to the A horizon; several mycorrhizal fungi in Fuji such as *Lactarius hatsudake* and *L. laeticolor*, which were collected in 2013, were exceptions. For *Pholiota lubrica* collected in 2011, this fungus absorbed radiocesium very quickly probably due to an abundance of its mycelia in the O horizon. On the contrary, the mycorrhizal *Tricholoma saponaceum* in Chichibu and *Catathelasma imperiale* in Chiba, $^{137}\text{Cs}^{/}\text{K}$ ratio in mushrooms was much lower than that in the O and A horizons. It was not clear whether features of the mushrooms or the soil environment resulted in the observed differences between fungi collected from Fuji and Chichibu/Chiba.

On a dry weight basis, $^{40}\text{K}$ concentration seemed high in mushrooms and low in the O or A (C/O) horizons. A reason for radiocesium contamination to be high in mushrooms appears to be because of potassium richness (Seeger 1978). The $^{40}\text{K}$ level, however, was similar for the triparties on a fresh weight basis (Fig. 12.10), suggesting no special mechanism of K absorption. Because most K exists as ions in the cytoplasm, the difference was due to a high water content in mushrooms (a water content of 90–95% is common). The high $^{137}\text{Cs}^{/}\text{K}$ ratio observed in *Russula emetica* was probably induced by heavy soil contamination. On the other hand,

![Fig. 12.9 $^{137}\text{Cs}^{/}\text{K}$ ratio in mushrooms and soils in 2015](image)

- Mushroom; O horizon; A (C/O) horizon. $^{137}\text{Cs}^{/}\text{K}$ ratio in *Russula emetica* was 6.9
- No data: radiocesium concentration was below the detection limit
mushroom/O or A (C/O) horizon ratios of $^{137}$Cs for $R$. emetica (See Sect. 12.6) was not higher compared with Fuji mushrooms, but much higher compared with mycorrhizal fungi in Chichibu and Chiba. Some physiological or ecological mechanisms for Cs accumulation might work also in the case of $R$. emetica.

12.8 Features of Radioactive Contamination with Different Date of Fallout (Fig. 12.11)

A high uptake of $^{137}$Cs by mushrooms, derived from nuclear weapons tests (NWT), was observed in Japan from the 1950s to 1960s (Muramatsu and Yoshida 1997; Sugiyama et al. 1994; Yoshida and Muramatsu 1996). This contamination originated from the global fallout by NWT, which peaked in 1963 (Komamura et al. 2006), and by the Chernobyl accident in 1986. NWT affected the wild mushrooms in Japan more than the Chernobyl accident. The contribution of the Chernobyl accident was estimated to be in the range of 7–60% and 10–30% on average in each study (Igarashi and Tomiyama 1990; Muramatsu et al. 1991; Shimizu et al. 1997; Yoshida and Muramatsu 1994; Yoshida et al. 1994). In the current study, ecological features of radioactive contamination in mushrooms and in soils were discussed by comparing the contamination from the Fukushima accident with those from NWT and the Chernobyl accident.
Shortly after the Fukushima accident, a large proportion of total $^{137}$Cs in the O horizon of soils from Chichibu, Fuji and Chiba were derived from the Fukushima accident, and the $^{134}$Cs/$^{137}$Cs ratio was constant among these research Forests. It showed a similar percentage contribution of contamination before the Fukushima accident. The mean contribution of the Fukushima accident to total contamination was roughly 88% in autumn 2011. This value decreased with time; 86% in 2012, 77% in 2013 and 65% in 2014. These results suggest that radiocesium released from the Fukushima accident moved relatively quickly out of the O horizon, whereas most the past residual $^{137}$Cs remained in the material cycle system on the soil surface. For example, $^{137}$Cs might have been sequestered inside mycorrhizal mycelia. However, it is far from a quantitative evaluation, because of the unstable occurrence of mushrooms between years and locations. In the A or C/O horizon, the mean ratio of Fukushima $^{137}$Cs to total $^{137}$Cs increased from 59% in 2011 to 73% in 2012, then decreased and stabilized at the equivalent level to the O horizon of about 65% in 2013 and 2014. The changes in the ratio appeared to be due to the transfer of $^{137}$Cs from the O horizon, and somewhat to the transfer out of the A horizon.

The proportion of pre-Fukushima $^{137}$Cs is high in mycorrhizal fungi, such as *Suillus grevillea*, *S. luteus*, *S. viscidus*, *Amanita caesareoides*, *Lyophyllum shimeji* and *Lactarius laeticolor* sampled in Fuji. Sugiyama et al. (2000) reported high $^{137}$Cs activities in *P. lubrica* and *S. grevillei* collected around Mt. Fuji in 1996. These fungal species can be characterized by their ability to retain radiocesium. Specifically, more than half the $^{137}$Cs was derived from pre-Fukushima fallout in *S. grevillea*, *A. caesareoides* and *L. shimeji*. Further, *Catathelasma imperiale* in Chiba had a low concentration of $^{137}$Cs, but the ratio of the pre-Fukushima $^{137}$Cs was also high. Thus,
the range of radiocesium concentrations found in mycorrhizal fungi is large. It is unusual that the contribution of pre-Fukushima $^{137}$Cs fallout remained high under the influence of fallout from the Fukushima accident. The mechanisms remain unclear how fungi with a high turnover rate of cells and tissues can retain pre-Fukushima $^{137}$Cs, in which the ratio is much higher than in the soil substrate. $^{137}$Cs deposited over a few decades may continue to be circulated in a closed system of fungal mycelium, which prevents its loss to the lower soil horizons.

### 12.9 Conclusion

In this chapter, some of the dynamics of radiocesium contamination in the forest ecosystem in relation to mushrooms was revealed. Radiocesium accumulation in several mycorrhizal mushrooms was similar to that reported after the Chernobyl accident, but not all mushrooms were contaminated equally. Biology and ecology of mushrooms, geographical, geological and pedological features may affect radiocesium dynamics in forests. Monitoring data of radiocesium concentration could evaluate the transfer of radiocesium from the litter to the soil layer or mushrooms and will provide useful information on the mechanisms of radiocesium accumulation in relation to potassium, and the selective retention of absorbed radiocesium in mushrooms. The number of samples and period of monitoring, however, was insufficient. Long-term monitoring of $^{137}$Cs is necessary to clarify more precisely the dynamics of the contamination, though monitoring of $^{134}$Cs is now becoming difficult because of its short half-life.

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