Graduate School of Agricultural and Life Sciences
The University of Tokyo

Tomoko M. Nakanishi
Keitaro Tanoi *Editors*

Agricultural Implications of the Fukushima Nuclear Accident

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Editors
Tomoko M. Nakanishi
Graduate School of Agricultural and Life Sciences
The University of Tokyo
Bunkyo-ku, Tokyo, Japan

Keitaro Tanoi
Graduate School of Agricultural and Life Sciences
The University of Tokyo
Bunkyo-ku, Tokyo, Japan

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Foreword

On March 11, 2011, a tremendous earthquake occurred in northeast Japan that caused a large accident at a nuclear power plant in Fukushima. This unexpected mishap led to the emission of radioactive nuclides from the power plant, which contaminated a large area, including cities, farmlands, mountains, and the sea, causing serious problems due to agricultural contamination. Immediately after the accident, ^{131}I, ^{137}Cs, and ^{134}Cs were the main radioactive nuclides detected in the fallout. As the day continued, ^{131}I decayed because of its relatively short half-life (half-life: 8 days), but the amount of ^{137}Cs (half-life: 30 years) and ^{134}Cs (half-life: 2 years) remained the same. However, in approximately 1.5 years, ^{134}Cs had decayed gradually, whereas the amount of ^{137}Cs was similar to that of the original radioactivity due to its long half-life.

From an agricultural perspective, one of the most important issues is the security of food safety for consumers and producers of various agricultural products. After 1 year, the government amended the new standards and enforced much lower radioactivity levels in food, including water and milk. However, some products had higher radioactivity levels than those allowed by the new standards, although they were not regarded as contaminated according to the provisional regulation levels.

In our Graduate School of Agricultural and Life Sciences at The University of Tokyo, many academic staff members have initiated research activities in their specific fields, such as the contamination of soil, plants, animals, and fish, and the activity of radiation in the environment. Some of these studies have involved cooperative research with other research organizations such as the Fukushima Agricultural Technology Center. The initial research results have been published in the Japanese journal *Radioisotopes*, written in a language that can readily be understood by Japanese readers without a technical background. We have also held several report meetings, every 3–4 months since November 2011, which are open to the public. This book contains the results that have already been reported in Japanese journals, at meetings, or in new studies.

The effects of radioactive contamination will persist for a long time into the future. Investigating the behavior and mechanisms of radioactivity is the only way to develop solutions to cope with the contamination. Thus, the most urgent issue addressed by this research is to understand how radioactivity is incorporated into foods and how radioactive contamination can be prevented in agricultural products.

Tokyo, Japan

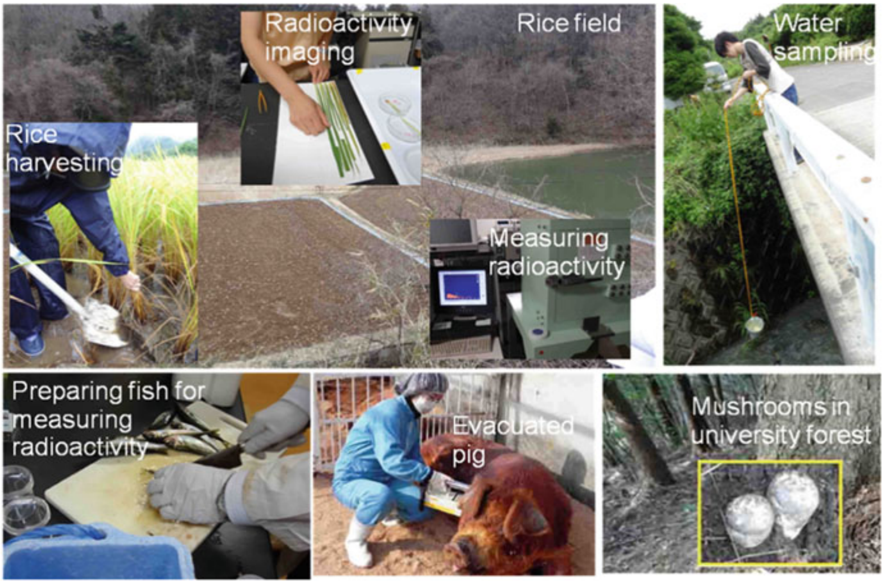
Hiromichi Nagasawa
Professor
Dean, Graduate School of Agricultural and Life Sciences
The University of Tokyo

Preface

Since the Fukushima Daiichi nuclear power plant accident in March 2011, contamination of places and foods has been a matter of concern. Unfortunately, agricultural producers have few sources of information and have had to rely on the lessons from the Chernobyl accident in 1986 or on information obtained from the International Atomic Energy Agency. However, as of this writing, data on the specific consequences of the Fukushima accident on Japanese agriculture remain limited. More than 80% of the land that suffered from the accident was related to agriculture or was in forests and meadows. The influence of the accident on agriculture was the most difficult to study because the activity in nature had to be dealt with. For example, when contaminated rice is harvested, scientists working on rice plants and soils and the study of watercourses or mountains have to collaborate to analyze or determine the vehicle by which the radioactivity accumulated and through which it spread in nature.

The situation with Fukushima differs from that with Chernobyl, needless to say, in terms of the nature of the soil, breeding species, types of grass or trees, and other aspects. When the nuclear accident at Fukushima is compared with that at Chernobyl, the contaminated area in Fukushima, both low and high in radioactivity, was approximately 6% of that in Chernobyl. The amount of fallout in Fukushima was approximately one-sixth and the distance to which the fallout spread was approximately one-tenth that in Chernobyl.

At the request of agriculturists in Fukushima, we at the Graduate School of Agricultural and Life Sciences at The University of Tokyo have been urgently collecting reliable data on the contamination of soil, plants, milk, and crops. Some of the objects of our activities can be seen in the following photos. Based on our data, we would like to comment on or propose an effective way of resuming agricultural activity. Because obtaining research results based on in situ experiments is time-consuming, we have been periodically holding research report meetings at our university every 3–4 months for laypeople, showing them how the contamination situation has changed or what type of effect can be estimated.



Although our research is still ongoing, we would like to summarize in this book our observations made during the one and a half years after the accident.

Tokyo, Japan Tomoko M. Nakanishi

Contents

Chapter 1
The Overview of Our Research

Tomoko M. Nakanishi

Abstract The overview of our research projects for Fukushima is presented including how they were derived. Then, where the fallout was found, right after the accident, is briefly summarized for soil, plants, trees, etc. The time of the accident was late winter, there were hardly any plants growing except for the wheat in the farming field. Most of the fallout was found at the surface of soil, tree barks, etc., which were exposed to the air at the time of the accident. The fallout found was firmly adsorbed to anything and did not move for months from the site when they first touched. Therefore, the newly emerged tissue after the accident showed very low radioactivity. The fallout contamination was not uniform, therefore, when radiograph of contaminated soil or leaves were taken, fallout was shown as spots. Generally, plants could not absorb radiocesium adsorbed to soil. Further findings are described more in detail in the following sections.

Keywords Fallout • Research in agriculture • Research project • Research site • The way of contamination

1.1 Research Project

After the accident of Fukushima Daiichi nuclear power plant, thousands of measuring data have been piling up, especially in the web sites of government agencies. However, most of them are two kinds of the data. One is the radio activities of the places, including soil, air dust or sea water and the other is the measurement of the foods. These are just the monitoring data and it is difficult to find out the research

T.M. Nakanishi (✉)
Graduate School of Agricultural and Life Sciences, The University of Tokyo,
1-1-1 Yayoi, Bunkyo-ku, Tokyo 113-8657, Japan
e-mail: atomoko@mail.ecc.u-tokyo.ac.jp

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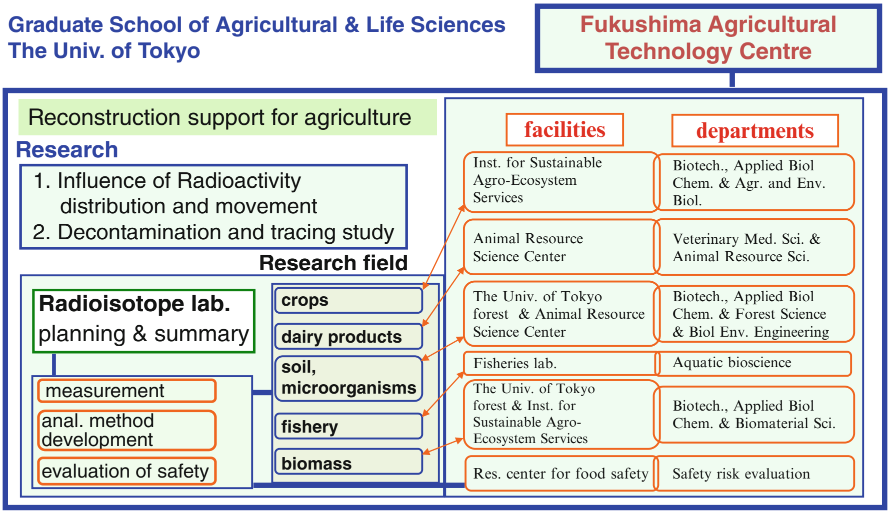

Fig. 1.1 Research project. About 40 academic staffs from all kinds of the facilities and departments of our Graduate School proposed research plans and they were classified into several groups. Wide ranges of the agricultural research have been going on

data related to agriculture, such as, how much amount of radioactivity was found or estimated when the plants were grown in the contaminated field or how much radioactivity was accumulated in mountains and what about the contamination of river water coming from the mountain, etc.

Right after the accident, many academic staffs in our Graduate School had started the research project for Fukushima. But soon we found that it was very difficult to pursue the research by individual researcher alone, since the target is very complicated and it is the study of nature itself. For example, when contaminated rice was found, discussion was needed not only from scientists of rice but also from those of soils or water flow. When our dean, Prof. H. Nagasawa, had asked us, right after the accident, what kind of the research we can do, about 40 academic staffs corresponded to our dean's request and proposed their research plans. In our faculty, wide ranges of the agricultural research have been going on, for plants, soil, animals, fish, etc. Many scientists are pursuing their own individual majoring field. Since the research plans proposed were from all kinds of the facilities and departments of our Graduate School (Fig. 1.1), they were classified into several groups as follows, and the research projects were started. However, there has not been any budget prepared for these projects. That means, most of them were started and developed based on voluntary activities.

(Research project group)

1. Influence of fallout (distribution and movement)
   (a) Crop plants and soils
   (b) Stock raising & dairy products
   (c) Fishery

(d) Environment, including wild life
(e) Radiation measurement & radiochemistry
(f) Science communication

2. Recovery of suffered agriculture (recovery from Tsunami effect)

(a) Crops production and soils (salt damage, farmland maintenance, etc.)
(b) Biomass production

These research project groups were sometimes merged or further divided along with the development of the research. It was our challenge and first trial to perform the research by the group of the scientists who had never discussed nor carried out the same project before, in such a wide range of research field. In this meaning, these projects are very unique, not only covering vast field of agricultural research but also consisted of so different types of the scientists.

1.2 Research Site

The main research sites were shown in Fig. 1.2. Since many staffs belong to the facilities attached to the Graduate School were participated, the research sites were distributed in wide range of the districts in Japan. A tremendous number of the samples collected at each site were sent to the radioisotope lab. of the Graduate School, located at the main university campus in Tokyo. Then, the radioactivity of the samples were measured by germanium semiconductor detector or NaI(Tl) counters and the radioactivity imaging was performed using imaging plates or real-time radioisotope imaging systems (Kanno et al. 2012; Kobayashi et al. 2012; Hirose et al. 2012), we developed.

Besides our facility sites, we started collaboration with Fukushima Agricultural Technology Centre, which is the largest agricultural research center in Fukushima prefecture, including Fruit & Tree Lab., right after the accident. This collaboration was in a large field of the research, including vegetables, serial plants or fruit trees, soils, etc.

We started field ecological study and decontamination research at Date city, Samegawa village and Iitate village. In the case of Date city, a big project has started to analyze the ecological circulation or movement of the radionuclides, including food chain of the wild lives. The flow of the radiocesium via water with respect to the feature of the landscape is also being studied.

The wild life researchers are performing their investigation as close as possible to the nuclear reactor site but it was prohibited to enter the place within 30 km from the reactor. They are collecting small animals or insects and studying the contamination level or distribution of the fallout. Then they analyzed the specific accumulation manner of the nuclides along with food chain.

Among our facilities, the highest radioactivity was counted at Animal Resource Science Center. Therefore, they prepared the haulage from contaminated grass and fed them to the cow to see how radiocesium is transferred to the milk. Several kinds

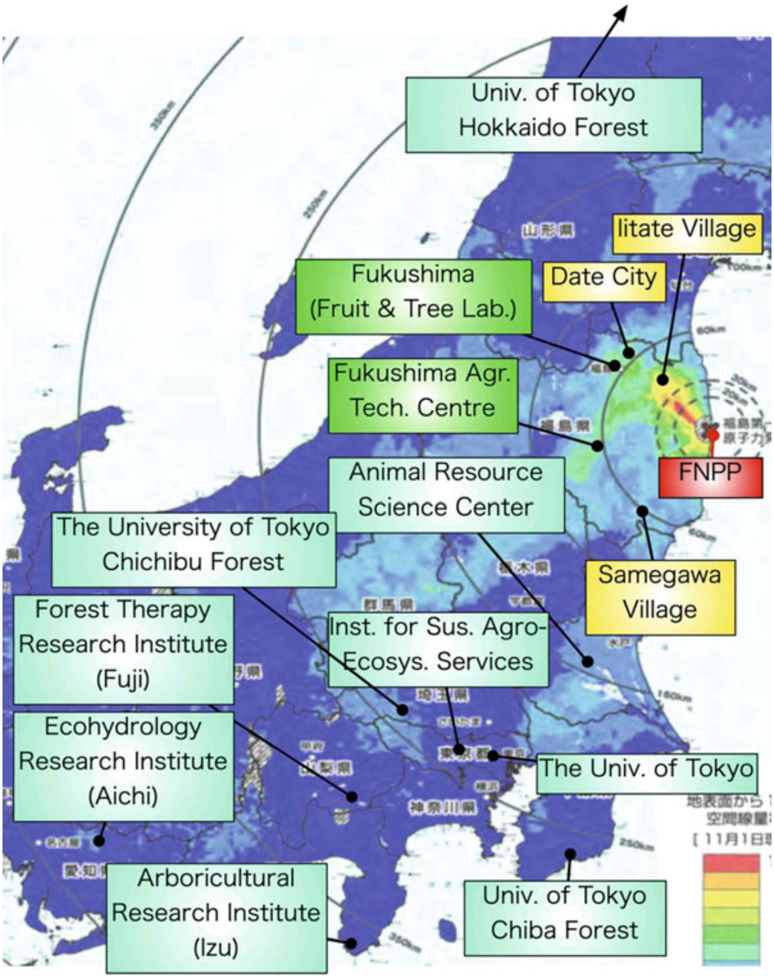

Fig. 1.2 Research site. The research sites were distributed in wide range of the districts in Japan. A tremendous number of the samples collected at each site were sent to the radioisotope lab. of the Graduate School, located at the main university campus in Tokyo

of highly contaminated stock animals were brought in from Fukushima prefecture to see what kind of influence can be seen in second or third generation. The wild animals captured at highly contaminated place in Fukushima were also brought in to find out the distribution of the radioactivity in tissue.

The researchers of marine biology are collecting the samples mainly at Ibaraki prefecture, adjacent to the Fukushima prefecture, to the south. They asked the fishery labor union to collect the fish from different depth in sea.

There is our farming land in Tokyo, called Institute for Sustainable Agro-Ecosystem Services. Though the contamination level was very low, since it is about

230 km far from the nuclear power plant, the influence of low level contamination to the agricultural products are studied. Especially for fruit trees, the radioactivity throughout the tree tissue was scrutinized. They found that the small amount of radioactivity can be transferred from bark skin to the xylem tissue, which was estimated as the main route for radiocesium accumulation in fruits.

In the case of The University of Tokyo Forests, litters or mushrooms were collected and the radioactivity was measured. It was noted that some kind of mushrooms selectively collected the very old radioactive fallout, about 40 years ago, when open test of atomic bomb was conducted.

1.3 Fallout on Soil

The initial information we got was as follows. In the case of soil, most of the radioactivity was detected in the surface, 2–3 cm of soil in the fields of Fukushima (Fig. 1.3). Many pipes were prepared on the ground and the radioactivity profile along with the depth was measured (Shiozawa et al. 2011). The left figure in Fig. 1.3 shows the radioactivity profile in the soil. When the farming soil was collected and the radioactivity images were taken by an imaging plate, radioactivity was found as spots. The radiograph of the soil showing that the contamination was not uniform in the soil suggested that the radioactive nuclides were adsorbed at particular site of the soil. The soil was crashed and separated to find out which part of the soil the radioactivity was accumulated. The highest radioactivity was measured in two fractions, the finest fraction, clay, and an organic layer, which was the debris of the fallen plant tissue and was not yet decomposed completely by microorganisms.

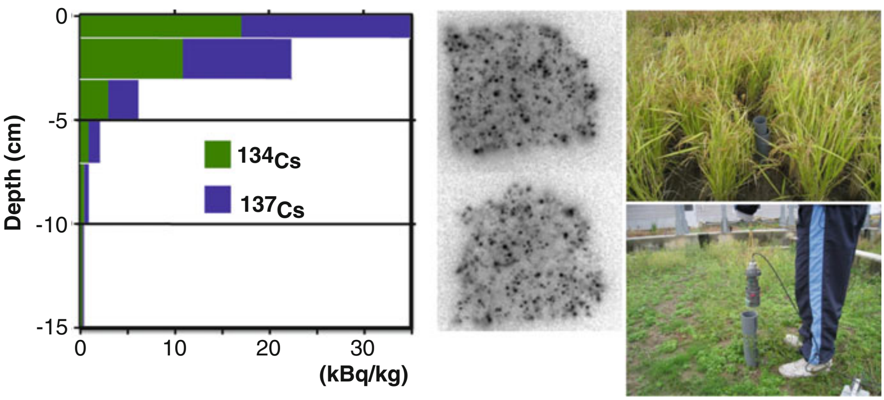

Fig. 1.3 Fallout on soil. Most of the radioactivity was detected in the surface of soil in the fields of Fukushima (*left*). The contamination was not uniform in the soil suggested that the radioactive nuclides were adsorbed at particular site of the soil (*middle*). Many pipes were prepared on the ground and the radioactivity profile along with the depth was measured (*right*)

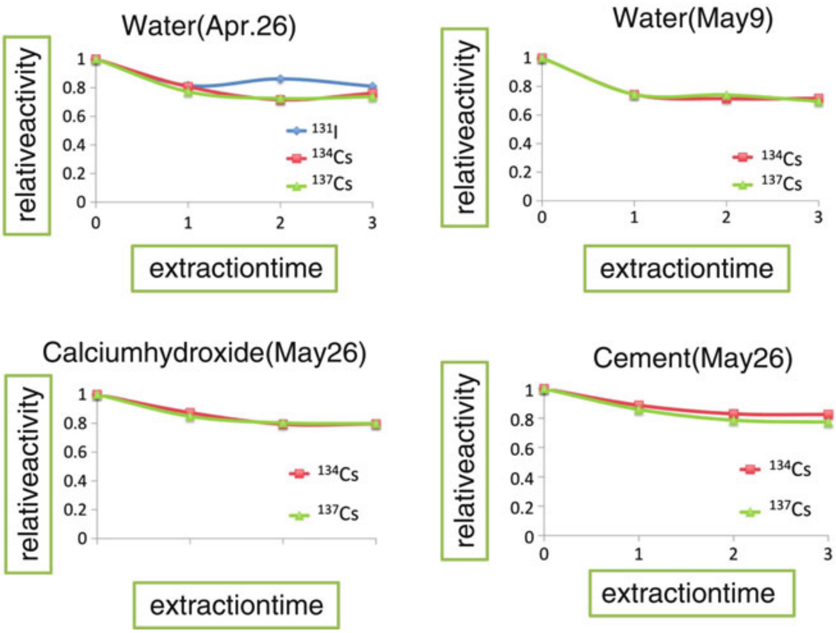

Fig. 1.4 Extraction of radioactive nuclides from paddy soil for a rice plant. Soil was washed under different treatments to find out in which condition the radioactivity can be removed from the soil

Then, the soils from different farming place were collected and were washed under different treatments to find out in which condition the radioactivity can be removed from the soil (Nogawa et al. 2011). As is shown in Fig. 1.4, the initial washing removed at most 20% of the radionuclides. However, further washing with solutions of caesium iodide, hydrated lime, fertilizer, and even cement did not remove any radioactivity, suggesting that most of the nuclides that remain in the soil were so firmly adsorbed to the soil that it is difficult to decontaminate the soil with chemical treatment.

Therefore, it was suggested that this thin contaminated surface soil can thus be collected and buried on site, leaving the land safe to work again, as the radionuclides are unlikely to be leached from the soil (Fujimura et al. 2012). The downward movement of the radiocesium in soil was further monitored.

1.4 Fallout on Plants

The way of contamination in the plants grown at the time of the accident was as follows. Two months after the accident, wheat grown in Fukushima had high levels of radioactivity, but the radioactivity was measured mainly in the older leaves, which had been expanding at the time of the accident (Tanoi et al. 2011). Leaves that emerged after the accident had far less radioactivity, and ears had only 1/2000th

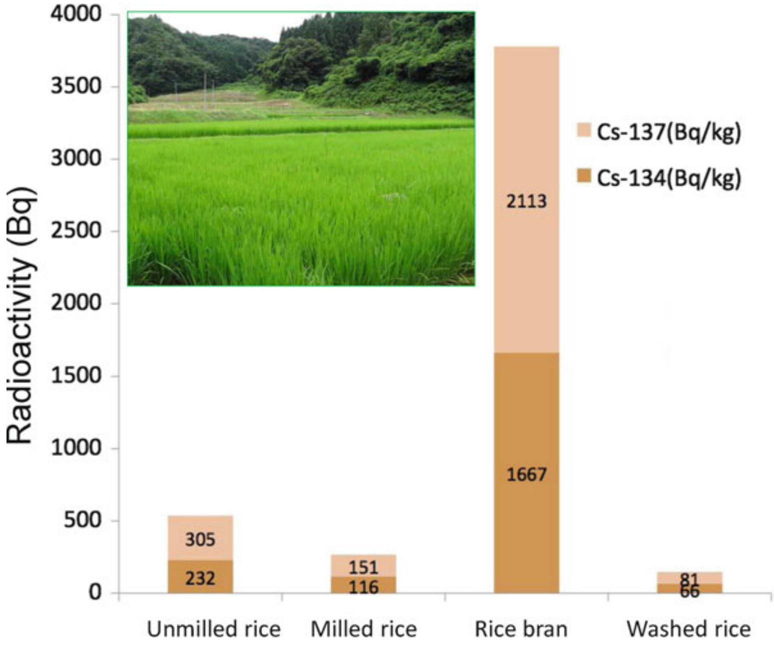

Fig. 1.5 Distribution of radioactivity in rice grain. Most of the radiocesium is accumulated in outer skin, rice bran. When the grain is milled, radioactivity is reduced to about half. By washing, the radioactivity was further reduced to about half

the radioactivity. These results suggested that the fallout adhered firmly to the leaf surfaces and that only very small amounts of the radionuclides had been absorbed and transferred to other organs. Radioisotope imaging has revealed the fallout as microscopic grains on the leaf surfaces, similar to the spot images of soils.

Since fallout was firmly adsorbed on soil, it was estimated that only small amount of radioactivity was absorbed by plant roots when grown in contaminated soil. However, in some rare cases, like that in Date city, the harvested rice grains showed high radioactivity, more than 500 Bq/kg (provisional regulation level), even though they were grown in the soil with low radioactivity, less than 5,000 Bq/kg. In such cases, the paddy rice field was surrounded by small forests and because of the landscape, the rice field was shaped as stepwise terrace (Fig. 1.5). To grow rice plants, a large amount of water is needed and the water is introduced as stream from the neighboring forest for this kind of the field. The water was conducted to the highest terrace of the field first and then it was introduced successively to the downward fields. However, the contamination level of the rice grain was different among the terrace fields. The highest radioactivity was not necessarily found in the rice grown in the field located at the highest level, i.e. closest to the mountain. The accumulation manner of radiocesium by rice grain was analyzed in laboratory using the contaminated soil (Nakanishi et al. 2012). Once radiocesium is dissolved with water, plants easily absorb and accumulate radiocesium but the problem is how fallout could be dissolved

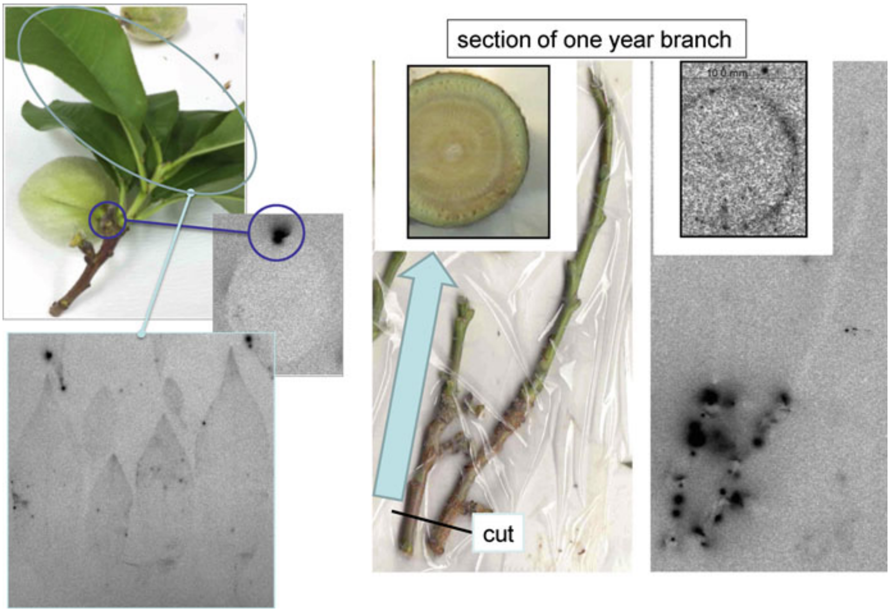

Fig. 1.6 Peach tree. The radiograph of fruit leaves and branches were taken by an imaging plate. In the case of a fruit, only bottom part, connecting to the branch, showed high radioactivity and the leaves emerged after the accident did not show any high radioactivity (*left*). In the case of branch, only the part already grown at the time of the accident was highly contaminated and newly emerged branch after the accident showed hardly any radioactivity. The section of the branch showed that only outer skin was highly contaminated (*right*)

as ions in water. The dissolved radiocesium ion could be derived from two ways, one from the litter in the mountain and the other from the organic matters in the rice field. Since water itself from the mountain is not contaminated, it was estimated that when the contaminated litter was decomposed by microorganisms, the radionuclides were adsorbed to the soil right after the decomposition. There must be some special condition to dissolve radiocesium in litter. In paddy field, organic matters at soil surface are decomposed especially during hot summer. The rice roots around this time grow horizontally to support the plants with developing ears firmly, therefore, the roots could be very close to the soil surface, decomposition site, to absorb radiocesium. Now further experiment is conducted to find out the source of the rice grain contamination.

Figure 1.5 shows the distribution of radioactivity in rice grain. Most of the radiocesium was accumulated in outer skin, rice bran. Therefore, when the grain was milled, radioactivity was reduced to about half. Then by washing, the radioactivity was further reduced to about half. To eat rice, water is added and steamed so that the radioactivity per kg unit is further reduced.

In the case of peach trees, the fallout contamination was found in the same way as that of wheat. That is, only the tissue already grown and exposed to the air, at the time of the accident, showed high radioactivity. Figure 1.6 shows radiograph of fruit, leaves and branches taken by an imaging plate. In the case of fruit, only bottom part of the fruit connecting to the branch showed high radioactivity. The radioactive nuclide was accumulated only at this part and did not move toward the fruit. The radiograph of the leaves showed that the leaves emerged after the accident did not show any high radioactivity. In the case of branch, radiograph showed that only the branch part already grown at the time of the accident was highly contaminated and newly emerged branch after the accident showed hardly any radioactivity. The dissection of the branch showed that only outer skin was highly contaminated. In the case of the other kind of trees, only bark was highly contaminated and pealing the bark was the most effective way for decontamination (Takada et al. 2012).

To remove radioactivity by plants was proposed, since plant roots excrete acids to mobilize nutrients. Plants grown in contaminated soil could therefore remobilize the radionuclides and accumulate them in their roots. However, in our common view, it is extremely difficult to find out or create the plants which accumulate high amount of radiocesium. In the case of phyto-remediation, the concentration of the accumulated target nuclides in the plant has to be much higher than that of soil. When phyto-remediation is taken into account, the most promising method now considered is to analyze the mobilization mechanism of adsorbed radionuclide in soil by the mushroom and introduce the function to the plants.

1.5 Others

On the university's farm in Ibaraki prefecture, about 160 km southwest of the Fukushima nuclear power plant, we fed cows with silage made from contaminated grass harvested in Ibaraki. After 5 days the milk contained about 1/100th the radioactivity; it had thus rapidly accumulated the radiocesium (Hashimoto et al. 2011).

On the university's farm in the Tokyo, about 230 km southwest of the nuclear power plant, we grew cabbage and potatoes in soil that had an activity of about 100 Bq/kg (15 cm depth). After 2 months, the radiocesium in the developed leaves before washing was 9 Bq/kg, much lower than the provisional regulation level of 500 Bq/kg (Oshita et al. 2011).

We have now expanded our investigations into a long-term study that will also cover trees and fisheries. In the following sections, further findings are described more in detail.

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Chapter 2
Behavior of Radiocesium Adsorbed by the Leaves and Stems of Wheat Plant During the First Year After the Fukushima Daiichi Nuclear Power Plant Accident

Keitaro Tanoi

Abstract The behavior of radiocesium in winter wheat after the accident in March 2011 was characterized on the basis of an investigation of radiocesium in wheat grown in open fields. The distribution of the radiocesium contamination of wheat was analyzed by determining the radiocesium concentration in each part approximately 2 months after the fallout occurred in agricultural fields, which was a short period after the hydrogen explosion occurred at Fukushima Daiichi nuclear power plant. At that time, only the leaves and stems, but not the grains, were contaminated directly by the fallout because the wheat growing in the fields was in juvenile phase, before heading. The radioactivity was more than 1,000 times higher in the leaves growing at the time of the accident than that in the panicles that developed later. Autoradiographic images captured using an imaging plate showed that the highest radioactivity was found in many spots on the old leaves, suggesting that the radionuclides were strongly bound to the leaves. Moreover, effects of the seed sowing date, which varied from October 8th to November 20th, 2010, on the radiocesium concentration in grains were investigated. The radiocesium concentration in the grains after harvest was correlated with the plant height measured on May 28th. These results suggest that the radiocesium in the grains was derived from leaves and stems where it had accumulated when the fallout occurred.

Keywords Autoradiography • Contamination • Radiocesium • Wheat

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K. Tanoi (✉)
Graduate School of Agricultural and Life Sciences, The University of Tokyo,
1-1-1 Yayoi, Bunkyo-ku, Tokyo 113-8657, Japan
e-mail: uktanoi@mail.ecc.u-tokyo.ac.jp

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2.1 Introduction

In mid-March 2011, the fields in the east of Japan, particularly part of Fukushima Prefecture, were contaminated with radiocesium and radioiodine after an accident at the Fukushima Daiichi nuclear power plant. There are many reports of agricultural contamination by the fallout in the air, mostly related to nuclear bomb tests from 1940s to 1970s and the incident at Chernobyl. In these reports, transfer factor (TF) was used as an index of the degree of radionuclide transfer from the soil to the edible parts of crops. There are few data related to TF of radiocesium in wheat in Japan, although Uchida et al. (2007) reported that TF (dry-weight basis) of stable Cs in husked wheat was in the order 10^{-4} to 10^{-3}.

The radiocesium concentration in husked wheat produced in 2011 in Fukushima Prefecture was measured, and most of the husked wheat was found to have an extremely low concentration of radiocesium, which was supported by the low TF of Cs in wheat. However, a high concentration of radiocesium, >100 Bq/kg, was detected in a small proportion of the husked wheat in 2011. In general, direct contamination of the edible parts had the highest effect on the level of contamination in the edible parts. However, in this case, the grains were not directly contaminated by fallout because the wheat growing in the fields was in the juvenile phase, before heading. Thus, other mechanisms of radiocesium transfer to grains exist in addition to soil-to-plant (or grains) transfer and direct contamination of grains.

To understand the mechanism of radiocesium transfer to the grains in 2011, we used wheat plants that were growing in the open fields before the accident, in which we clarified the distribution of radiocesium contamination. The effects of the seed sowing date on the concentration of radiocesium in grains from the open field were also investigated.

2.2 Materials and Methods

2.2.1 Analysis of Radiocesium in Each Plant Part

The winter wheat cultivars "Kinuazuma," "Yukichikara," and "Abukumawase" were sowed directly (0.7-m furrow, 0.8 kg/a) on the wheat field (gray lowland soil; $N:P_2O_5:K_2O = 1:1:1$ kg/a as the basal fertilizer) in mid-fall, 2010. Ammonium acetate was used as a top-dressing at 0.3 kg/a on February 25th, 2011. For monitoring the growth, the number of expanded leaves was counted for ten plants belonging to each cultivar.

Each plant part was collected on May 26th, 2011 (2 months after the fallout). Gamma ray analysis was performed using a germanium semiconductor detector for the "Kinuazuma" cultivar. The parts used in the analysis were the panicle, stem, flag leaf 1(FL1; the top of the flag leaf), FL2 (the leaf below FL1), FL3 (the leaf below FL2), and "other leaves" (older leaves). The samples were measured using a germanium semiconductor detector (GEM type, Seiko EG&G). The gamma ray energies used to measure ^{134}Cs and ^{137}Cs activities were 604.7 and 661.6 keV, respectively.

Autoradiography was performed for the three cultivars. Leaves from each position were collected and fixed onto Kent paper. After wrapping them in polypropylene film, the samples were exposed to an imaging plate (IP; BAS-IP MS2025, Fuji Film) for 7–10 days at −80 °C. IP was scanned using a high performance image analyzer (FLA-5000, Fuji Film) at 100-μm resolution with 16-bit data size and the data obtained was the photostimulated luminescence value.

2.2.2 Effects of the Seed Sowing Date on the Radiocesium Concentration in Grains

The wheat cultivar "Fukuakari" was sowed directly (0.8 kg/a) on the wheat field (gray lowland soil; $N:P_2O_5:K_2O = 1:1:1$ kg/a as the basal fertilizer) on October 8th, October 20th, November 8th, and November 20th, 2010. Ammonium acetate was used as a top-dressing at 0.3 kg(N)/a on February 25th, 2011. The plant heights were measured on March 28th, 2011. The concentration of radiocesium in the grains was determined after harvest using a germanium semiconductor detector, as described in Sect. 2.2.1.

2.3 Results and Discussion

2.3.1 The Radiocesium Was Tightly Bound to the Leaves That Had Already Expanded During the Fallout

2.3.1.1 The Concentration of Radiocesium in Each Plant Part Sampled from the Wheat Cultivar "Kinuazuma" on May 26th, 2011

From the gamma spectra of the leaf samples, only radiocesium (^{134}Cs and ^{137}Cs) was identified as the fission products; therefore, the concentration of radiocesium was determined (Fig. 2.1).

The concentration of radiocesium was highest in "other leaves," and it declined drastically with the leaf position; the newest leaves contained the lowest radiocesium concentration. The panicles had the lowest concentration of radiocesium, which was more than 1/1000th of the "other leaves."

2.3.1.2 Imaging Analysis of Radionuclides in Leaves from the Wheat Cultivars "Kinuazuma," "Yukichikara," and "Abukumawase" Sampled on May 26th, 2011

Using the same samples that were analyzed by the germanium detector, we performed imaging analysis using IP to identify the distribution of radionuclides on the leaves. IP was more sensitive to beta rays than gamma rays, as is the case with

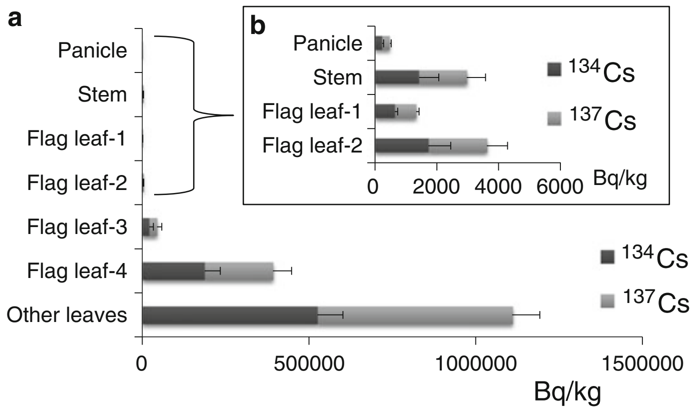

Fig. 2.1 The radiocesium concentration in the plant parts of the wheat cultivar "Kinuazuma" sampled on May 26th, 2011. (**b**) is an enlarged plot of (**a**), which contains the panicle, stem, FL1, and FL2. The *error bars* indicate the standard deviation (n = 3)

positron emitters; the 511-keV gamma rays made little contribution to the IP analysis compared with the beta rays (Noguchi and Suzuki 2001). The possible beta-ray emitters of fission products and their daughters are ^{131}I (half life: 8.03 days), ^{86}Rb (18.6 days), ^{134}Cs (2.07 years), ^{136}Cs (13.16 days), ^{137}Cs (30.08 years), ^{89}Sr (50.53 days), ^{90}Sr (28.79 years), ^{90}Y (64 h), ^{91}Y (58.51 days), ^{140}Ba (12.75 days), ^{140}La (1.68 days), among others. From the analysis using the germanium detector, it is likely that the autoradiographic images indicated ^{134}Cs and ^{137}Cs in this study. It is also possible that ^{90}Sr contributed but this was considered to be low because the ratio of ^{90}Sr/radiocesium(^{134}Cs+^{137}Cs) was approximately 1/600–1/19000 in soils sampled from the eastern part of Japan (http://radioactivity.mext.go.jp/ja/contents/6000/5808/24/194_Sr_0724.pdf: in Japanese). The contribution of ^{40}K, a natural beta-ray emitter, was considered to be trivial because there were no signals from a rice leaf without fallout contamination with 1 week contact with IP. There are a few reports of multi-nuclides being analyzed simultaneously and separately using shielding and image processing where the energies were significantly different from each other (Ishibashi et al. 2010). However, the high beta-ray energies emitted by both ^{134}Cs and ^{137}Cs meant that it was impossible to analyze them separately. Thus, it is suggested that the autographic images indicate the distribution of radiocesium.

The distribution of radiocesium is shown in Figs. 2.2 and 2.3. In Fig. 2.2, there are many active spots on FL4 and FL5, which were leaves that had already expanded by March 26th, 2011 (Table 2.1). On the other hand, there were hardly any spots on FL1, FL2, or FL3 (indicated as blue arrows), which had not emerged by March 26th, 2011 (Table 2.1). This indicated that the trend in the activities determined using the germanium detector and autoradiography were almost the same. The radiocesium distribution pattern captured by the imaging plate was also the same as the other cultivars ("Yukichikara" and "Abukumawase"), as indicated in Fig. 2.3 and Table 2.1.

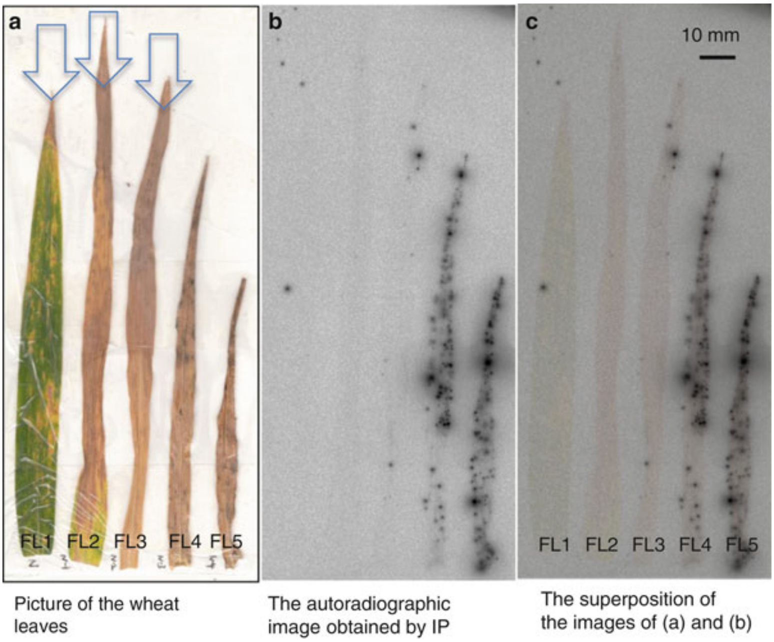

Fig. 2.2 Autoradiographic image of the wheat cultivar "Kinuazuma" obtained using an IP. The samples are from the same field as those shown in Fig. 2.1. "FL" indicates a flag leaf whereas the leaf positions are numbered from the top of FL to the bottom. The leaves indicated by *blue arrows* had not emerged by March 26th, 2011, which is also indicated in Table 2.1. The image (**c**) is a superposition of the images (**a**) and (**b**). There were three replicates for each analysis

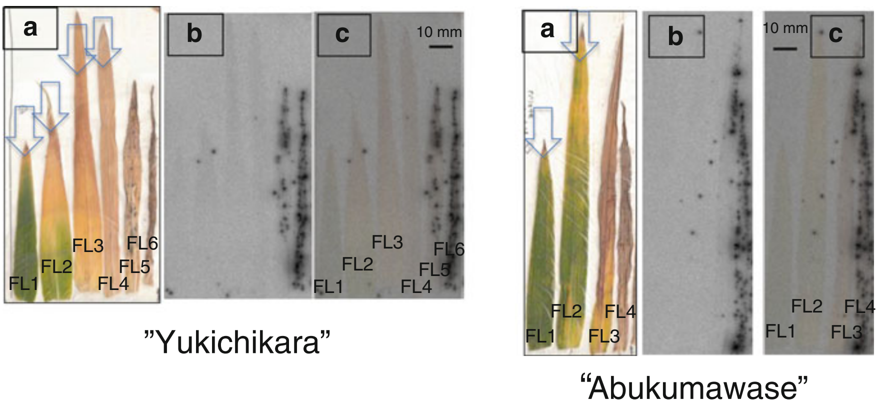

Fig. 2.3 Autoradiographic images of the wheat cultivars "Yukichikara" and "Abukumawase" obtained using an IP. The samples were collected on the same day as "Kinuazuma," which is shown in Fig. 2.2. "FL" indicates a flag leaf and the leaf position is numbered from top to the bottom. The leaves indicated by *blue arrows* had not emerged by March 26th, 2011, which is also indicated in Table 2.1. (**a**) Shows the visible image, (**b**) is the autoradiographic image obtained using an IP, whereas image (**c**) shows a superposition of the images (**a**) and (**b**). There were three replicates for each analysis

Table 2.1 The average number of expanded leaves of the wheat cultivars in March 2011 (n = 10)

	Number of expanded leaves		Number of leaf position from bottom					
	1 March	20 March	FL1	FL2	FL3	FL4	FL5	FL6
"Kinuazuma"	5.7	6.5	10	9	8	***<u>7</u>***	***<u>6</u>***	***<u>5</u>***
"Yukichikara"	6.4	7.4	13	12	11	10	9	***<u>8</u>***
"Abukumawase"	6.6	7.4	10	9	***<u>8</u>***	***<u>7</u>***	***<u>6</u>***	***<u>5</u>***

The data were provided by Arai Y., Nihei N., Takeuchi M. and Endo A. in Fukushima Agricultural Technology Centre
The leaf number which is underlined, italic and bold type was expanded at the day of 20 March 2011
FL flag leaf

Table 2.2 The growth and development of the wheats

Seeding date	Heading date	Ripening date	Plant height (cm) 21 December	28 March	Harvest time	No. of spikes (per m^2)	Grain weight (kg/a)
8 October	7 May	18 June	31	34	73	561	58
20 October	8 May	18 June	14	19	75	634	74
8 November	13 May	22 June	9	9	71	502	70
20 November	15 May	22 June	5	7	72	551	68

The data were provided by Arai Y., Nihei N., Takeuchi M. and Endo A. in Fukushima Agricultural Technology Centre
No. numbers

The analysis of the radiocesium concentration in each plant part showed that there was a significantly lower radiocesium concentration in the panicles that had not emerged in mid-March 2011, whereas a high concentration of radiocesium was detected on the leaves that had already expanded by mid-March 2011 when the fallout from the accident occurred. The spotty contamination on the leaves remained for up to 2 months after the fallout. These results suggested that the contamination, which was assumed to have been caused by radiocesium, was bound tightly to the surfaces of the leaves, whereas an extremely small amount of radiocesium entered the leaves and translocated to the panicles. The chemical form of caesium on the surfaces of leaves was not determined.

2.3.2 The Plant Size at the Time of the Fallout in Mid-March 2011 Was the Main Factor That Affected the Radiocesium Concentration in Grains

The growth parameters in the field are shown in Table 2.2. The plant height was more when the seeds were sown earlier. After the hibernation period, the plants from the October 20th sowing showed the most vigorous growth of the four different seeding dates. The differences in the heading and ripening dates for the different sowing dates varied by 8 and 7 days, respectively.

Table 2.3 The concentration of radiocesium in grains

	Radiocesium in grain after harvesting		
Seeding date	^{134}Cs (Bq/kg)	^{137}Cs (Bq/kg)	Total radiocesium (Bq/kg)
8 October 2010	129	154	283
20 October 2010	68	77	145
8 November 2010	11	13	24
20 November 2010	7	8	15

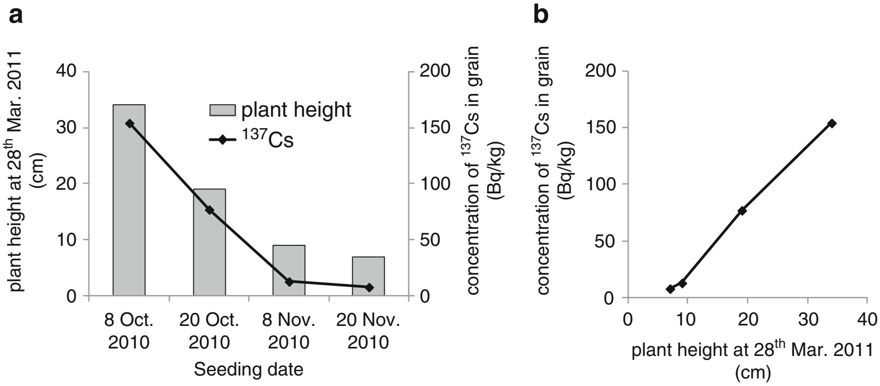

Fig. 2.4 (**a**) The relationship between the plant height on March 28th, 2011 (shown as *grey bar*) and the concentration of ^{137}Cs in the grains during harvest (shown as a *polygonal line*). (**b**) Plot of the plant height vs the concentration of ^{137}Cs in the grains. These data were provided by Arai Y., Nihei N., Takeuchi M. and Endo A. at the Fukushima Agricultural Technology Centre

The concentration of radiocesium in the grains decreased with the date of sowing (Table 2.3 and Fig. 2.4). The relationship among the sowing day, the plant height in mid-March 2011, and the ^{137}Cs concentration in the grain after harvesting is shown in Fig. 2.4. The ^{137}Cs concentration in the grain after harvesting was correlated with the plant height on May 28th, 2011 (Fig. 2.3b). The results strongly suggest that the radiocesium in the grain was derived from the leaves and stems that had already emerged by mid-March 2011, when the fallout occurred.

Komamura et al. (2006) reported that wheat grains in Japan were directly contaminated by fallout from the Chernobyl disaster and the concentration of ^{137}Cs in grain was higher than that in rice because heading had not occurred in rice plants by the end of April, when the disaster occurred. In wheat, there was a strong correlation between the date of head emergence and the concentration of ^{137}Cs in the grains (Komamura et al. 2006). For radiocesium, these reports indicate that there was a greater impact from the direct contamination of grains than indirect contamination which occurred through its transport from the soil to the edible parts of plants. Thus, the present study demonstrated the behavior of radiocesium in the leaves and/or stems and that it was transferred to the grains. This mechanism was the primary factor that determined the relatively high concentration of radiocesium in the grains produced in 2011.

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Chapter 3
Radiocesium Absorption by Rice in Paddy Field Ecosystems

Keisuke Nemoto and Jun Abe

Abstract Although most of the radiocesium fallout that deposited in paddy fields after the Fukushima nuclear disaster in March 2011 was expected to be bound to clay in the soil resulting in a very low soil-to-plant transfer function, a radiocesium contamination level of >500 Bq/kg was detected in brown rice grown in several hilly areas of Fukushima Prefecture in the autumn of the same year. The likely source of the radiocesium was fallout deposited on organic matter in the paddy fields and litter in mountain forests, from which runoff water flowed into irrigation channels that ultimately lead to the paddy fields. This problem appears to have been caused by conditions specific to lowland rice paddy fields, which are wetland ecosystems. Integrated studies of the soil, water, and plants from an ecological viewpoint are necessary to understand the mechanism of radiocesium absorption by rice before commercial rice production in the affected areas can be resumed.

Keywords Irrigation water • Paddy field ecosystems • Radiocesium • Rice (*Oryza sativa* L.)

Abbreviations

Cs	Cesium
N	Nitrogen

K. Nemoto (✉) • J. Abe
Graduate School of Agricultural and Life Sciences, The University of Tokyo,
1-1-1 Yayoi, Bunkyo-ku, Tokyo 113-8657, Japan
e-mail: unemoto@mail.ecc.u-tokyo.ac.jp; junabe@agrobio.jp

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3.1 Introduction

The nuclear disaster in Fukushima in March 2011 released considerable amounts of various radionuclides and contaminated extensive areas of farmlands. Radiocesium comprised the majority of the radionuclides released and special attention needs to be paid to ^{137}Cs because of its long half-life.

In general, plants absorb radioactive materials: (1) directly from the surface through their aerial parts or (2) through root uptake. After the radioactive material was released into the atmosphere, it adhered to the surfaces of the aerial parts of plants where direct absorption occurred, which was the major source of contamination in plants immediately after the release of the radioactive material. The radioactive material then entered the environment and moved into the soil, where it was absorbed through the roots resulting in long-term contamination of crops. The level of root absorption is greatly affected by the behavior of the radioactive material in the soil and the soil-to-plant transfer factor depends on the specific soil type because radioactive materials such as cesium are strongly bound to the soil granules (Yamaguchi et al. 2012). Because the typical farm soil in the affected area (mainly Fukushima) was a gray lowland soil, which binds cesium strongly, the radiocesium contamination of various crops through root absorption was much lower than the provisional regulation level (500 Bq/kg) in most cases during 2011.

A low level of radiocesium contamination was also expected in rice (*Oryza sativa* L.). Half way through the investigation of radioactive contamination (pilot survey) of rice in mid-September of 2011, the radiocesium concentration in brown rice was below the detection limit at most test sites and the highest contamination level was approximately 1/4th of the provisional regulation level. However, in the subsequent investigation, the radiocesium concentration was close to or >500 Bq/kg in many samples of brown rice from several localities in the hilly areas of Fukushima Prefecture (e.g., Obama district in Nihommatsu City, Onami district in Fukushima City, and Oguni district in Date City), which surprised the whole of Japanese society. The radiocesium absorption level in the rice grown in these paddy fields was extraordinarily—100 times or more than that in the flatland areas of Fukushima Prefecture such as Koriyama City. The mechanism underlying this unusual absorption was unclear, and a rapid investigation is necessary before the crop production can be resumed in the affected area. In this paper, we will summarize our most current knowledge about this problem.

3.2 "Seasonality" in the Radiocesium Absorption Level of Rice

The temporal pattern of radiocesium absorption by plants (i.e., the time course of absorption throughout the plant growth stages) can be estimated from the spatial distribution of the absorbed material in the plant body, provided the material has low mobility in the living plant (Tanoi et al. 2011). We used this approach to investigate the distribution of radiocesium in rice plants.

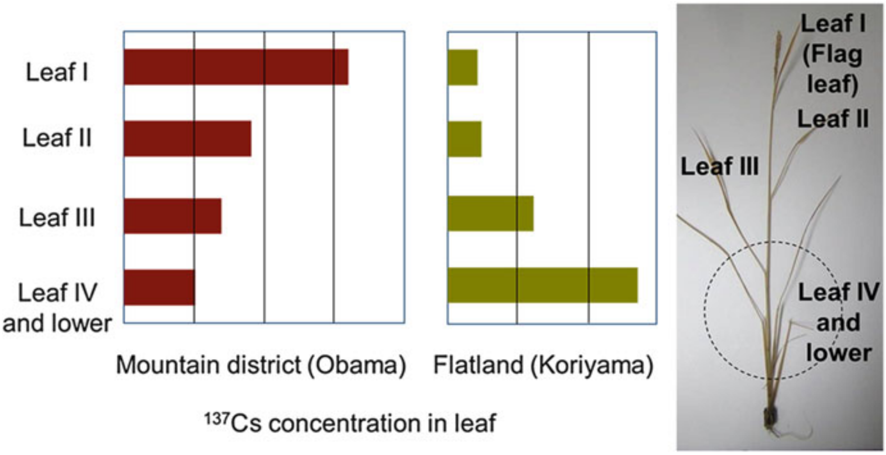

Fig. 3.1 Distribution of radiocesium in rice. The ^{137}Cs concentration was evaluated using an imaging plate method (Tanoi et al. 2013)

The radiocesium concentration in leaves decreased acropetally from the lower leaves (old leaves) to the upper leaves (new leaves) in rice plants grown in a paddy field at Fukushima Agricultural Technology Center in Koriyama City, where the radiocesium concentration of brown rice was only approximately 5 Bq/kg. In contrast, the radiocesium concentration increased acropetally in the successive leaves of rice plants grown in a paddy field in Obama, where 470 Bq/kg radiocesium was detected in brown rice (Fig. 3.1). This distribution pattern suggests that considerable amount of radiocesium was absorbed by the rice roots in midsummer in Obama when the upper leaves were growing actively.

The most likely explanation of these findings is the promotion of organic matter decomposition under high temperatures. Organic matter decomposition is enhanced greatly in paddy fields when the air temperature exceeds 30 °C, which could have released radiocesium from radiocesium-contaminated organic matter. Various types of organic matter such as crop residues and weeds could have been sources of radiocesium in the paddy fields in Obama, Onami, and Oguni (Shiozawa 2012). Furthermore, radiocesium derived from litter deposited in mountain forests may have entered the mountain runoff and flowed into the paddy fields where it was absorbed by rice. In fact, many of the paddy fields that exceeded the provisional regulation level used mountain runoff as an irrigation source because it is very rich in nutrients such as potassium and magnesium (Fig. 3.2). Although the concentration of ionized radiocesium in the mountain runoff was rather low, the radiocesium concentration in the suspended solids (colloid) increased to several Bq/L after rainfall (unpublished data). Thus, it is important to determine how the suspended organic material decomposes to release ionized radiocesium into the irrigation water.

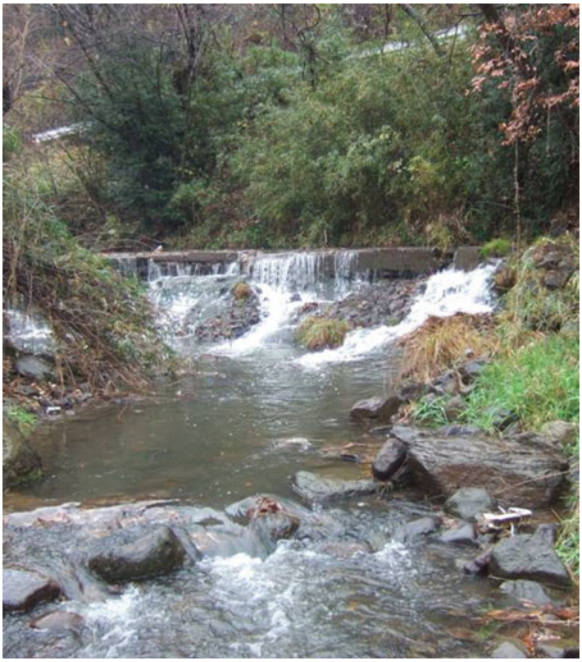

Fig. 3.2 Mountain runoff water flowing into a river that is often used for paddy field irrigation in mountain districts

3.3 Radiocesium Absorption Reflects the Features of the Paddy Field Ecosystem

In paddy field ecosystems, the accumulation and decomposition of organic matter and the input of irrigation water are essential factors that determine the sustainability of production. Between the autumn and the spring, crop residue decomposition is suppressed by low temperatures and anaerobic conditions, whereas intensive organic matter decomposition occurs in summer, resulting in the release of nutrients required for plant growth in abundance. Irrigation water also provides an abundance of natural nutrients; runoff from mountain forests is particularly rich in potassium and magnesium. Our ancestors understood these ecological consequences based on the experiences of generations of farmers and they sited their paddy fields in locations with a rich forest in the upper stream catchment. There is a saying in Japanese that “Rice yield depends on the soil, wheat and barley on the manure,” which suggests the sustainability of paddy ecosystems. Our hypothesis about radiocesium contamination of rice predicts that the mechanism of contamination is inextricably linked to the unique features of nutrient flow and cycling in paddy field ecosystems (Fig. 3.3).

The flooding of paddy fields may enhance radiocesium absorption in some cases. It is well known that direct cesium absorption from water is much easier compared with absorption from soil. We conducted a hydroponic culture experiment where the culture solution contained radiocesium released by the nuclear disaster at 1 Bq/L

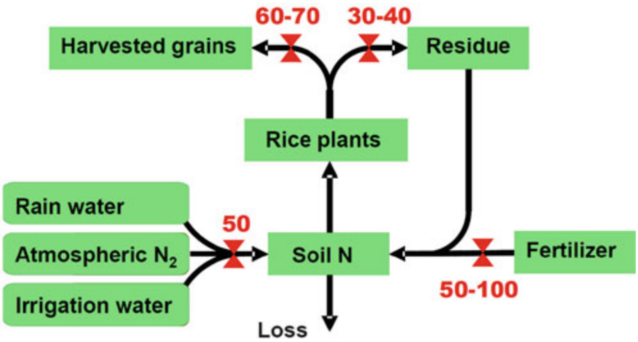

Fig. 3.3 Nutrient flow and cycling in the paddy field ecosystem. This diagram shows the nitrogen cycle in a typical paddy field in Japan. The numbers indicate the level of nitrogen input or output (kg/ha) per year

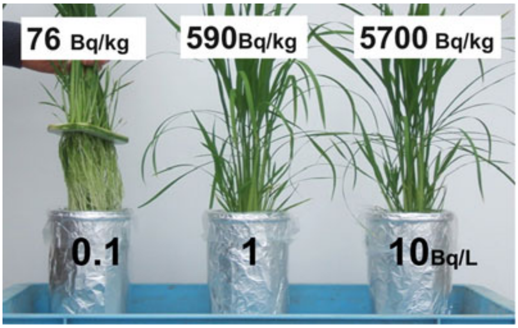

Fig. 3.4 Uptake of radiocesium by rice in hydroponic culture. Radiocesium fallout was collected from wheat leaves affected by the Fukushima nuclear disaster and the fallout was added to the hydroponic solution at three different concentrations. The concentration of radiocesium that accumulated in the rice shoots was measured using a germanium semiconductor detector after 26 days of cultivation

and rice plants accumulated radiocesium concentrations of almost 600 Bq/kg (dry matter) in their shoots (Fig. 3.4). This absorption level was 1,000 times higher than that from soil. This unusually high absorption level was undoubtedly because of the artificial culture conditions where the whole root system was exposed to the solution medium during hydroponic culture, which is not expected to occur in actual paddy fields. Nevertheless, we estimated that absorption of radiocesium by roots directly from water may have occurred before the radiocesium bound to soil, given that the water permeability of the soil was very low in most paddy fields where the radiocesium concentration exceeded the provisional regulation level (500 Bq/kg) (Shiozawa 2012). In these conditions, the water and/or soil in the paddy fields may have remained stagnant with radiocesium-contaminated water for some period, which may have caused root absorption. There were also reports of radiocesium contamination in wasabi (Japanese horse radish; *Eutrema wasabi*) from some areas, which is often cultured using the water runoff from mountains. This also suggests the possibility of radiocesium absorption from water by roots.

Given the remarkably high efficiency of radiocesium absorption from water by roots, particular attention should be given to radiocesium contamination of the water used for commercial hydroponic vegetable production, which is often recommended as a way of avoiding radiocesium transfer from the soil to vegetables.

The effect of the exchangeable potassium concentration in the soil is another key factor. It is widely known that a low potassium concentration in the soil enhances cesium absorption by plants. A joint research project in Onami district by the Ministry of Agriculture, Forestry, and Fisheries (MAFF) and Fukushima Prefecture reported a low concentration of exchangeable potassium in paddy fields where the radiocesium concentration was high in brown rice. This report suggested that the low exchangeable potassium concentration in the paddy soil may have reinforced the radiocesium absorption by rice roots in areas where brown rice contamination exceeded the provisional regulation level. Our pot experiment using contaminated paddy soil from Obama also showed that the radiocesium uptake by rice seedlings was reduced to approximately 1/10th after the application of potassium chloride, which indicated the effectiveness of potassium fertilizer application to contaminated paddy fields as a countermeasure for absorption of radiocesium by roots.

However, we would like to point out that the conventional rice cultivation methods used in the problem area are still quite reasonable and effective given the low input requirements of sustainable agriculture. Local farmers intensively utilized the rich mineral nutrients in runoff water to produce highly palatable rice while minimizing the use of chemical fertilizers. Thus, rice plants absorbed sufficient potassium from the paddy fields, although the potassium concentration of the soil was low. However, the nuclear disaster destroyed the well-intended efforts of farmers who wanted to produce sustainable, high quality rice in Fukushima.

3.4 Behavior of Radiocesium in Organic Matter

As discussed above, the problem appeared to be caused by the specific conditions in lowland rice paddy fields, which are wetland ecosystems. Many useful agricultural and plant studies were reported after Chernobyl (e.g., Ehlken and Kirchner 2002), but the experience and knowledge acquired from the areas affected by the Chernobyl disaster are not directly applicable to solving the rice production issues in Fukushima. Thus, original perspectives and approaches based on agricultural research in monsoonal Asia are required to prevent radiocesium contamination of rice, which are different from those required in upland farm areas. To understand the mechanism of radiocesium contamination of rice and solve the problem, we need to analyze the flow of radiocesium through forests, mountain streams, and paddy fields.

In the hydroponic experiment mentioned above, we extracted radiocesium from the fallout deposited on field-grown wheat leaves. Unexpectedly, we found that the fallout was relatively insoluble and only a small percentage of the radiocesium could be

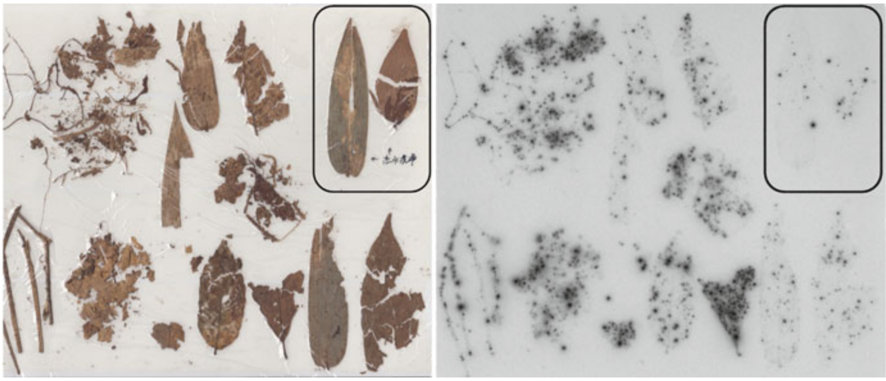

Fig. 3.5 Radioactivity in fallen leaves collected from the forest litter in Nihonmatsu-shi 8 months after the Fukushima disaster. *Left*, leaves; *right*, radioactivity detected using the imaging plate. The two leaves in the box were rinsed before measurements but they still contained scattered radiocesium

extracted by a boiling water treatment followed by nitrate leaching. We have very little knowledge about this fallout, including its chemical form and properties, but huge amounts of this relatively insoluble radioactive fallout are still bound to organic matters in paddy fields and litter in mountain forests (Fig. 3.5). Because it will take several years before the litter on the forest floor decomposes completely, the release of radiocesium from the litter to the environment will probably last for a long period. Thus, it is necessary for agricultural, soil science, plant nutrition, and forestry scientists to collaborate in the continued long-term monitoring of radiocesium in regional ecosystems.

3.5 Can Breeding Resolve the Problem?

We have discussed the possible mechanism of radiocesium contamination in rice from an ecological perspective; besides, breeding new rice cultivars that absorb less radiocesium is another important approach that may solve the problem. Thus, we screened rice varieties to acquire basic information relevant to future breeding efforts. Over 100 rice varieties were grown using highly contaminated soil and their radiocesium uptake capacities were measured during the vegetative stage. The radiocesium uptake during the vegetative phase was generally higher in *japonica* varieties compared with *indica* varieties. The only exception was Pokkali rice, a famous salt-tolerant *indica* variety, which had a higher radiocesium uptake capacity than any of the *japonica* varieties we tested. However, the overall range of genetic variation in the radiocesium uptake capacity was only threefold.

We also need to study the radiocesium accumulation level during the reproductive stages before we conclude the study; however, the present results suggest that the genetic variation in radiocesium uptake by rice is not sufficiently high to breed a new variety that could resolve the problem. Therefore, the integration of breeding with other countermeasures such as cultivation methods and civil engineering with special attention to ecophysiological and environmental aspects will be required before rice culture in the affected areas can be resumed.

3.6 The Diagnosis of Radiocesium Absorption in Individual Paddy Fields

It appeared that the high levels of radiocesium contamination in brown rice were caused by a complex interaction between local factors and features of the individual paddy fields. The local factors included large amounts of radioactive fallout and contaminated water that tended to accumulate in areas with mountainous geography, whereas the paddy field-specific factors included the types of organic matter and runoff water that provided a source of radiocesium, the exchangeable potassium content, the type of clay in the soil, and water percolation. Furthermore, because the decomposition rate of organic matter can differ in the leaf litter produced by various trees, the end of the radiocesium contamination risk for rice will also depend on the organic matter type.

Solving the problem of radiocesium contamination in rice demands the clarification of general factors in affected areas and the detection of radiocesium absorption in individual paddy fields. Unfortunately, detailed data were not available for individual paddy fields during 2011 because the rice harvested from individual paddy fields was gathered when the brown rice contamination level exceeded 500 Bq/kg. Thus, it is necessary to conduct further field studies to diagnose individual paddy fields. We are conducting rice cultivation field experiments in some areas of Date City this year (2012), where radiocesium contamination levels exceeding 500 Bq/kg were detected in some brown rice samples in 2011. We intend to diagnose individual paddy fields to support the resumption of rice production by native farmers in these areas.

Acknowledgments The studies reported in this chapter were performed as collaborative research by three groups from the Graduate School of Agricultural and Life Sciences of the University of Tokyo (Laboratory of Radioplant Physiology, Laboratory of Land Resource Science, Laboratory of Crop Ecology and Morphology) and Fukushima Agricultural Technology Centre.

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Chapter 4
Cesium Uptake in Rice: Possible Transporter, Distribution, and Variation

Toru Fujiwara

Abstract Here, we review cesium uptake and accumulation in rice. Cesium is an alkaline metal, and its uptake is affected by potassium nutrition. Several transporters for cesium have been described. The distribution of cesium in the rice plant body differs from that of potassium, suggesting differential transport/storage mechanisms for cesium and potassium in rice. Cesium concentration in rice differs among cultivars, and it would be possible to determine the gene(s) responsible for cesium uptake/accumulation in future. This knowledge will form the foundation for the development of low radiocesium-accumulating cultivars.

Keywords Accumulation • Cesium • Cultivars • Potassium • Transporter • Uptake

4.1 Introduction

After the Fukushima Daiichi nuclear power plant accident in March 2011, radioactive nuclides were emitted and widely diffused in the environment, including across agricultural areas in Japan. Radiocesium, one of the emitted nuclides, has relatively long half lives (2.06 years for ^{134}Cs, 30.2 years for ^{137}Cs), and contamination of the environment and foods with radiocesium has been and will be of concern for a long time. Radiocesium deposited in soil sticks to soil particles; however, a small portion is redistributed into a soil solution that is readily absorbed by plants. Uptake and transport of cesium (Cs) by plants is an important determinant of the degree of radiocesium contamination of foods. Elucidating the uptake and transport processes involved and their mechanisms is important for minimizing the contamination of agricultural products. In this short article, aspects of Cs transport are reviewed with a major focus on rice, and future perspectives are also discussed.

T. Fujiwara (✉)
Graduate School of Agricultural and Life Sciences,
The University of Tokyo, Tokyo, Japan
e-mail: atorufu@mail.ecc.u-tokyo.ac.jp

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4.2 Possible Transporters Involved in Cs Uptake

Several factors are likely to affect radiocesium uptake from the soil. A very good review of such factors was published recently (Yamaguchi et al. 2012). The radiocesium from the Fukushima accident was mostly present on the soil surface. Given the uneven distribution of radiocesium, the root architecture is likely to affect its uptake. Plants with roots near the soil surface may absorb more radiocesium than those with deeper root systems. Recently, a gene involved in the determination of root architecture in rice, namely "deep roots and shallow roots," has been identified (Uga et al. 2009). However, to my knowledge, the relationship between root architecture and radiocesium uptake has not yet been established, despite this being an important aspect of radiocesium uptake from the soil by rice. Lateral distribution of radiocesium will change over time because of natural and human activities, and depending on the distribution, desirable root architectures may also change over time.

Cs is an alkaline metal that is taken up by plants in the form of Cs^+. Potassium (K) is an essential plant nutrient that is a major component of fertilizers. Cs transport has been discussed in conjunction with K transporters. It is well established that the mechanism of Cs absorption by a root is similar to the mechanism of K absorption, following Michaelis–Menten kinetics (White and Broadley 2000). Inhibition of Cs absorption by the addition of K has also been shown in hydroponics and soil cultures (Smolders and Tsukada 2011), suggesting that Cs is taken into plants through K transporters. It is also known that the antagonism of K in Cs absorption is restricted, and under excessive Cs/K ratios, the effect is very much limited.

A number of K transporters have been identified in plants, and Cs transport has been demonstrated for several of these K transporters. The KUP/HAK/KT family, the family of high-affinity K^+ transporters, has been shown to participate in Cs absorption (White and Broadley 2000). Yeast expressing *Arabidopsis thaliana* KUP1 absorbs K^+, and its activity is affected by the presence of Cs^+ (Fu and Luan 1998). Under high K conditions, *v*oltage-*i*nsensitive *c*ation *c*hannels (VICCS), independent cation channels, participate in Cs absorption (White and Broadley 2000). In the case of *A. thaliana*, VICCs are coded by the AtCNGC and AtGLR gene families. Although to my knowledge, no experimental evidence is currently available, rice homologs of the abovementioned transporters are likely to play a role in Cs uptake and distribution. Notably, these transporters represent only a subset of all the transporters involved in Cs uptake and transport in plants. As discussed below, Cs and K distribution within rice plants differs. This could possibly be attributed to differences in the relative transport of K and Cs among transporters or to the presence of Cs-specific transporters.

After absorption by roots, Cs is loaded to the xylem to reach the aerial portions of plants; this is followed by phloem transport to reach the grains. Different types of transporters are involved in these processes. In the case of K, the loading of K is mediated by SKOR; however, its involvement in Cs transport has not been demonstrated.

4.3 Cs-Specific Transporters?

Plants grown in soil contain a number of nonessential elements such as Cs. Nonradioactive Cs is present in the environment and foods. Cs is not an essential element for plants or animals. Cs is not very toxic to plants and animals compared with other heavy metals such as cadmium (Cd) and arsenic (As). It has long been believed that plants do not have specific transporters for nonessential toxic elements and that their uptake is mediated by transporters for essential nutrients that are chemically similar. For example, As in the form of arsenate is taken up by phosphate transporters (Zhao et al. 2009), and arsenite is taken up by aquapolin-like proteins (Ma et al. 2008; Kamiya et al. 2009). Cd transport is mediated by IRT1, an iron transporter (Nakanishi et al. 2006), and more recently, Os Nramp5 has been reported to function as a manganese (Mn) and Cd transporter (Ishimaru et al. 2012; Sasaki et al. 2012). A few years ago, a sensational report (Ueno et al. 2010) was published in PNAS in which the authors claimed that rice HMA3 is a transporter that is highly specific to Cd. This claim was based on their analysis of transgenic plants, which reportedly showed altered Cd accumulation without changes in other major metal contents. HMA3 is present on the vacuolar membrane and regulates Cd transport from roots to shoots. This claim of a Cd-specific transporter led to increased interest in the field, and the authors also discussed possible reasons why Cd-specific transporters had evolved. However, according to one of the authors, their manuscript did not include several transgenic plants overexpressing Os HMA3 at a much higher level than the two transgenic lines reported in the manuscript. These strong overexpressors had reduced Zn concentration in the grains but were not included in the manuscript. The authors claimed that Os HMA3 is highly specific to Cd. One of the authors presented further analysis of the transgenic plants reported in the PNAS paper at an annual meeting of the Japanese Society of Soil Science and Plant Nutrition in Tottori, Japan in September 2012. The concentrations of Zn in the roots of transgenic plants were shown to be reduced, further suggesting that OsHMA3 is not actually Cd-specific at all. The general concept of toxic metals being taken up by plants through transporters for essential elements still stands, and it is probably also the case for Cs transport in rice. Notably, however, the relative specificity for Cs differs among K transporters, and this may be a basis for the differential distribution of Cs and K in rice plants, as described below.

4.4 Cs Distribution in Rice

After being taken up by rice, Cs is transported and distributed to the aerial portions of plants. Radiocesium distribution was studied Tsukada et al. (2002a, b). In their studies, the patterns of radiocesium derived from fallout were examined in field-grown rice. They found that 65% of the radioactivity was recovered in straws, 10% in polished rice, 10% in bran, and 10% in husks. The overall distribution patterns were somewhat similar to those for K; however, the relative concentration differed among tissues. Cs tends to accumulate in old leaves rather than young leaves. Such differential distribution can be attributed to differing specificity of the transporters. Alternatively, Cs and K may differ in their ability to bind to materials in living cells.

4.5 Variation in Rice Cs Concentrations Among Different Cultivars

Needless to say, it is crucial to reduce radiocesium contamination of foods. One practical way to reduce contamination is to use cultivars that accumulate less Cs in their grains. For this purpose, it is important to understand that there is variation in Cs uptake among plant species and cultivars. In the case of Cd, a large variation in Cd concentration in the leaves and grains among different cultivars of rice was used to identify genes important for Cd transport and to generate rice cultivars with reduced Cd accumulation or high Cd accumulation for phytoremediation.

In *A. thaliana*, quantitative trait locus (QTL) analysis of ecotypes with different Cs concentrations was used to identify the genetic locus that regulates Cs uptake in plants. Cs accumulation was found to be mainly regulated by QTLs in chromosomes 1 and 5 (Kanter et al. 2009). AtCNGC1 is very close to QTL on the 5th chromosome, and the amino acid sequence of CNGC1 reportedly differs between cultivars, suggesting that CNGC1 participates in Cs absorption (White and Broadley 2000).

A large variation in Cs concentration has been reported among different plant species, including crops and vegetables. Rice is a low accumulator of Cs. The transfer factor for radiocesium from the soil to rice grains is mostly in the range of 0.001–0.0002. This means that when rice is grown in soils of 5,000 Bq/kg it will accumulate 5–25 Bq/kg radiocesium in its polished grains, and I believe that this is below the provisional regulation level (500 Bq/kg) as well as the new standard (100 Bq/kg) for radiocesium contamination in food set by the Japanese government.

To determine the degree of variation among rice cultivars, Ishikawa et al. measured the concentrations of nonradioactive Cs among different specimens from the World Rice Core Collection obtained from Tsukuba (Yamaguchi et al. 2012). There was a 30-fold difference in Cs concentrations in the grains of these different rice cultivars. Our group also conducted a field study in Fukushima Prefecture in 2011 (Fig. 4.1), and we found large variation among cultivars (more than 20-fold). Notably, the difference observed under field conditions will also be caused by the uneven distribution of Cs in the field; however, the observed difference was far greater than the uneven distribution of Cs that is normally observed, suggesting a wide variation in Cs concentration among rice cultivars. It is expected that this variation will facilitate the identification of the gene responsible for Cs transport in rice and that this can be used to reduce Cs concentrations in the grains in future.

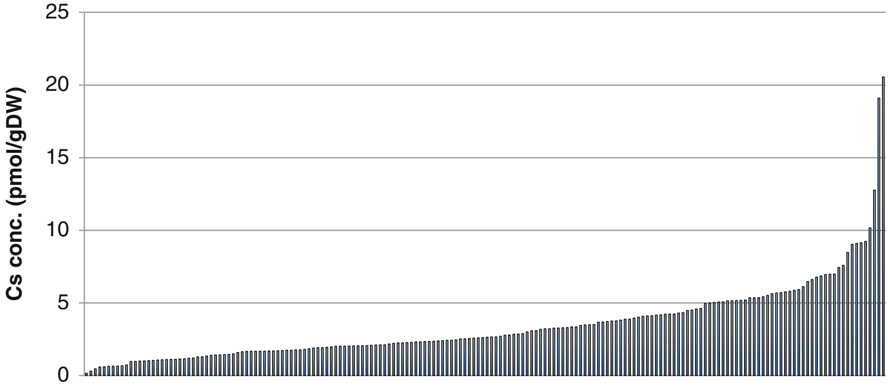

Fig. 4.1 Distribution of cold Cs (Cs-133) concentration in different rice cultivars grown in a rice field in Fukushima. More than 100 cultivars representing Japanese and overseas were grown in a rice field in Fukushima Prefecture in 2011. Cold Cs (Cs-133) concentration in brown rice was determined. Concentration was arranged in an ascending order. Concentration of each cultivar is shown by a *bar*. A wide range of difference in Cs concentration was observed

4.6 Future Perspectives

Many studies have established the physiological characteristics of Cs uptake, transport, and accumulation in plants, including rice; however, the molecular mechanisms of Cs transport and accumulation in rice remain largely unknown. Several transporters have been identified in *A. thaliana* that affect Cs transport; however, studies on rice are necessary to reduce radiocesium uptake and transport into grains. As seen in previous studies on toxic elements, it is most likely that the transporters will be identified through molecular and genetic approaches. The relatively large variation in Cs accumulation in rice grains may allow us to identify the transporter and/or other genes involved in Cs transport and accumulation.

As mentioned above, Cs transporters responsible for Cs uptake and distribution are most likely to be transporters of (an) essential element(s). Cs transporters identified in *A. thaliana* are K transporters, and if such a gene is mutated, it is likely that K transport would be affected. However, in the case of Cd accumulation in rice, Ishikawa et al. successfully reduced the Cd concentration in grains by screening mutant Koshihikari for reduced Cd concentration, and this did not require molecular understanding of the transport process. It turned out that the gene disrupted in their mutant that caused the reduced Cd accumulation in the grains was Os Nramp5, an Mn transporter. According to their press release and their presentation at the annual meeting of the Japanese Society of Soil Science and Plant Nutrition, the growth of this particular mutant line in the field was identical to that of the wild-type Koshihikari. I expect that this mutant line will be cultivated in farms in Japan in the near future and that this will reduce the risk of Cd uptake by humans. Notably, the Tos17 disruption line of Os Nramp5 grows poorly in the field (Sasaki et al. 2012), suggesting the importance of selecting an appropriate allele for practical purposes.

It should be possible to screen mutant elite Japanese cultivars for reduced Cs accumulation in grains using a similar approach. This may allow us to identify the responsible gene(s) using a forward genetic approach, and with such basic understanding, we will be able to reduce Cs contamination not only in rice but also in other plant/vegetable species.

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Chapter 5
Time-Course Analysis of Radiocesium Uptake and Translocation in Rice by Radioisotope Imaging

Natsuko I. Kobayashi

Abstract The pattern of real-time ^{137}Cs uptake by plants was visualized using a macroscopic real-time radioisotope imaging system. We found that ^{137}Cs was easily taken up by rice plants only when it was dissolved in a liquid medium. In contrast, only a small amount of ^{137}Cs was taken up when the same liquid medium containing ^{137}Cs was added to the soil and supplied to the rice plants. This result demonstrates the intensive Cs adsorptive property of the soil. When rice was grown hydroponically in K-sufficient environment followed by K withdrawal for 2 days, ^{137}Cs was taken up easily compared with the rice without K deficiency. The application of K was shown to be an effective method for preventing radiocesium uptake by rice plants. However, K-deficiency was found to have little effect on the xylem loading process of Cs. Understanding the Cs translocation processes and evaluating the factors affecting each process based on experimental evidence could facilitate the development of an agricultural technique to reduce the radiocesium content in the edible parts of plants.

Keywords Imaging • K deficiency • Radiocesium • Rice • Tracer experiment • Xylem loading

Abbreviations

Cs	Cesium
IP	Imaging plate
K	Potassium

N.I. Kobayashi (✉)
Graduate School of Agricultural and Life Sciences, The University of Tokyo, 1-1-1 Yayoi, Bunkyo-ku, Tokyo 113-8657, Japan
e-mail: anikoba@mail.ecc.u-tokyo.ac.jp

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Macro RRIS	Macroscopic real-time radioisotope imaging system
P	Phosphorus
PSL	Photo-stimulated luminescence
ROI	Region of interest

5.1 Introduction

Limiting radiocesium uptake by plants from soil is important for reducing the ingestion of this pollutant. In addition to taking measures for preventing further entry of radiocesium into farmlands, inhibiting the soil-to-plant transfer of radiocesium by developing appropriate agricultural techniques may be effective. Cs belongs to the alkali metal group and has chemical and physical properties similar to other alkali metals. In plants, Cs is known to show similar behavior to K, which is an alkali metal and an essential macronutrient for organisms. Plants take up Cs through K^+ transporters and the K^+ channel pathway, and increase in the K concentration in a culture medium reduces the Cs uptake by plants due to the competition between K^+ and Cs^+ (Zhu and Smolders 2000). The exchangeable K^+ concentration in paddy soil showed a significant negative correlation with the radiocesium content in brown rice produced in the Fukushima region in 2011 (http://www.pref.fukushima.jp/keieishien/kenkyuukaihatu/gijyutsufukyuu/05gensiryoku/240112_tyukan.pdf). Thus, it was assumed that K fertilization to maintain the soil K^+ concentration at approximately 250 mg/kg, which is the recommended level suggested by the government, would be an effective technique for reducing the soil-to-plant transfer of radiocesium.

Preventing radiocesium accumulation, particularly in the edible parts of plant, may further reduce the risk of food contamination. Cs behaves similar to K in plants, as mentioned above. However, it is not known whether Cs behaves exactly the same as K in plants because it has not been sufficiently investigated and its behavior may differ depending on the species. A different Cs:K concentration ratio among tissues has been found in several grasses (Menzel and Heald 1955) and trees (Gommers et al. 2000; Robison et al. 2009), whereas the Cs:K concentration ratio among tissues was almost uniform in some tropical trees (Staunton et al. 2003). Furthermore, Robison et al. (2009) reported that the radiocesium distribution pattern within a coconut tree was altered by K fertilization. In *Arabidopsis*, mutants lacking individual K transporters had a specific Cs:K concentration ratio in their shoots (Hampton et al. 2005), indicating a relationship between specific properties of the K transport system and the Cs distribution pattern.

According to these studies, it may be necessary to understand the primary processes of Cs translocation separately within plants, such as the root uptake, xylem loading, and remobilization, before elucidating the Cs translocation mechanism and developing an agricultural technique for reducing the radiocesium content in edible parts. Therefore, we performed several types of ^{137}Cs tracer experiments in our laboratory.

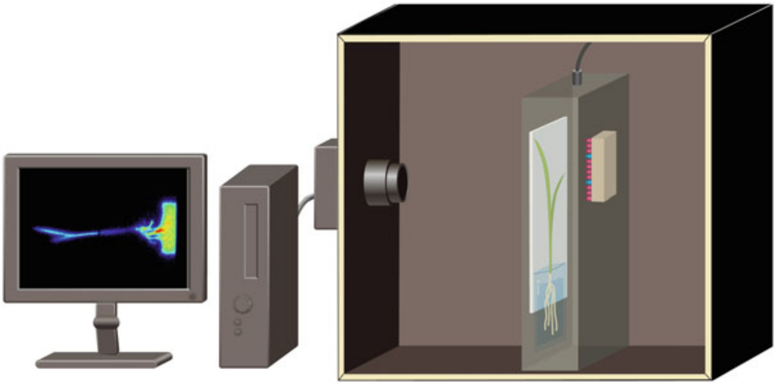

Fig. 5.1 Macro RRIS. The system consisted of a plant box with a fiber optic scintillator, a signal detection and multiplication component, and the monitor. The system had a viewing area of 10 cm × 20 cm

5.2 Application of Macro RRIS for Quantification of ^{137}Cs Signals

To understand each translocation process separately, the real-time imaging analysis of ^{137}Cs migration through a whole plant could provide convincing information based on the non-destructive and continuous analysis of a single plant. Therefore, we evaluated the performance of a macroscopic real-time radioisotope imaging system (macro RRIS; Fig. 5.1; Nakanishi et al. 2009; Kanno et al. 2012) for ^{137}Cs determination. The results showed that macro RRIS, using an Al sheet to cover the scintillator surface as light shielding when detecting ^{137}Cs in illuminated conditions, could be used to image a spot containing 5 Bq ^{137}Cs in 1 μl during an integration period of 5 min (Fig. 5.2a). The detection performance of macro RRIS was lower than that of an imaging plate (IP) in terms of its sensitivity in the low radioactivity range (<10 Bq) and quantitative performance in the high radioactivity range (>400 Bq), but we demonstrated that macro RRIS could be applied efficiently for real-time imaging analysis using ^{137}Cs within the proper radioactivity range (Fig. 5.2b).

5.3 Uptake of ^{137}Cs by Rice Plants from Liquid or Soil Media

The strong adsorption of radiocesium by soil components, particularly clay, is known to occur immediately after radioactive fallout. This was clearly demonstrated by the imaging analysis (Fig. 5.3). When ^{137}Cs was added to the liquid medium, a large amount of ^{137}Cs, which was sufficient to produce a ^{137}Cs distribution image during an integration period of 10 min, was absorbed by the rice root and translocated to the shoot (Fig. 5.3a). In contrast, when the same amount of ^{137}Cs was added to the

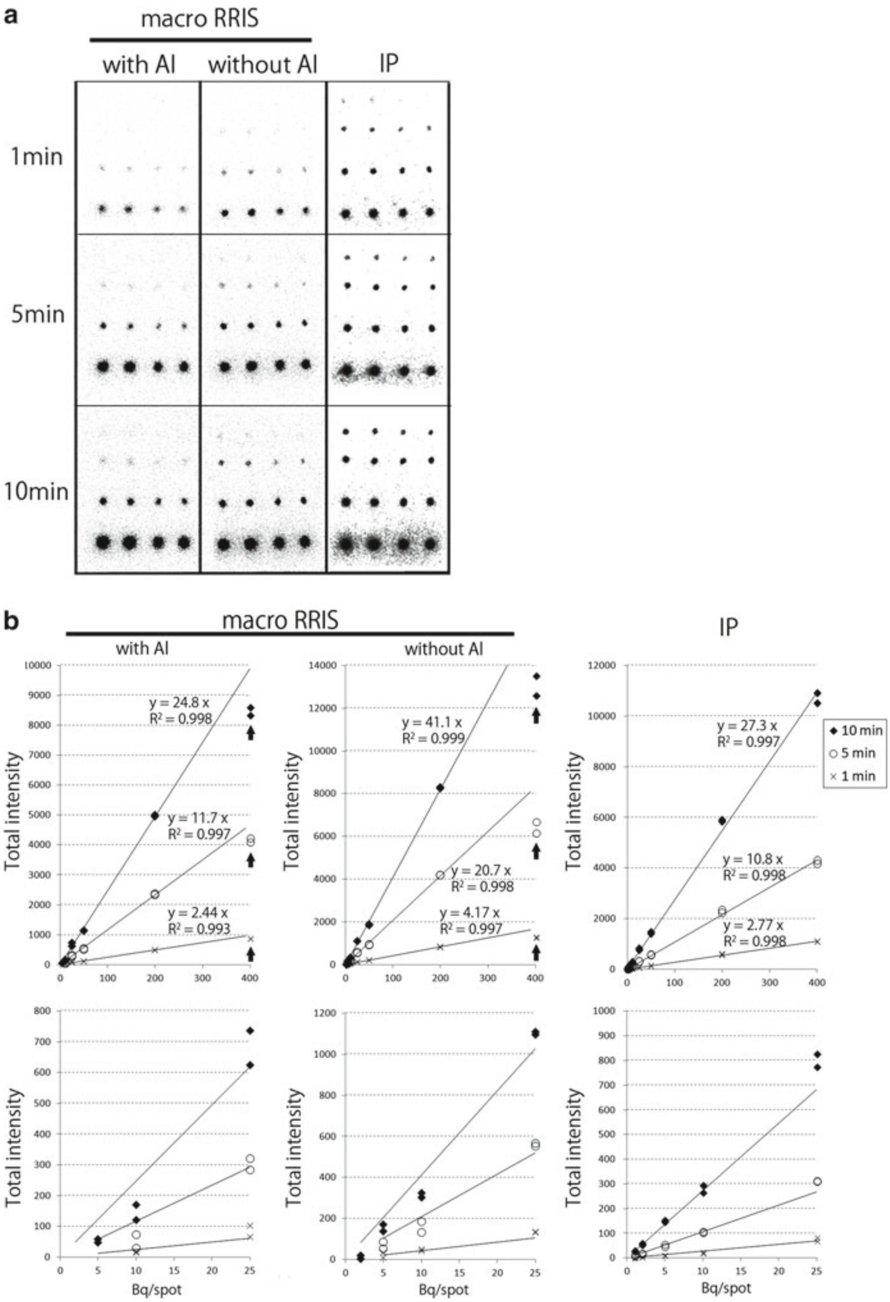

Fig. 5.2 Comparison of performance of macro RRIS and IP in the analysis of ^{137}Cs. (**a**) Images of a ^{137}Cs standard solution spotted on paper at concentrations ranging from 1 Bq (*top right*, two spots) to 400 Bq (*bottom left*, two spots), which were analyzed using macro RRIS and IP. Effects of the Al plate covering the scintillator surface were also examined. The standard solution was

soil medium, the rice plants absorbed only a little amount of ^{137}Cs even after 20 h, which was slightly detectable by IP (Fig. 5.3a, b). Immediately after the introduction of ^{137}Cs into the liquid medium, the ^{137}Cs signal in the liquid medium started to decrease and reached a plateau within 5 h, demonstrating that almost all ^{137}Cs added to the medium was absorbed by the rice (Fig. 5.3c). In the leaves, the ^{137}Cs signal increased almost linearly for 5 h, after which the rate of increase declined drastically, although plenty of ^{137}Cs was still present in the root (Fig. 5.3b). This characteristic ^{137}Cs uptake and translocation pattern was investigated further and has been presented in the following paragraph.

5.4 Distribution Pattern of Radiocesium in Soil

When ^{137}Cs was added to the soil, the ^{137}Cs signal was distributed uniformly throughout the soil (Fig. 5.4). However, radiocesium was found to be distributed non-uniformly in the surface soils collected from the Fukushima region, where some particles showed intense radioactivity (Fig. 5.4). The different radiocesium distribution patterns in the ^{137}Cs-added soil and the field soils contaminated by fallout suggested that the ^{137}Cs status of soil, particularly its chemical form, may be different, although the same radioactivity concentration was detected in both soils. This also implied that the results obtained in the soil-to-plant transfer analysis using soil with ^{137}Cs may not accurately reflect the natural situation in the field after contamination by fallout.

5.5 Observation of the Xylem Loading Activity Using Macro RRIS

As mentioned in Sect. 5.3, the rate of increase in the ^{137}Cs content declined significantly several hours after complete absorption of ^{137}Cs (Fig. 5.3c). There are two possible explanations for this observation. One is a decline in the physiological activity of the plant due to the specific experimental conditions, e.g., uncontrolled humidity, the possibility of severe mineral deficiency after the ^{137}Cs decline in the liquid medium within several hours (Fig. 5.3c), and the slight bending of the leaf blade within the plant box (Fig. 5.3a). If this was the case, only the initial increase in the curve could represent the physiological activity due to a combination of the

Fig. 5.2 (continued) detected using macro RRIS with an integration time of 1, 5, or 10 min, or using IP by maintaining contact with the spot for 1, 5, or 10 min. (**b**) Calibration curve for ^{137}Cs detected by macro RRIS with or without Al, and by IP. The signal intensity detected by macro RRIS and the PSL value recorded by IP were quantified based on the images in (**b**). The approximate curves and their approximate linear equations are also presented. The area between 1 and 25 Bq is magnified and shown in the *bottom* row

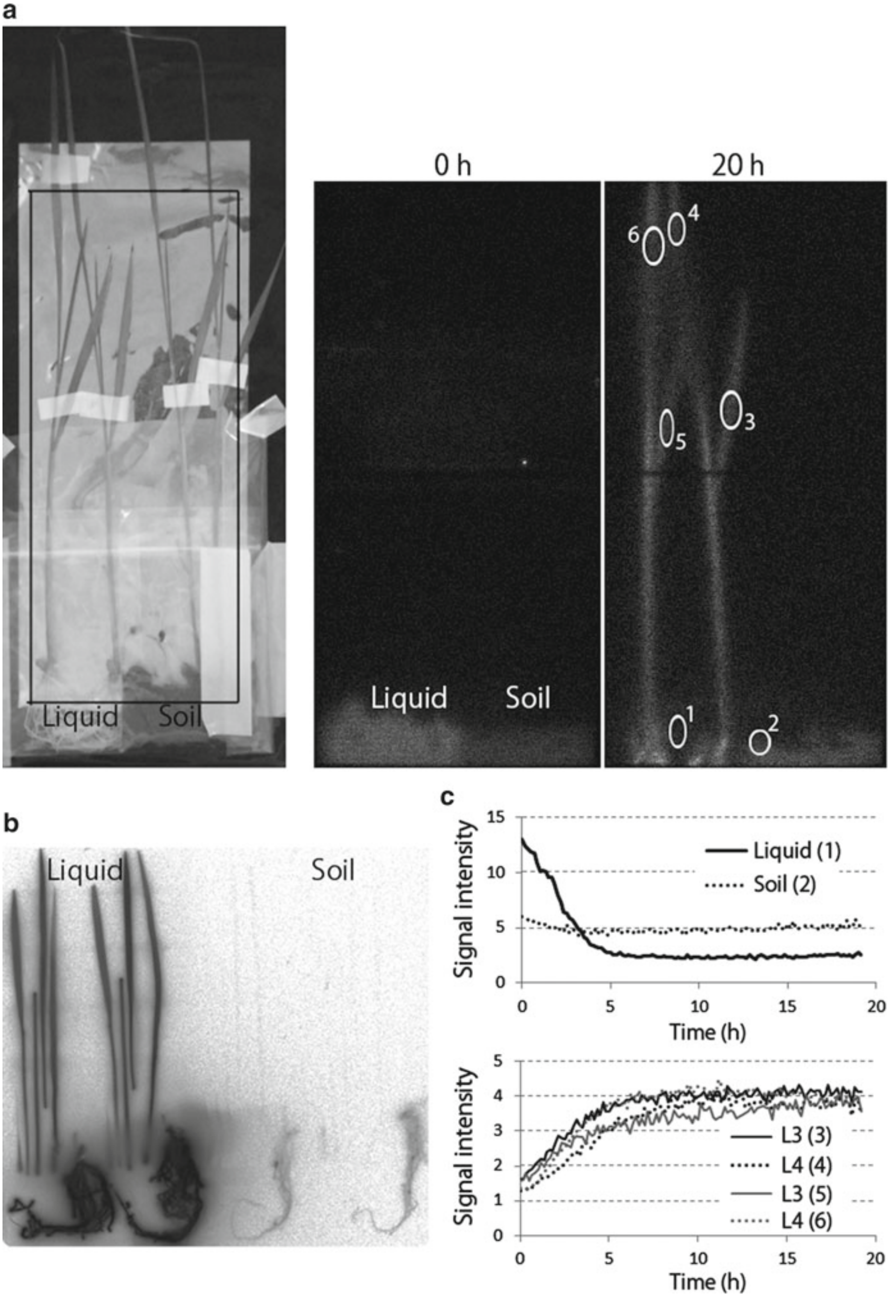

Fig. 5.3 ^{137}Cs uptake by the rice seedlings planted in liquid or soil. (**a**) Rice seedlings placed in the plant box and real-time images captured by the macro RRIS at the start of the experiment (0 h) and the end of the experiment (20 h). The integration time for each image was 10 min. The seedlings were grown in half-strength Kimura B liquid medium (K^+=0.27 mM) until they developed three expanded leaves (L2, L3 and L4) under 16:8 h of *light*:*dark* conditions at 30 °C. Two seedlings

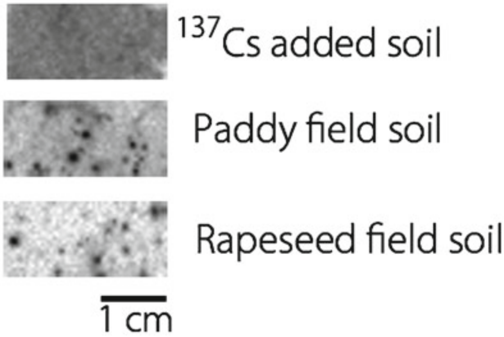

Fig. 5.4 The IP images of the soil with ^{137}Cs, and the surface soils collected from the paddy field and the rapeseed field in Fukushima district in June 2011. Bar = 1 cm

uptake and the xylem loading activity. Another explanation was that the ^{137}Cs increase in the shoot was composed of two distinct components. After the ^{137}Cs entered the root cytosol, some of the ^{137}Cs entered the cellular compartment for storage, whereas another part of the ^{137}Cs entered the xylem immediately and was translocated to the shoot through the xylem. If this was the case, the rapid translocation of ^{137}Cs to the shoot within 5 h (Fig. 5.3c) can be interpreted as the xylem loading activity, and the slowly increasing curve that appeared after 5 h (Fig. 5.3c) may be explained by the ^{137}Cs remobilization activity.

To distinguish these two possibilities, ^{137}Cs was resupplied to rice seedlings that had already been treated with ^{137}Cs for 19 h (Fig. 5.5). Then, ^{137}Cs content in the shoot increased again (Fig. 5.5). In contrast, the ^{137}Cs content did not change when the liquid medium was replaced with fresh medium that lacked ^{137}Cs, i.e., the renewing of experimental conditions did not restore the ^{137}Cs translocation from root-to-shoot (Fig. 5.5). Based on this result, the initially increasing curve was assumed to represent the xylem loading activity.

5.6 Effects of K Deficiency on the ^{137}Cs Uptake and Xylem Loading Activity

K-deficient plants usually accumulate greater amounts of Cs than K-sufficient plants (Zhu and Smolders 2000). A similar phenomenon was observed when K-sufficient and K-deficient rice seedlings were supplied with equal amounts of

Fig. 5.3 (continued) were planted in 3 ml of liquid medium or the soil medium (3 g of soil plus 3 ml of liquid medium) containing 50 kBq ^{137}Cs. The soil was collected from a paddy field in Fukushima district. The soil contained no radiocesium derived from fallout because the soil was collected from a deep part of the paddy field (5–10 cm from the surface). In the real-time image, six ROIs are shown. ROIs 1 and 2 indicate the liquid medium component. ROIs 3 and 5 are the L3 blades, whereas ROIs 4 and 6 are the L4 blades. (**b**) The IP image of rice seedlings after macro RRIS imaging was completed. The leaves were separated and cut into a root part with the bottom part of the shoot, L2, L3 sheath, L3 blade, L4 sheath, L4 blade, and L5 blade (L5 had not developed its sheath yet). The ^{137}Cs signal was hardly detected in L2 and L3 sheath. (**c**) Time course of ^{137}Cs signal accumulation in the six ROIs

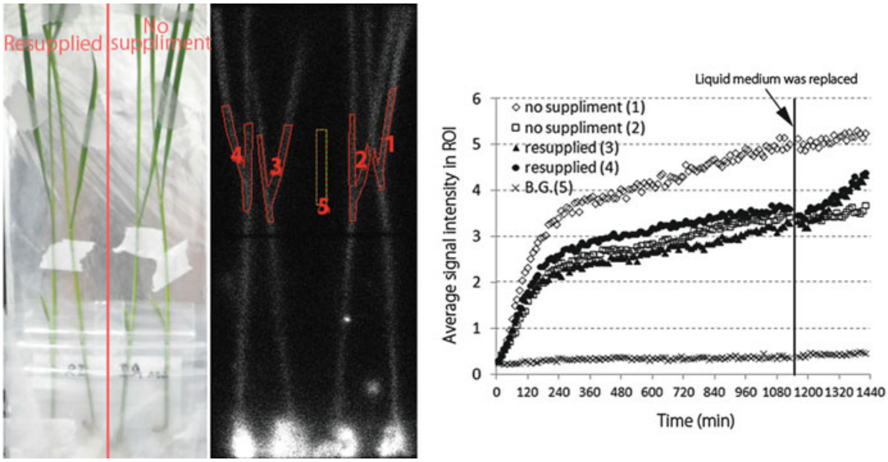

Fig. 5.5 Time course of ^{137}Cs signal accumulation in rice leaves after the liquid medium ($K^+=0.27$ mM) containing ^{137}Cs was replaced by liquid medium with or without ^{137}Cs. ^{137}Cs uptake images of four rice seedlings grown hydroponically for 2 weeks captured 19 h (1,140 min) after ^{137}Cs was introduced into the liquid medium at an intensity of 10 kBq/ml. Next, ^{137}Cs was resupplied for 4.5 h to two seedlings by replacing the liquid medium with 2 ml of fresh liquid solution containing 20 kBq ^{137}Cs. For the other two seedlings, the liquid medium was replaced by the fresh liquid medium without ^{137}Cs (no supplement). The integration time for each image was 10 min. ROIs 1 and 2 are the rice shoots without ^{137}Cs supplementation. ROIs 3 and 4 are the rice shoots that had ^{137}Cs resupplied at 1,140 min. ROI 5 represents the background signal intensity

^{137}Cs in a 0.27 mM K^+ condition (Fig. 5.6a). ^{137}Cs was rapidly accumulated in the K-deficient rice root (Fig. 5.6b), demonstrating that the ^{137}Cs uptake activity was increased significantly by a 2-day K-deficiency treatment. This increase in the ^{137}Cs uptake activity was quantified to determine the signal intensity in the liquid medium of K-deficient plants, where the ^{137}Cs signal decreased rapidly (Fig. 5.6c, d). However, the increase in the ^{137}Cs content of the leaves was not affected significantly by K-deficiency (Fig. 5.6d). This suggested that the ^{137}Cs xylem loading activity was not affected by the 2-day K-deficiency treatment. Alternatively, it is possible that K deficiency actually enhanced the xylem loading activity and that it also inhibited transpiration from the leaves, which could have negatively affected the root-to-shoot translocation of elements and the translocation rate to the shoots of K-deficient plants resulted in a similar manner to the K-sufficient plants. However, this possibility was rejected because the leaves of K-sufficient and K-deficient plants had the same transpiration rate (Table 5.1). In conclusion, 2 days of K deficiency enhanced the Cs uptake by the roots, whereas it had little effect on the Cs xylem loading process. Engels and Marschner (1992) suggested

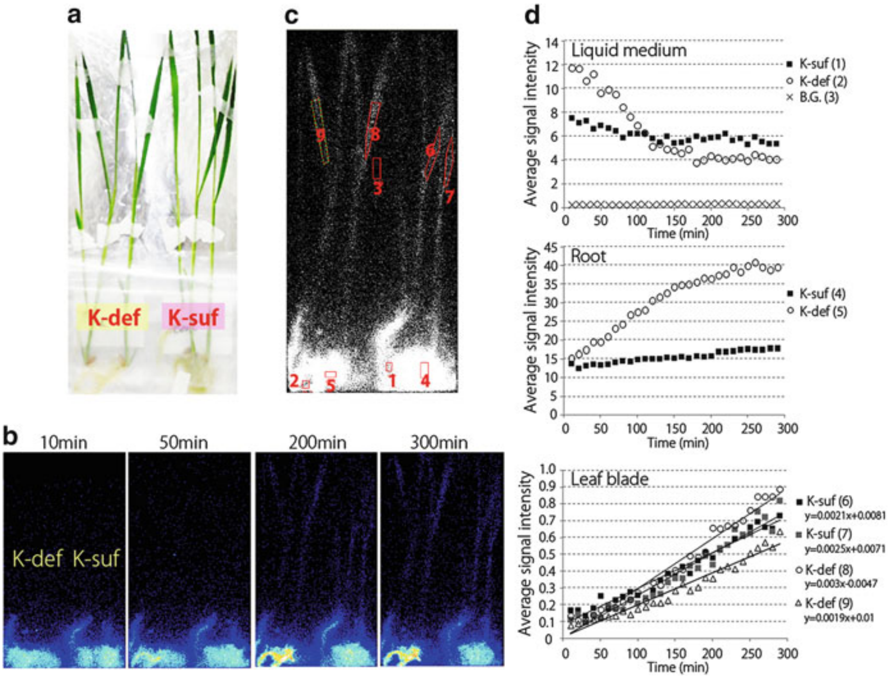

Fig. 5.6 Effects of K deficiency on the ^{137}Cs uptake and translocation pattern. (**a**) Rice samples placed in the plant box. The *red flame* indicates the viewing field of the macro RRIS. Three K sufficient seedlings and 3 K-deficient seedlings were prepared, although we only analyzed two seedlings in each set. K-sufficient plants were grown in liquid medium containing 3.5 mM K^+ for 2 weeks. For the K-deficiency treatment, the rice seedlings were grown in the K-sufficient medium for 12 days and transplanted to the K-free liquid medium, before further culture for 2 days. The K-sufficient and K-deficient seedlings were treated with normal liquid medium with added ^{137}Cs (10 kBq/ml). (**b**) Part of the sequential images captured by macro RRIS at 5 h (300 min). The integration time for each image was 10 min. (**c**) Positions of nine ROIs. ROIs 1 and 2 are the liquid medium component. ROI 3 represents the background signal intensity. ROIs 4 and 5 are the roots. ROIs 6–9 are the L3 blades of each seedling. (**d**) Time course of the ^{137}Cs signal decrease in the liquid medium (*top*), and its accumulation in the root (*middle*) and the leaf blade (*bottom*). The approximate curves are shown for the ^{137}Cs signal in the leaf blade and the linear approximate equations are shown under each sample name. *K-def* K-deficient, *K-suf* K-sufficient

Table 5.1 Transpiration rates from leaf blades L3 and L4 of K-sufficient and K-deficient rice plants

	L3		L4	
	K-suf	K-def	K-suf	K-def
Transpiration rate (mmol $H_2O/m^2/s$)	1.17	1.02	1.14	1.45
SD	0.189	0.0687	0.0833	0.201

No significant difference was observed between the samples

Sample size = 3; SD is the standard deviation of the three samples; *K-suf* K-sufficient, *K-def* K-deficient

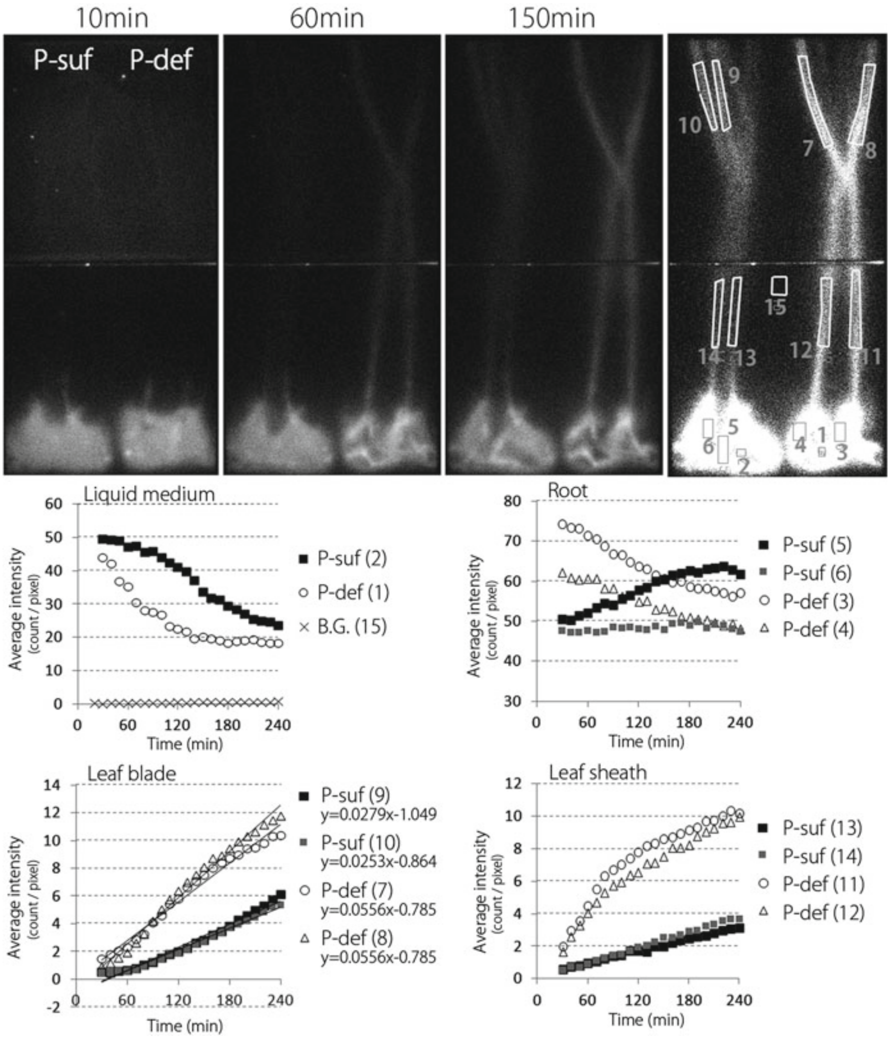

Fig. 5.7 Effects of P deficiency on ^{32}P uptake and translocation. For the P-sufficient and P-deficient samples, two seedlings were grown in normal half-strength Kimura B liquid medium (phosphate = 92 μM) for 2 weeks and two seedlings were grown normally for 6 days followed by cultivation in P-free liquid medium for 6 days, respectively. The P-sufficient and P-deficient seedlings were then placed into normal liquid medium containing ^{32}P (2 kBq/ml). After recording the ^{32}P images for 4 h, 14 ROIs were selected, i.e., the liquid medium component (ROIs 1 and 2), the roots (ROIs 3–6), the leaf blades (ROIs 7–10), and sheaths (ROIs 11–14), as indicated, to determine any time-dependent changes in the signal intensity. ROI 15 represents the background signal intensity. The approximate curves and their linear approximate equations are added in the corresponding figure to demonstrate the ^{32}P translocation kinetics in the leaf blade. *P-suf* P-sufficient, *P-def* P-deficient

that the K-xylem loading process and root uptake process were regulated separately in maize. Assuming that Cs migration was basically regulated by the K control system, the results presented in this study are consistent with those reported by Engels and Marschner (1992).

5.7 Enhanced P Xylem Loading Activity in P-Deficient Rice Plants

With respect to the xylem loading activity, it is known that the root-to-shoot transport of P is induced by P deficiency through the upregulation of P transporter gene expression (Ai et al. 2009). To confirm this property of P using macro RRIS and to confirm our experimental characterization of the xylem loading activity, we analyzed the ^{32}P uptake and translocation pattern using macro RRIS. After comparing the declining curve of the ^{32}P signal in the liquid medium from P-sufficient rice plants with that of P-deficient plants, we confirmed the positive effect of P-deficiency on the phosphate uptake activity (Fig. 5.7). After being absorbed by the roots, phosphate was shown to be transported to the shoots immediately and it never accumulated in the roots of P-deficient rice (Fig. 5.7). There was an evident increase in the P content of the leaves of P-deficient rice. These observations confirmed the stimulatory effect of 6 days of P deficiency on the P-xylem loading activity and uptake activity.

5.8 Conclusion and Future Perspectives

Using macro RRIS, we analyzed Cs uptake and xylem loading processes separately. In contrast to the positive effect of P deficiency on the uptake and xylem loading of P, K-deficiency had an initially positive effect on Cs uptake but not on Cs-xylem loading process. Other than these processes, the remobilization of Cs in the leaves and roots, the xylem-to-phloem transfer activity, and Cs exclusion from the roots are suggested as potential factors that affected the radiocesium distribution patterns in plants. Given that the Cs distribution in the edible parts is known to vary depending on the plant species, understanding these Cs translocation processes and evaluating the environmental factors that affect the Cs distribution pattern in rice, a model plant, could help to develop crop-specific agricultural techniques for reducing radiocesium accumulation in the edible plant parts. Further tracer experiments are required to evaluate each Cs translocation process and are currently underway.

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Chapter 6
Vertical Migration of Radiocesium Fallout in Soil in Fukushima

Sho Shiozawa

Abstract The vertical migration of radiocesium fallout in the soil was monitored for 1 year at several locations in Fukushima after the nuclear power plant explosion. We determined the vertical gamma ray intensity profiles in boreholes in the soil using a scintillation survey meter with a lead collimator to restrict the incoming radiation, only allowing horizontal detection. The average migration distances of radiocesium at two time points were accurately determined based on the difference in the depth of the centroids of two gamma ray intensity profiles. The results showed that although the convective velocity of radiocesium was unexpectedly as high as 1/10th of the velocity of the infiltrating rainfall water 2–3 months after the nuclear plant accident, the velocity decreased to 1/100th–1/200th of that of the water after 6–12 months. This indicated that strong fixation of radiocesium to clay particles occurred during the initial 2–3 months. Radiocesium uptake by plant roots may have decreased remarkably along with the mobility of radiocesium in the soil.

Keywords Collimator • Distribution coefficient • Gamma ray • Migration velocity of Cs • Sorption • Water flux in soil

Abbreviation

Cs Cesium

S. Shiozawa (✉)
Graduate School of Agricultural and Life Sciences, The University of Tokyo,
1-1-1 Yayoi, Bunkyo-ku, Tokyo 113-8657, Japan
e-mail: asho@mail.ecc.u-tokyo.ac.jp

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6.1 Retardation of Cesium Migration as a Result of Fixation to Soil Solids

Cesium (Cs) is soluble in water as a univalent cation. In general, the cation is fixed on any solid surface with a negative electric charge, such as soil particles or organic matter in soil. The fixed cation is exchanged by any other cation; in a manner that the concentrations of the fixed cations are in equilibrium with the concentrations of other cations in the soil water. Plant roots can easily absorb this form of Cs. This adhesion of Cs on soil solids is referred to as "weak fixation" in the present study. On the other hand, Cs strongly fixes to other types of clay crystals such as silicate sheets. The strongly fixed Cs ions on these clay particles are rarely replaced by other cations as soluble ions, and thus are seldom absorbed by plant roots.

The average water flux (q) in the surface soil over time is calculated as the amount of precipitation minus the soil evaporation, i.e., approximately q = 1,000 mm/year in Japan as well as in Fukushima. The corresponding average velocity of water molecules (v) is calculated as the water flux divided by the volumetric water content (θ), the value of which is, in case of a typical θ value of 0.5 is approximately v = q/θ = 2 m/year. However, the average velocity of Cs is retarded because of its fixation to soil solids. The ratio of the average velocity of Cs relative to that of water molecules is calculated based on the ratio of Cs present in the soil water to Cs in the total soil, including fixed Cs (Fig. 6.1), which is given by the following equation:

$$\frac{1}{\mathrm{Re}} \equiv \frac{\text{velocity of Cs}}{\text{velocity of } \mathrm{H_2O}} = \frac{\text{Cs in soil water}}{\text{Cs in overall soil}} \tag{6.1}$$

Assuming a sorption equilibrium of Cs concentrations on solids and water, and reversible exchange between the two phases, the distribution coefficient K_d can be introduced, which is defined as follows:

$$K_d \equiv \frac{\text{Cs concentration in solid phase [Bq / kg]}}{\text{Cs concentration in liquid phase [Bq / L]}} \tag{6.2}$$

Re can be calculated using K_d as follows:

$$\mathrm{Re} = 1 + \frac{K_d \rho_b}{\theta} \approx \frac{K_d \rho_b}{\theta} \tag{6.3}$$

where ρ_b is the dry bulk density. In general, K_d indicates the mobility (immobility) of a specific ion in a specific soil. Re defined in Eq. (6.1) is an inverse indicator of the mobility of Cs in the soil and also an indicator of the difficulty in Cs uptake by plant roots because plant roots absorb Cs in the form of ions from the soil water adjacent to the roots.

The IAEA handbook (IAEA 2010) provides mean values of K_d for Cs as K_d = 1,200 L/kg for 469 soils, K_d = 530 L/kg for sand, and K_d = 270 L/kg for organic soil. The K_d values for Cs are one to two order greater than radiostrontium (Sr), which is another typical radionuclide released by nuclear explosions (Sr was not released during the Fukushima accident), i.e., its mean value is K_d = 52 L/kg;

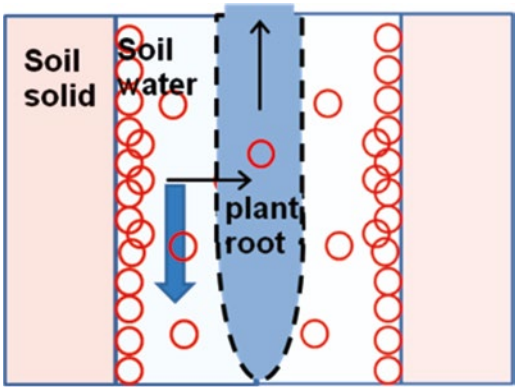

Fig. 6.1 Effects of cesium (Cs) fixation to soil solids. Cesium molecules dissolved in the soil water could only migrate with the water and were absorbed by the plant root

therefore, Cs is much less mobile in soil. The values of K_d measured in 30 Japanese paddy soils ranged from $K_d = 270$ L/kg to $K_d = 17,000$ L/kg with a geometric average of 2,300 L/kg (Kamei-Ishikawa et al. 2008). However, their K_d values were obtained in laboratory equilibrium conditions after shaking the suspended solutions in test tubes for several days; thus, the actual value of Re in the field could be very different from that calculated in the laboratory based on the K_d value. Given typical values of $\theta = 0.5$ and $\rho_b = 1.0$ g/cm^3, the value for Re calculated from K_d is approximately twice of that of K_d.

Field investigations in Sweden after the Chernobyl accident (Rosen et al. 1999) showed that the initial velocity of Cs during the first year (1987) was 5–10 mm/year, which decreased to 1–4 mm/year in the next 5–8 years, indicating that the strong fixation of Cs to soil particles proceeded gradually for years. Thus, the values of Re may be approximately 100–1,000. This investigation also showed that the Cs velocity in organic soil was several times that in mineral soil.

6.2 Cs Concentration Profile in Soil 2 Months After the Fallout

Figure 6.2 shows vertical concentration profiles of radiocesium (^{134}C and ^{137}Cs) determined in an undisturbed paddy field in Fukushima Prefecture before plowing on May 24th, 2011, approximately 2 months after the radiocesium fallout (Shiozawa et al. 2011). The top 0–15 cm of the soil was sampled, which was separated into six layers of 1–5 cm thickness. The resulting concentration profile indicated that 88% of ^{134}Cs + ^{137}Cs was present within the 0–3 cm layer and 96% within the 0–5 cm layer. However, although most of the radiocesium remained within the top few centimeters of the surface layer, some of it had already reached the 10–15 cm layer. The mean traveling distances of ^{134}Cs and ^{137}Cs were calculated from the profile as 1.74 cm. On the other hand, the mean traveling distance of water molecules during the 70-day period was estimated based on the (precipitation − evaporation)/(volumetric water content) as approximately 20 cm. Therefore, the convective

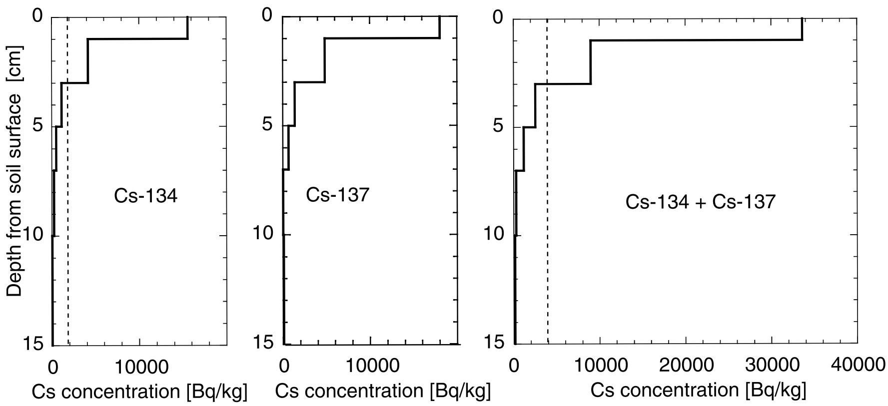

Fig. 6.2 Radiocesium concentration profiles in the soil on May 24th, 2011, for an undisturbed paddy field (*solid line*) and a plowed paddy field (*dashed line*) (Shiozawa et al. 2011)

velocity of Cs was 1/10th of that of water due to the sorption of Cs on soil particles. However, this Cs velocity was two to three orders greater than that calculated based on laboratory determined distribution coefficients (K_d) for Japanese paddy soils (Kamei-Ishikawa et al. 2008), indicating that the movement of Cs in the field was very different from the sorption equilibrium.

It is difficult to determine Cs migration distances of several millimeters by comparing the two vertical concentration profiles obtained by soil sampling at different time points because the Cs concentration profiles are not in exactly the same soil positions. They vary considerably even if they are obtained at the same time. Thus, we developed a method for measuring Cs migration distances of several millimeters in the soil over a period of several months by comparing two vertical gamma-ray intensity profiles, which were measured using a scintillation survey meter inside monitoring boreholes.

6.3 Method Used to Monitor the Cs Migration Distance in a Borehole

6.3.1 Collimator and Monitoring Pipe

To measure the intensity of horizontal incoming γ-rays at each depth in the soil, the gamma-ray detector was enclosed by a barrier (lead) that had a slit window around the detector to restrict any incoming radiation except for that in the horizontal direction. A cylindrical scintillation probe (Health Physics Instruments, Model 5000) measuring 35 mm in diameter was covered with a handmade lead collimator that

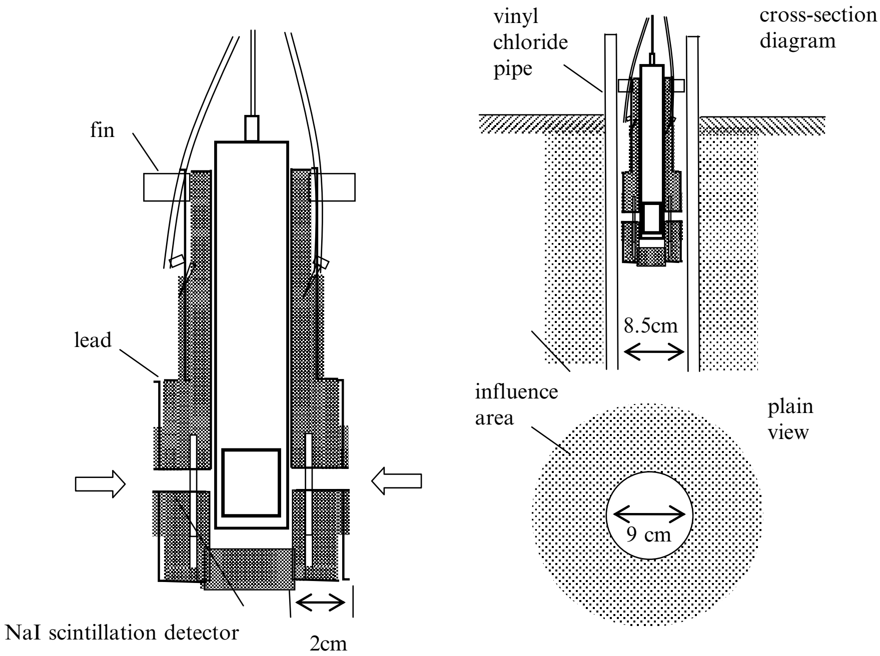

Fig. 6.3 Lead collimator for the scintillation probe and the soil gamma-ray distribution monitoring pipe in a borehole

had a slit window around the scintillation detector (Fig. 6.3). The collimator consisted of an upper part (main body) and a lower part, which were separated by the window. The lower part could slide in an acryl guide cylinder and the base was fixed onto the main body such that the vertical width of the slit window was variable in a range of 0–2 cm. The lower part of the collimator was connected to the upper part by a rubber band, whereas pieces of polystyrene spacers were placed in the window to determine the size of the window. The horizontal depth of the slit window was 2 cm. The collimator was made from 2-mm thick lead plate. The thickness of each part ensured that radiation entering the detector from any direction had to pass through more than 1.5 cm of lead. The width of the window was determined as 0.8 cm after several test measurements, based on the sensitivity of the measured profiles and the time required for measurements.

In each of monitoring places we made a borehole in the soil using a hand auger, into which we inserted a vinyl chloride pipe that measured 0.6–1.3 m in length, 85 mm internal diameter, and 89 mm external diameter. The scintillation probe with a lead collimator was suspended in the monitoring pipe using a rope to measure the gamma-ray intensity at each depth. We added a fin to the root of the collimator to prevent it from tilting in the pipe.

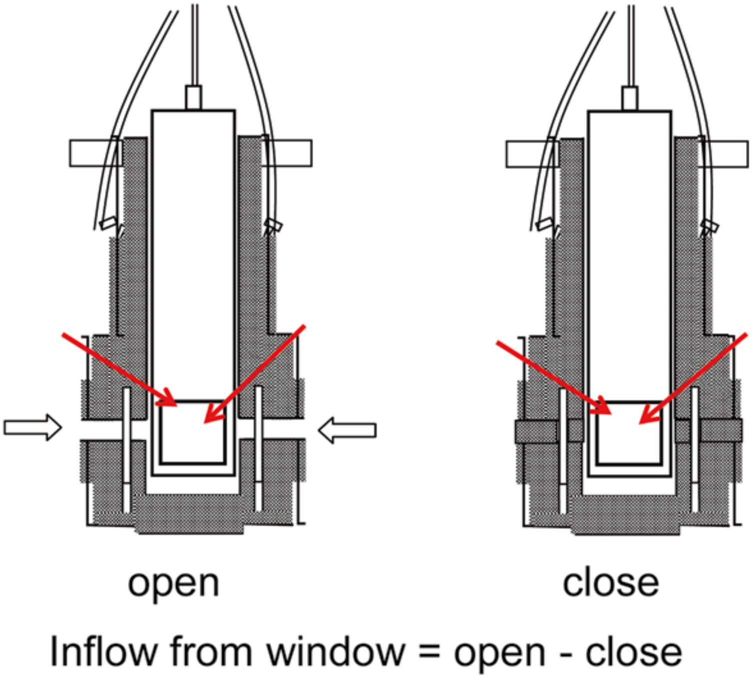

Fig. 6.4 Method for correcting insufficient lead insulation of the collimator

6.3.2 Method for Correcting Insufficient Collimator Lead Shielding

The thickness of the lead shielding of the collimator was insufficient and the lead itself emitted some gamma rays. These noise, radiation leaks, and any other undesirable radiation had to be subtracted from the measured radiation intensity to determine the radiation that exclusively entered the slit window. In order to cancel the noises, we added a measurement in which the slit window was plugged with lead (Fig. 6.4). The correct value of the radiation intensity at each depth in the monitoring pipe was obtained by subtracting the value measured with the plugged window collimator at the same depth from the value measured with the open window collimator. Using this method, the corrected radiation distribution reflected the actual distribution of Cs concentrated at the soil surface more sharply, as shown in Fig. 6.5. The time required to measure the radiation intensity at each depth in the monitoring hole was 25–50 s, depending on the amount of Cs present (Bq/m^2) at each site.

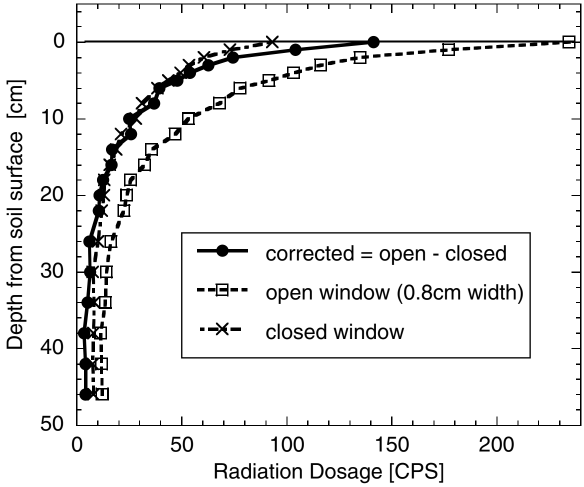

Fig. 6.5 Effects of correction for leakage through the lead insulation of the collimator. An example measured in Koriyama is shown

6.3.2.1 Calculation of the Cs Migration Distance

The average Cs migration distance was determined based on the center of the vertical radiation intensity (or concentration) profiles, which was calculated as follows:

$$\langle x \rangle = \frac{\sum_i x_i (\mathrm{c_i} - \mathrm{c_0}) \Delta x_i}{\sum_i (\mathrm{c_i} - \mathrm{c_0}) \Delta x_i} \tag{6.4}$$

where $\mathrm{c_i}$ is the measured radioactive intensity (or Cs concentration) at depth of x_i, and Δx_i is the interval of the measured depth, and $\mathrm{c_0}$ is the average radioactive intensity (or Cs concentration) in the deep soil layer where $\mathrm{c_i}$ does not decrease with depth.

For the profile obtained by soil sampling where $\mathrm{c_i}$ was the actual Cs concentration, the center of the profile $\langle x \rangle$ in Eq. (6.4) indicates the average Cs migration distance during the period after Cs fallout (late March 2011). For the profiles measured in situ in the monitoring holes, the average Cs migration distance between the two time points was determined by the difference in the centers of the two profiles.

6.4 Results and Discussion

Figure 6.6 shows a comparison of the vertical radiation profiles at two time points, which were measured in the monitoring boreholes at each location, and the radiocesium concentration profile at the first time point for the sampled soil in each hole (right-hand side). The soils in the locations were not disturbed after the Cs fallout. It was apparent that the vertical radiation profiles measured in situ in the monitoring

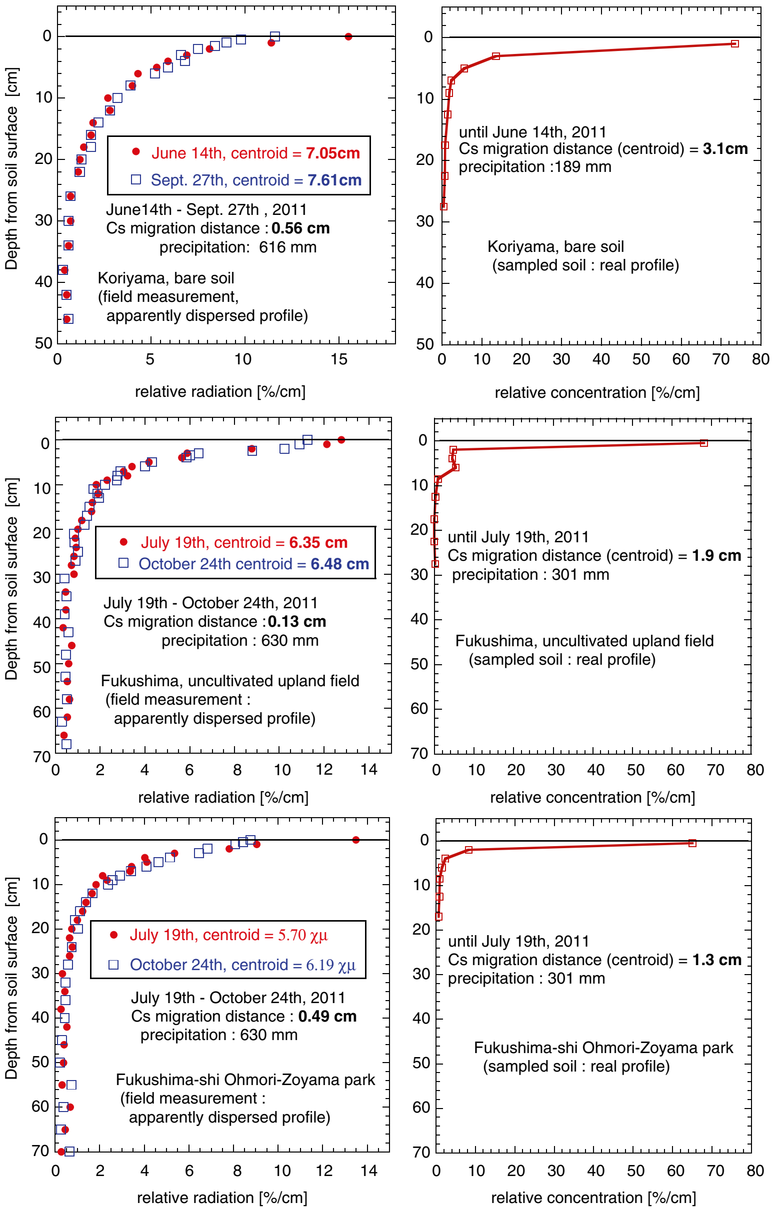

Fig. 6.6 Vertical radiation profiles at 2 time points measured in each monitoring borehole (*left-hand side*) and the radiocesium concentration profile at the 1st time point for the sampled soil from each hole (*right-hand side*). The soils were not disturbed in the locations after the radiocesium fallout. The depth of the centroid of each radiation/concentration profile is shown

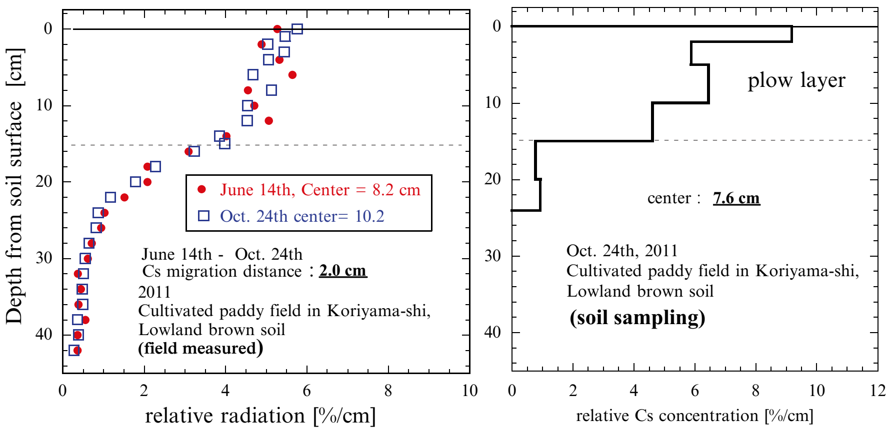

Fig. 6.7 Comparison of the vertical radiation profiles measured in the monitoring borehole (*left-hand side*) and the radiocesium concentration profile of the sampled soil (*right-hand side*) for a cultivated paddy field where a 0–15 cm layer was plowed and puddled with water in May 2011 after the radiocesium fallout

boreholes were highly dispersed and the location of the center was much deeper than the actual concentration profiles obtained from the sampled soil. This suggested that the radiation profiles measured in the monitoring holes did not accurately reflect the actual concentration profiles when the gamma-ray source was concentrated in a thin soil surface layer. There are two reasons for the measured radiation profiles to be dispersed. First, the slit window of the collimator was not sufficiently narrow compared with its depth to restrict radiation incoming from inclined direction other than horizontal direction. Second, the scintillation survey meter used in the study detected gamma rays scattered throughout the soil as well as those emerging directly from the radiation source (Cs). Therefore, the in situ radiation profiles were more dispersed than the actual concentration profiles when the source was concentrated in a thin surface layer. Figure 6.7 shows the profiles for a cultivated paddy field where a 0–15 cm deep plowed layer was mixed and puddled before the monitoring hole was created. For such vertically distributed gamma-ray source, the radiation profile measured in the hole accurately reflected the gamma-ray source concentration profile.

Although radiation profile measured in the monitoring hole was considerably dispersed and did not reflect the actual radiocesium concentration distribution, the difference in the depth of the centers of the profiles at the two time points may accurately reflect the average migration distance of the radiocesium during this period.

The migration distances between the two time points detected by in situ radiation monitoring and the initial migration distances obtained from the first soil samples are summarized in Table 6.1, with the precipitation data during these periods. The initial migration distance for 2–3 months after the radiocesium fallout (late March 2011) until the first soil sampling was 13–22 mm, which was approximately 1/10th–

Table 6.1 Average distance of radiocesium migration in the soil based on the centroids of the radiation profiles measured in the monitoring boreholes

	Field containing flowering trees (Koriyama-shi) Brown lowland soil 620 kBq/m^2	Uncultivated upland field (Fukushima-shi) Brown lowland soil 350 kBq/m^2	Ohmori-Zoyama Park (Fukushima-shi) Brown forest soil 480 kBq/m^2
Period 1 from March 11th, 2011	To June 14th, 2011	To July 19th, 2011	To July 19th, 2011
Depth of the centroid (for soil sampling)	**21.6 mm**	**19 mm**	**13 mm**
Precipitation (R)	189 mm	301 mm	301 mm
Period 2	To September 29th, 2011	To October 21th, 2011	To October 21th, 2011
Distance between the centroids of the two profiles	**9.7 mm**	**2.3 mm**	**4.9 mm**
Precipitation (R)	616 mm	630 mm	630 mm
Period 3	To March 28th, 2012	To March 27th, 2012	
Distance between the two centroids	**5.4 mm**	**2.0 mm**	
Precipitation (R)	340 mm	315 mm	

The three locations were bare soil that received radiocesium fallout

1/20th of the water molecule migration rate, given that the average migration distance of water molecules (q/θ) was similar to or twice the depth of precipitation (R) based on the Japanese meteorological conditions and the soil water content in the locations. However, after June 2011, the velocity decreased to 1/10th of that in the early period, i.e., to approximately 1/200th of water molecules.

In the 2–3 months after the radiocesium fallout in late March 2011, Cs was fairly mobile accompanied by the soil water movement; however, its mobility decreased remarkably after this period. Further evidence also suggested that Cs remained fairly mobile until May 2011, i.e., 2 months after the Cs fallout, because the soil samples from the cultivated paddy field shown in Fig. 6.7, which were taken from the subsoil below the plowed layer (below 15 cm) in late May (before water filling), contained high levels of Cs (15–20%). This suggested that Cs was moved from the plowed soil into the undisturbed sublayer by rainfall, after plowing in early May.

The results shown in Table 6.1 indicate that strong Cs fixation occurred over a course of few months. The Cs fallout accompanied weak rainfall during late March 2011, after the nuclear plant accident. It is considered that the form of Cs present in the fallout was mostly that of soluble ion and not the form attached to solid particles because a large amount of the Cs fallout was fixed to the smooth surfaces of trees and their leaves by Coulomb force. The fixed Cs on trees was not washed away by rainfall (≥600 mm), and half of the radiocesium fallout was still present on trees above the ground in the summer of 2011. When the radiocesium fallout came in contact with the soil surface, it was immediately fixed to the soil by Coulomb force

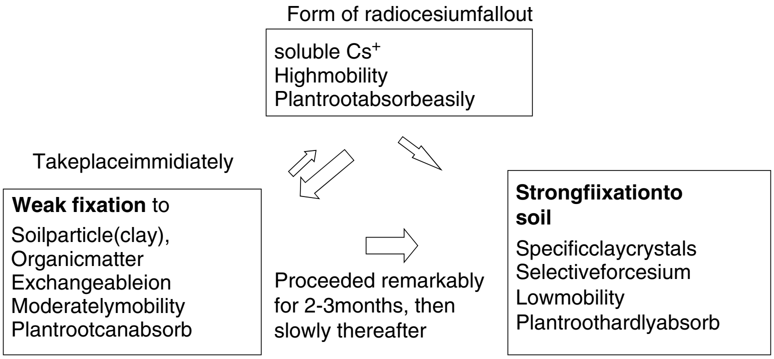

Fig. 6.8 Forms of radiocesium present in the soil and their dynamics

and to any organic material that covered the soil surface. This initial fixation to the soil or organic material was weak because of which the fixed Cs could be replaced by other cations in the soil water that were present as soluble ions and thus, they were relatively mobile in the water flow. Although weak fixation may occur immediately, Cs can also be gradually transferred into a state of strong fixation at specific sites on the inner surfaces of silicate crystal clay sheets (Fig. 6.8). The radiocesium migration analysis in the present study indicated that the transfer to strong fixation typically proceeded over a course of few months after the fallout, until June 2011 in the soils in Fukushima. The radiocesium concentrations in vegetable samples produced in Fukushima Prefecture dropped to almost "not detected" after July 2011; however, previously, some samples were detected (>10 Bq/kg) for which the radiocesium was considered to be transferred from the root. The transfer of radiocesium from weak fixation to strong fixation must have prevented the plant root uptake as well as the decreased rate of migration in the soil.

6.5 Conclusion

The velocity of the convective movements of radiocesium was 1/10th–1/20th of that of water molecules in the soil during the first 3–4 months after the Cs fallout, from late March 2011 to June or July 2011, but it declined rapidly to 1/100th–1/200th of water molecules during the next 8–9 months (up to March 2012). This remarkable reduction in the Cs migration rate indicated a change in the Cs transfer from weak fixation to strong fixation to soil over a course of few months, which would also have been accompanied by a remarkable reduction in the Cs transfer from soil to plants through root uptake. Therefore, according to the long-term field investigations of Cs fallout after atmospheric nuclear weapon testing and the Chernobyl accident, the Cs mobility in soil will decrease slowly year after year.

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Chapter 7
Radioactive Nuclides in Vegetables and Soil Resulting from Low-Level Radioactive Fallout After the Fukushima Daiichi Nuclear Power Plant Accident: Case Studies in Tokyo and Fukushima

Seiichi Oshita

Abstract Vegetables and field soils about 60 and 230 km away from the Fukushima Daiichi nuclear power plant were examined for ^{134}Cs and ^{137}Cs radioactivity. The total ^{134}Cs and ^{137}Cs transferred was <7 Bq/kg wet weight in potato tubers grown in fields where the total ^{134}Cs and ^{137}Cs concentration in the soil was ≤1,235 Bq/kg dry weight. For the edible parts of lettuce and cabbage, the total ^{134}Cs and ^{137}Cs concentrations were lower than the detection limit. In this case, the maximum value in soil was 651.2 Bq/kg dry weight.

Keywords Cabbage • Cesium-134 • Cesium-137 • Lettuce • Low level • Potassium-40 • Potato • Soil

7.1 Introduction

The nuclear power plant accident that occurred in Fukushima Prefecture in March 2011, caused a wide spread of radioactive nuclides over various parts of Japan. This raised the immediate concern that agricultural products might become directly contaminated by radioactive nuclide fallout. Therefore, an investigation of the contamination of vegetables by radiocesium (^{134}Cs and ^{137}Cs) was initiated about 2 months after the accident (Oshita et al. 2011, 2013).

For this investigation, vegetable samples were collected from the fields at the Institute for Sustainable Agro-ecosystem Services, Graduate School of Agricultural and Life Sciences, The University of Tokyo, located in Nishitokyo City, Tokyo. Samples were also collected from fields near a mountain village in Fukushima Prefecture.

S. Oshita (✉)
Department of Biological and Environmental Engineering, Graduate School of Agricultural and Life Sciences, The University of Tokyo, 1-1-1, Yayoi, Bunkyo-ku, Tokyo 113-8657, Japan
e-mail: aoshita@mail.ecc.u-tokyo.ac.jp

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Another possible problem, which could have developed over time in addition to direct contamination, was the transfer of radioactive nuclides through root absorption (indirect contamination). Therefore, the transfer of radioactive nuclides from the soil was studied by intentionally spreading high-concentration radioactive nuclides onto culture media. Previous studies examined rice plants (Tensho et al. 1959; Yonezawa and Mitsui 1965; Tsumura et al. 1984), grapes (Zehnder et al. 1995), mushrooms (Ban-nai et al. 1994) and field crops (Uchida et al. 1987; Ban-nai et al. 1995; Ban-nai and Muramatsu 2002). Another study assessed the effects of environmental radioactivity on foodstuffs (Schwaiger et al. 2004). To examine radionuclide transfer coefficients, ^{137}Cs at an extremely high concentration was used in many cases such as the paper by (Ban-nai et al. 1995) [490 kBq/pot (about 163 kBq/kg, as the one pot was filled with 3.0 kg of the soil)] and that by (Broadley and Willey 1997).

In the results reported here, the amounts of radiocesium (^{134}Cs + ^{137}Cs) transferred from the field soil to vegetables at a low concentration in the soil of ≤1,235 Bq/kg dry weight were measured in order to determine the transfer of radiocesium in vegetable-producing fields.

7.2 Materials and Methods

7.2.1 Vegetable and Soil Samples

Samples were obtained from two different locations. One was the agricultural research field at the Institute for Sustainable Agro-ecosystem Services located in Nishitokyo City, Tokyo (altitude approximately 60 m above sea level; this site is referred to as "Nishitokyo City"). This site was about 230 km away from the Fukushima Daiichi nuclear power plant. Potatoes (Irish Cobbler) and cabbage (YR Rakuzan) that grew in this field and the soil around these vegetable-producing fields were collected on 16 May 2011 (40 days after the settled cabbage planting and 47 days after potato planting), on 27 May 2011 (51 days after the settled cabbage planting and 58 days after potato planting) and on 20 June 2011 (75 days after the settled cabbage planting and 82 days after potato planting). Soil samples, mostly consisting of surface soil (depths of 0–5 cm), were sampled at the time of vegetable sampling. In addition, on 11 June 2011, more soil samples were collected at 5-cm intervals and at depths as great as 35 cm below the ground surface.

Potato (Kita-Akari), cabbage (Shikidori) and lettuce (Berkley) samples were collected from a mountain village (altitude approximately 420 m above sea level) located in the southeastern part of Fukushima Prefecture and about 60 km away from the Fukushima Daiichi nuclear power plant. They were sampled on 18 July 2011, 99 days after potato planting, 78 days after the settled cabbage planting and 49 days after the lettuce planting. Soil was sampled from each field at 5- or 10-cm intervals and at depths as low as 30 cm below the ground surface.

7.2.2 Radioactive Nuclide Concentration Measurements

We measured the concentrations of ^{134}Cs and ^{137}Cs, both of which have a long half-life, as well as naturally occurring ^{40}K using a germanium semiconductor detector (ORTEC, SEIKO EG&G Co. Ltd.). For the samples collected on 16 May 2011, the measurement time was set to 50,000 s for the vegetable samples and 18,000 s for the soil samples. For all other samples, the measurement time was set to 3,600 s.

7.3 Results and Discussion

7.3.1 Concentrations of Radioactive Nuclides in Vegetables and Soil (Tokyo)

Table 7.1 shows the measured concentrations of the radioactive nuclides ^{134}Cs, ^{137}Cs and ^{40}K in samples of potato leaves (both washed and unwashed), roots, tubers and soil. Among the potato samples collected 47 days after planting, an extremely small amount of radiocesium was detected in the washed (^{137}Cs was detected but ^{134}Cs was not) and unwashed (both ^{134}Cs and ^{137}Cs were detected) leaf samples. However, radiocesium (^{134}Cs and ^{137}Cs) was not detected in the unwashed leaf sample of potatoes collected 58 days after planting, although a very small amount was detected in the washed sample.

These results contradicted our predictions. This could have been because the detection limit for the unwashed leaf sample was about twice as high as that for the washed leaf sample, as indicated in Table 7.1. Because the amount of the unwashed leaf sample was less than that of the washed leaf sample, we suspected a greater detection limit for the unwashed leaf sample.

No radiocesium was detected in the root samples. With regard to tubers, which are the edible parts of plants, ^{137}Cs was detected at a concentration of 2.4 Bq/kg wet weight in the sample collected 58 days after planting. Although the concentration of ^{134}Cs was lower than the detection limit, the measurement is provided as a reference only. ^{134}Cs and ^{137}Cs were not detected in the sample collected 82 days after planting.

To explain these results for tubers, we considered potassium (K) to be an important factor for tuber growth. Translocation and storage of carbohydrates occurs in staple crops rich in starch, such as potatoes, and potassium ions (K^+) move around in these plants. A report on daikon radish described that cesium behaved like potassium in plants, in that the potassium concentrations in root tubers decreased after a certain stage of plant growth and cesium exhibited the same tendency (Ban-nai and Muramatsu 2002).

Comparison of the concentrations of ^{40}K based on this hypothesis showed that the concentrations in the samples collected 82 days after planting were about 45% of those in the samples collected 58 days after planting. We estimated that the normal ^{40}K concentration in potato tubers was 124.6 Bq/kg wet weight. This was based on the assumption that the potassium concentration in potato tubers is 410 mg/100 g

Table 7.1 Concentration of ^{134}Cs, ^{137}Cs and ^{40}K of potato and soil in Tokyo

			Nishitokyo City (May 16, 2011)		Nishitokyo City (May 27, 2011)			Nishitokyo City (June 20, 2011)		
			47 days		58 days			82 days		
Days from settled planting			Moisture content (% w.b.)	Concentration (Bq/kg[a])	Moisture content (% w.b.)	Concentration (Bq/kg[a])	Detection limit (Bq/kg[a])	Moisture content (% w.b.)	Concentration (Bq/kg[a])	Detection limit (Bq/kg[a])
Potato	Leaf (washed)	^{134}Cs	–	N.D.	90.0	3.8	2.9			
		^{137}Cs		1.8		4.9	3.4		–	
		^{40}K				380.3	32.6			
	Leaf (without wash)	^{134}Cs	–	4.3	90.0	N.D.	6.0			
		^{137}Cs		4.5		N.D.	8.5		–	
		^{40}K				N.D.	77.9			
	Root (washed)	^{134}Cs	–		91.7	N.D.	–		N.D.	0.7
		^{137}Cs		–		N.D.	–	88.7	N.D.	0.7
		^{40}K				508.7	144.4		107.5	13.4
	Tubor (washed)	^{134}Cs	–		–	1.2[b]	1.8		N.D.	0.3
		^{137}Cs		–		2.4	2.1	80.6	N.D.	0.3
		^{40}K				338.3	18.9		151.2	8.2
Soil (potato)		^{134}Cs	25.9	57.2	32.3	65.4	7.5	10.3	34.4	4.5
		^{137}Cs		73.3		101.5	8.8		51.0	4.6
		^{40}K		265.0		185.3	68.7		180.0	47.5

[a]Bq/kg wet weight for potato and Bq/kg dry weight for soil
[b]Less than detection limit

(Standard Tables of Food Composition in Japan, http://www.mext.go.jp/b_menu/shingi/gijyutu/gijyutu3/toushin/05031802.htm) and the radioactivity of 1 g of potassium is 30.4 Bq/g, also considering the natural abundance of ^{40}K.

The value of 124.6 Bq/kg wet weight was almost 82% of the value of 151.2 Bq/kg wet weight measured in the sample collected 82 days after planting. This indicated that the measured concentration of ^{40}K corresponded closely to the potassium concentration in tubers. Because the potassium concentration would naturally decrease to about 45% in the plants collected 58 days after planting, as with ^{40}K, we considered that the concentration of radiocesium, which behaves like potassium, would be reduced by half in the sample collected 82 days after planting and thus become undetectable.

Table 7.2 shows the concentrations of the radioactive nuclides ^{134}Cs, ^{137}Cs and ^{40}K in the samples of cabbage leaves (edible parts and outer leaf, washed and unwashed), roots and soil. Radiocesium was detected only in the samples of the outer leaf (non-edible parts) collected at 40 days and 75 days after settled planting. Because the concentrations were lower in the samples collected 75 days after planting than in the samples collected 40 days after planting, we assumed deposition of radioactive fallout.

It should be noted that the measurements obtained for the samples collected 75 days after planting indicated that the concentration of ^{137}Cs was lower than that of ^{134}Cs, although the detection sensitivity should have been higher for ^{137}Cs. One reason for these results was assumed to be the low concentrations in the samples that were close to the detection limit. However, this needs to be clarified. The major reason for radiocesium becoming undetectable in the outer leaf samples collected 51 days after planting was believed to be the six times higher detection limit for these samples than that for samples collected 75 days after planting.

Figure 7.1 shows the vertical distributions in the soil for radioactive ^{134}Cs, ^{137}Cs and ^{40}K. The "depth" axis in this figure shows negative values because depth was measured using the bottom of the ridge as the reference point (zero) when collecting soil samples from the potato field. In the soil samples from the potato and cabbage fields, distribution of radiocesium was observed in the plow layer, which was 15 cm below the ground surface. The concentrations were particularly high in the soil samples from the surface and ridges. Tables 7.1 and 7.2 show that the radiocesium concentration in the surface soil varied depending on the day of sampling. We assumed that this indicated a two-dimensional concentration distribution.

7.3.2 Concentrations of Radioactive Nuclides in Vegetables and Soil (Fukushima Prefecture)

Table 7.3 shows the concentrations of the radioactive nuclides in samples of potato tubers (washed) and aerial parts (leaf and stem, washed). Table 7.4 shows the concentrations in samples of edible cabbage parts (unwashed and washed), outer leaves (washed) and roots (washed). The concentrations in the samples of edible lettuce parts (unwashed and washed) and roots (washed) are shown in Table 7.5. Figure 7.2 shows the distributions of radioactive ^{134}Cs and ^{137}Cs concentrations in the soil samples collected at different depths in each field.

Table 7.2 Concentration of ^{134}Cs, ^{137}Cs and ^{40}K of cabbage and soil in Tokyo

			Nishitokyo City (May 16, 2011)		Nishitokyo City (May 27, 2011)			Nishitokyo City (June 20, 2011)		
			40 days		51 days			75 days		
Days from planting			Moisture content (% w.b.)	Concentration (Bq/kg[a])	Moisture content (% w.b.)	Concentration (Bq/kg[a])	Detection limit (Bq/kg[a])	Moisture content (% w.b.)	Concentration (Bq/kg[a])	Detection limit (Bq/kg[a])
Cabbage	Edible part (washed)	^{134}Cs	–		–				N.D.	0.7
		^{137}Cs						91.0	N.D.	0.8
		^{40}K							76.9	10.1
	Edible part (without wash)	^{134}Cs	–		–				N.D.	0.4
		^{137}Cs						93.1	N.D.	0.4
		^{40}K							66.4	5.9
	Outer leaf (washed)	^{134}Cs		N.D.	92.3	N.D.	2.3		N.D.	0.5
		^{137}Cs		N.D.		N.D.	2.9	91.9	0.6	0.5
		^{40}K				135.4	29.4		94.6	7.8
	Outer leaf (without wash)	^{134}Cs		3.4	92.3	N.D.	2.9		1.0	0.5
		^{137}Cs		4.2		N.D.	3.4	90.8	0.8	0.6
		^{40}K				133.3	34.9		101.8	8.1
	Root (washed)	^{134}Cs	–		92.2	N.D.	–		N.D.	0.4
		^{137}Cs				N.D.	–	91.8	N.D.	0.5
		^{40}K				134.1			58.0	8.3
Soil (cabbage)		^{134}Cs	27.3	42.3	34.4	102.2	7.6	38.8	59.4	10.1
		^{137}Cs		52.9		132.0	9.1		82.6	11.6
		^{40}K		286.6		190.6	71.8		195.7	106.5

[a]Bq/kg wet weight for cabbage and Bq/kg dry weight for soil

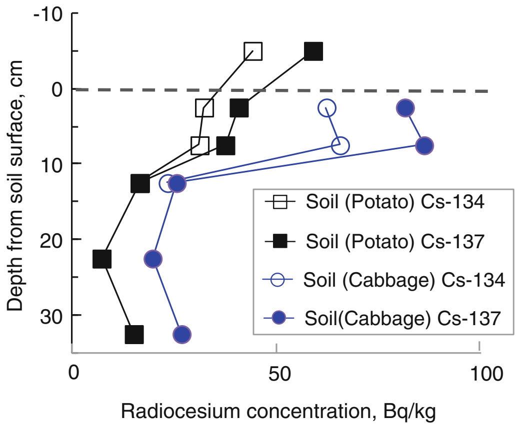

Fig. 7.1 Concentration of ^{134}Cs and ^{137}Cs at each depth in soil observed on 9 June 2011 at Institute for Sustainable Agro-ecosystem Services in Nishitokyo City

Table 7.3 Concentration of ^{134}Cs, ^{137}Cs and ^{40}K of potato in Fukushima Prefecture (99 days after planting)

	Tuber, washed (n = 1[b])			Aerial part (leaf + stem), washed (n = 1)		
Nuclide	Moisture content (% w.b.)	Concentration[a]	Detection limit[a]	Moisture content (% w.b.)	Concentration[a]	Detection limit[a]
^{134}Cs	78.4	3.4	1.2	79.2	58.6	5.2
^{137}Cs		3.3	1.1		52.7	5.0
^{40}K		305.1	13.7		581.0	47.5

[a]Bq/kg wet weight
[b]n = 1 means the mean value of tubers with 2.8 kg wet weight

An extremely small amount of radiocesium (^{134}Cs and ^{137}Cs) was detected in the potato tuber sample (Table 7.3). In this case, ^{134}Cs and ^{137}Cs in the soil at depths from 0 to 5 cm were detected at 568.6 and 666.4 Bq/kg dry weight (total of 1,235 Bq/kg dry weight), respectively (Fig. 7.2). With regard to the transfer of radiocesium, one study investigated the transfer rate from plant roots to organs in hydroponic conditions (Uchida et al. 1987).

They found that in root crops and leaf vegetables, the transfer rate of ^{137}Cs, determined by (activity of plant sample/day)/(mean activity of culture solution), to roots is faster by about one order of magnitude than that to other plant organs located above the ground. In contrast, in root vegetables (daikon radish) growing in soil culture, the transfer factor for leaves is higher than that for tubers, with a ratio of 4.1 for ^{137}Cs (Ban-nai and Muramatsu 2002). Regardless of which hypothesis is adopted, the concentration of radiocesium in the aerial parts shown in Table 7.3 was thought to be too high to have resulted from uptake through root absorption. Thus, the high concentration was assumed to be due to the deposition of radioactive fallout.

The transfer factor from soil to potato tubers was calculated for reference. Based on the concentrations of ^{137}Cs in the tuber and soil samples (583 Bq/kg dry weight as a mean value for vertical positions from −10 to 10 cm; not shown in Fig. 7.2), the transfer

Table 7.4 Concentration of ^{134}Cs, ^{137}Cs and ^{40}K of cabbage in Fukushima Prefecture (78 days after planting)

	Edible part, unwashed (n=2)			Edible part, washed (n=2)			Outer leaf, washed (n=2)			Root, washed (n=4)		
Nuclide	Moisture content (% w.b.)	Concentration[a]	Detection limit[a]	Moisture content (% w.b.)	Concentration[a]	Detection limit[a]	Moisture content (% w.b.)	Concentration[a]	Detection limit[a]	Moisture content (% w.b.)	Concentration[a]	Detection limit[a]
^{134}Cs	93.8	N.D.	2.7	93.3	N.D.	4.10	92.4	N.D.	3.7	88.5	N.D.	17.1
^{137}Cs			2.7			4.47			3.5			16.5
^{40}K		230.3	34.5		230.65	48.90		309.2	45.0		N.D.	191.6

[a]Bq/kg wet weight

Table 7.5 Concentration of ^{134}Cs, ^{137}Cs and ^{40}K of lettuce in Fukushima Prefecture (49 days after planting)

	Edible part, unwashed (n = 3)			Edible part, washed (n = 3)			Root, washed (n = 1)		
Nuclide	Moisture content (% w.b.)	Concentration[a]	Detection limit[a]	Moisture content (% w.b.)	Concentration[a]	Detection limit[a]	Moisture content (% w.b.)	Concentration[a]	Detection limit[a]
^{134}Cs	95.2	N.D.	5.2	94.9	N.D.	4.4	88.9	N.D.	5.5
^{137}Cs		N.D.	6.4		N.D.	5.3		6.1	5.3
^{40}K		192.7	60.7		171.5	52.5		211.8	62.9

[a]Bq/kg wet weight

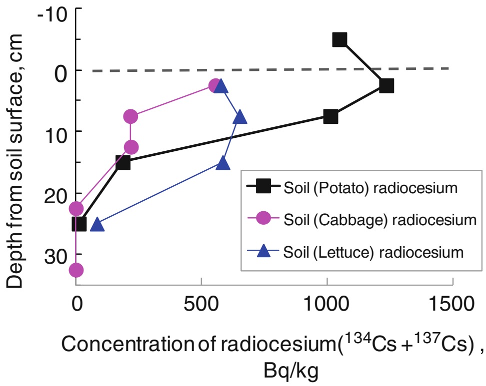

Fig. 7.2 Concentration of radiocesium (^{134}Cs + ^{137}Cs) at each depth in soil observed on 18 July 2011 in Fukushima Prefecture

factor was estimated to be 0.0057 using the following equation: (^{137}Cs concentration in tuber; fresh vegetable)/(^{137}Cs concentration in soil; dry soil). This value was smaller than the index value of 0.067 (http://www.maff.go.jp/j/press/syouan/nouan/110527.html) provided by the Japanese Ministry of Agriculture, Forestry and Fisheries (MAFF), which was derived by the same equation. When the sum of the ^{134}Cs and ^{137}Cs concentrations was used in this calculation, the transfer coefficient was 0.0061.

In cabbage, no radiocesium was detected in samples of the outer leaf (washed) and root (washed). In lettuce, a very small amount of ^{137}Cs was detected in a root sample but not in any other plant organ sample. In these cases, the maximum concentrations of ^{134}Cs and ^{137}Cs in the soil were found at depths ranging from 0 to 5 cm of 291.8 and 359.4 Bq/kg dry weight (total of 651.2 Bq/kg dry weight), respectively (Fig. 7.2). Data provided by MAFF in 2011 yielded the transfer factor of radiocesium from soil to vegetables in which the concentrations in soil were close to the values observed in Fukushima in our investigation (Ministry of Agriculture, Forestry and Fisheries, http://www.s.affrc.go.jp/docs/nogyo_gizyutu/pdf/3_1.pdf). Although we did not observe the transfer of radiocesium from the soil to cabbage or lettuce, it will be important to continue obtaining data on the transfer of radiocesium from soil with low contamination for further study.

The measured concentrations of radioactive nuclides in the soil showed a vertical distribution pattern within the plow layer for ^{134}Cs and ^{137}Cs, with surface soil having a high concentration level (Fig. 7.2). These results corresponded with the results obtained in Nishitokyo City (Fig. 7.1).

7.4 Conclusion

We investigated the transfer of radiocesium from soil with low contamination to vegetables. An extremely small amount of ^{137}Cs was detected in the tuber samples of potatoes collected 58 days after they were planted in a dry field with a ^{137}Cs concentration of 50–100 Bq/kg dry weight. However, radiocesium was not detected in potato samples collected 82 days after planting. Potassium is known to be transported in plants to adjust the osmotic pressure in cells. We suspected that cesium behaved in much the same way as potassium, which resulted in a lower concentration in potatoes at harvesting season.

In potatoes grown in a dry field with a ^{137}Cs concentration of 530–670 Bq/kg dry weight, a very small amount of ^{137}Cs was transferred to the tubers. This transfer coefficient was calculated to be 0.0057, although this should be considered as a reference value only. This value is smaller than the index value provided by the Japanese MAFF. However, it should be noted that this index value was determined from data in the range of 0.00047–0.13. In this investigation, any detectable transfer of cesium from the soil to the edible parts of cabbage or lettuce was observed. Because the transfer factor varies depending on soil characteristics, it will be necessary to collect more data for soil with a low contamination level and conduct more studies on the transfer of radiocesium from soil to vegetables in the fields where agricultural production is continuing.

Our investigation of soil did indicate a vertical distribution pattern for radiocesium concentrations, with a higher concentration in surface soil. We anticipate that by making the concentrations of radiocesium more uniform (reduced) by land cultivation, deep plowing or other soil mixing methods, the transfer of radiocesium will be reduced.

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Chapter 8
Radioactivity in Agricultural Products in Fukushima

Naoto Nihei

Abstract The radioactivity measurements performed by Fukushima local government, from March 2011 to March 2012, is presented. Agricultural products (cereals, vegetables, and fruit trees), forest products (mushrooms and edible wild plants), marine products (saltwater and freshwater fish), and stock farm products (beef, pork, and raw milk) were measured for radioactivity before shipment. In March 2011, 19% of the samples contained >500 Bq/kg of radiocesium, followed by 12% in April, 10% in May, 7% in June, and 1–2% after July. Only 3% of the samples investigated contained >500 Bq/kg, whereas the remaining 97% contained less than the provisional regulation level of 500 Bq/kg in 2011.

Keywords Agricultural products • Forest products • Marine products • Monitoring • Radiocesium • Radioiodine • Stock farm products

8.1 Introduction

Fukushima Prefecture is the third largest in Japan in terms of area. Fukushima Prefecture is blessed natural environment, and produces agricultural and livestock including rice, cucumbers, tomatoes, green beans, asparagus, peaches, nashi pears, autumn bellflowers, baby's breath flowers, beef, Japanese chicken, and fishery products such as Pacific saury, flounder, and carp. All the produce from Fukushima Prefecture was of high ranking in Japan.

The Tohoku Region Pacific Coast earthquake occurred on March 11, 2011, at 14:46 h. Immediately after the earthquake, accidents occurred at the Fukushima Daiichi power station operated by the Tokyo Electric Power Company. Radioactive

N. Nihei (✉)
Department of Agriculture, Forestry, and Fisheries, Fukushima Prefecture, Japan
e-mail: nihei_naoto_01@pref.fukushima.lg.jp

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Fig. 8.1 The germanium semiconductor detectors at Fukushima Agricultural Technology Centre

materials released during the accident reached farmlands in Fukushima and neighboring prefectures and contaminated the soil and agricultural products. To ensure the safety of agricultural, forestry, and marine products, "Environmental Radiation level Emergency Monitoring for Agricultural, Forestry, and Fishery Products" (hereafter referred to as monitoring) was implemented as an emergency response measure by the government's Nuclear Emergency Response Headquarters under the Act on Special Measures Concerning Nuclear Emergency Preparedness. Monitoring was performed before the shipment of agricultural product. After considering the quantity of production, the value of shipments, and the possible intake levels, the products and the sampling sites were decided in consultation with municipalities. The investigative program required almost 1 week for each item: agricultural products (cereals, vegetables, and fruit trees), forest products (mushrooms and edible wild plants), marine products (saltwater and freshwater fish), and stock farm products (beef, pork, and raw milk).

The extracted samples were washed with tap water and the edible portions were chopped finely. The samples were packed in a container and ten samples were measured at a time using the germanium semiconductor detector (CANBERRA) at Fukushima Agricultural Technology Centre (Fig. 8.1). The detection limit for radiocesium was approximately 10 Bq/kg. The Ministry of Health, Labour, and Welfare specified provisional guideline levels for radioactive materials in foods under the Food Sanitation Law immediately after the disaster. The provisional guideline levels for radiocesium were 200 Bq/kg in raw milk and 500 Bq/kg in cereals, vegetables, meat, fish, and shellfish. The concentration of radioiodine was 300 Bq/kg in raw milk and 2,000 Bq/kg in vegetables, fish, and shellfish. There are no provisional regulation levels for radioiodine in cereals and meat. If radioactive materials were detected at levels exceeding the provisional guidelines, the government would ask the municipalities to suspend shipments or limit consumption. The results of the monitoring have been released on the homepages of Fukushima Prefecture and the Ministry of Health, Labour, and Welfare.

Fukushima Prefecture: http://www.new-fukushima.jp/monitoring.php
Ministry of Health, Labour, and Welfare: http://www.mhlw.go.jp/stf/houdou/2r9852000001m9tl.html

By the end of March 2012, approximately 450 items were monitored to produce 19,000 data points (Table 8.1). This paper describes the impact of the radioactive materials dispersed during the disaster on agricultural products based on the results of the monitoring from the disaster to March 2012.

8.2 Radioactive Material Concentration in Each Type of Agricultural Product

We considered the samples that were harvested from March to June 2011 and from July to March 2012 (Table 8.2). The results varied greatly depending on time. Although the provisional guideline level for radiocesium was 500 Bq/kg, the new legal guideline level was reduced to 100 Bq/kg on April 1, 2012. The radiocesium concentration in agricultural products was classified as below the detection limit, from the detection limit to 100 Bq/kg, from 100 to 500 Bq/kg, and >500 Bq/kg.

8.3 Samples Harvested from March to June 2011

Figure 8.2 shows the concentration of radiocesium and radioiodine in the samples harvested from March to June 2011.

The maximum concentration of radiocesium in spinach (*Spninacia oleracea* L.) was 34,000 Bq/kg. Of the spinach samples harvested in March 2011, 10% contained radiocesium less than the detection limit, whereas 85% contained >500 Bq/kg. In April, 18% contained radiocesium less than the detection limit and 26% contained >500 Bq/kg. In May, 58% contained radiocesium less than the detection limit and no sample contained >500 Bq/kg. The maximum concentration of radioiodine in spinach was 8,400 Bq/kg. In March 2011, 80% of the spinach samples contained >2,000 Bq/kg of radioiodine, which dropped to 5% in April and to 0% in May 2011. The concentration of radiocesium in broccoli [*Brassica oleracea* L. (italica group)] showed the same monthly changes as spinach; the maximum concentration of radioiodine was 4,400 Bq/kg. The maximum concentration of radiocesium in cucumber

Table 8.1 The number of monitoring samples tested in each month

	2011										2012			
	March	April	May	June	July	August	September	October	November	December	January	February	March	Total
Vegetables and fruit trees	115	376	404	608	720	730	733	1,008	708	294	110	135	180	6,121
Cereals	0	0	0	0	43	104	1,170	802	192	22	0	1	0	2,334
Stock farm products	142	89	81	75	116	138	763	819	733	718	593	794	827	5,888
Forest products	21	103	214	92	55	81	197	220	25	42	10	9	14	1,083
Marine products	2	18	80	221	248	282	338	420	495	237	186	581	449	3,557
Total	280	586	779	996	1,182	1,335	3,201	3,269	2,153	1,313	899	1,520	1,470	18,983

Table 8.2 The analyzed food products

Classification		Investigation period: March 2011 to June 2011	July 2011 to March 2012
Vegetables and fruit trees	Leafy vegetables	Spinach, broccoli	Spinach, broccoli, cabbage Chinese chives, Chinese cabbage
	Fruit vegetables	Cucumber	Cucumber, tomato, strawberry, pumpkin, tepary bean, eggplant
	Stem and root vegetables	–	Asparagus, sweet potato, colocasia esculenta, Japanese radish, carrot, potato
	Fruit trees	Plum	Peach, pear, persimmon, apple, grape, chestnut, kiwi fruit
Cereals		–	Wheat, rice, buckwheat, soybean, azuki bean
Stock farm products	Meat	Beef, pork, chicken	Beef, pork, chicken
	Other	Original milk, chicken egg	Original milk, chicken egg
Forest products		Mushroom (shiitake mushroom, maitake mushroom etc.)	Mushroom (shiitake mushroom, maitake mushroom etc.)
Marine products		Saltwater fish (olive flounder, righteye flounder etc.) freshwater fish (ayu, Japanese dace, seema etc.)	Saltwater fish (olive flounder, fat greenling, righteye flounder etc.) freshwater fish (Japanese char, chub, pond smelt etc.)

(*Cucumis sativus* L.) was 27 Bq/kg. In March 2011, 50% of the cucumber samples were below the detection limit, which increased to 92% in April, 95% in May, and 100% in June. The maximum concentration of radioiodine was 36 Bq/kg. Radioiodine was detected in March and April but not in May or the subsequent months. The concentrations of radioactive material in leafy vegetables such as spinach and broccoli were the highest in March. Immediately after the accident, the crops were contaminated by the direct deposition of radioactive materials. Thus, high concentrations of radioactive materials were detected in cultivated leafy vegetables. But they declined over time. The reduction in the concentration of ^{131}I, with a short half-life, and the reduction in radioactive emissions from the damaged nuclear power station have gradually reduced the effects of direct contamination. On the other hand, the concentration of cucumber which was also grown at the disaster site was lower than that of spinach and broccoli because the measured component (the edible part) was not the leaf, which were most affected by the deposition of radioactive material, but the fruit.

Plum (*Prunus mume*) sampling commenced in May 2011 and the maximum concentration of radiocesium was 760 Bq/kg. In May 2011, 25% of plum samples contained >500 Bq/kg of radiocesium, which dropped to 14% in June, whereas

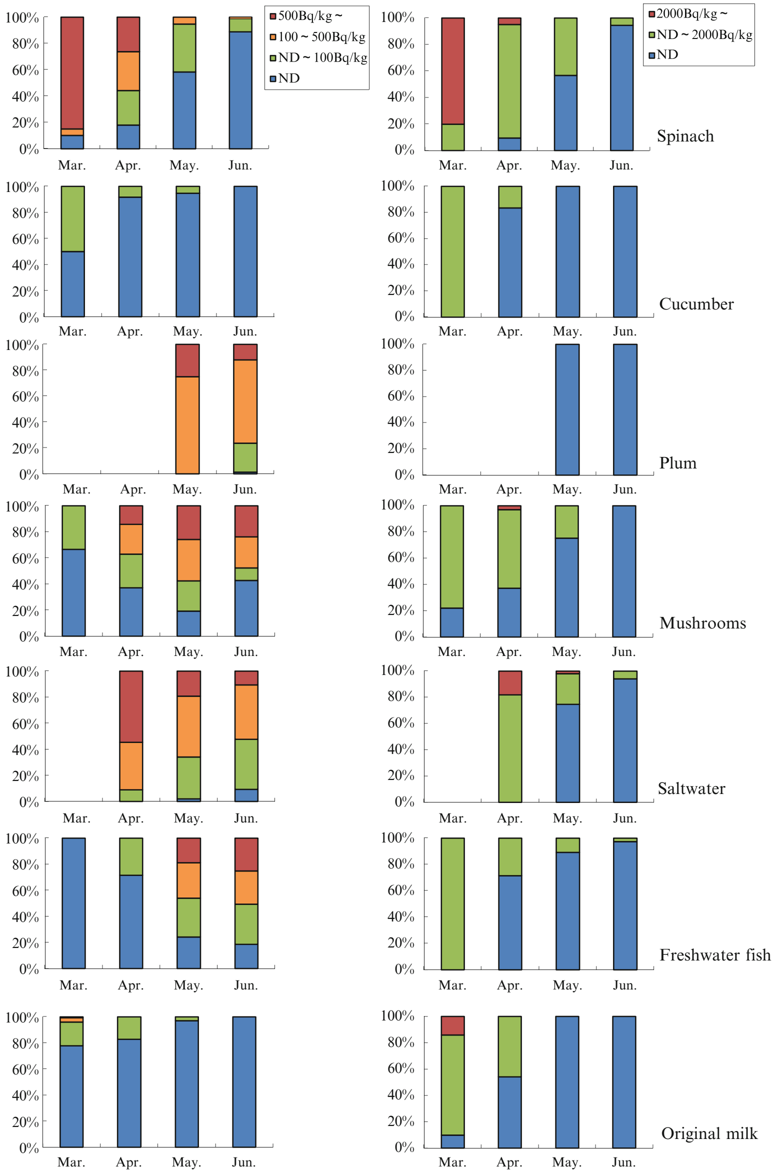

Fig. 8.2 Radioactive material concentrations in different products (from March 2011 to June 2011). *Left figures*: radiocesium; *right figures*: radioiodine

radioiodine was not detected. Because the plums were still in the flowering season at the time of the disaster, radiocesium did not affect the fruits directly. However, because the radioactive material was deposited on the entire tree, the concentration in the fruit was >500 Bq/kg initially.

The edible mushrooms that were investigated comprised shiitake mushrooms (*Lentinula edodes*), maitake mushrooms [*Grifola frondosa* (Dicks. ex Fr.) S. F. Gray], and other types. The concentration of radiocesium in mushroom sample did not exceed 500 Bq/kg in March 2011. However, 14% of the mushroom samples harvested in April contained >500 Bq/kg, followed by 26% in May and 24% in June. The maximum concentration of radiocesium was 12,000 Bq/kg and the maximum concentration of radioiodine was 13,000 Bq/kg. The high concentration of radioactive material in mushrooms agreed with previous reports (Battiston et al. 1989; Sugiyama et al. 1993). The radiocesium concentration in mushroom remained high even after May. The high mushroom concentration was not only because of the direct deposition of radioactive material on mushrooms but also because mushrooms actively absorbed and accumulated radiocesium from the soil.

The monitoring of saltwater fish commenced in April 2011. Olive flounder (*Paralichthys olivaceus*), fat greenling (*Hexagrammos otakii*), righteye flounder (*Pleuronectidae*), and other species were inspected. None of the saltwater fish sample harvested in April 2011 contained radiocesium less than the detection limit, whereas 45% contained radiocesium ranging from the detection limit to 500 Bq/kg, and the remaining 55% contained >500 Bq/kg. In May, 2% contained radiocesium less than the detection limit, 79% contained radiocesium ranging from the detection limit to 500 Bq/kg, and 19% contained >500 Bq/kg. In May, 9% contained radiocesium less than the detection limit, 80% contained radiocesium ranging from the detection limit to 500 Bq/kg, and 11% contained >500 Bq/kg. The maximum concentration of radiocesium was 14,400 Bq/kg. In April, 82% contained radioiodine ranging from the detection limit to 2,000 Bq/kg, whereas 18% contained >2,000 Bq/kg. In May, 74% contained less than the detection limit, 23% contained radioiodine ranging from the detection limit to 2,000 Bq/kg, whereas 2% contained >2,000 Bq/kg. In June, 94% contained less than the detection limit, 6% contained radioiodine ranging from the detection limit to 2,000 Bq/kg, and none had >2,000 Bq/kg. The maximum concentration of radioiodine was 12,000 Bq/kg.

Freshwater fish were harvested from rivers in the prefecture and the species inspected included ayu (*Plecoglosseus altivelis*), Japanese dace (*Tribolodon hakonensis*), and seema (*Oncorhynchus masou masou*). In March, 100% contained radiocesium less than the detection limit. In April, 71% contained radiocesium less than the detection limit and 29% contained radiocesium ranging from the detection limit to 100 Bq/kg. In May, 27% contained radiocesium ranging from 100 to 500 Bq/kg and 19% contained >500 Bq/kg. In June, 25% contained radiocesium ranging from 100 to 500 Bq/kg and 25% contained >500 Bq/kg. The radiocesium concentration in freshwater fish increased after May. In March, 100% contained radioiodine ranging from the detection limit to 2,000 Bq/kg. In April, 71% contained less than the detection limit, 29% contained radioiodine ranging from the detection limit to 2,000 Bq/kg. In May, 89% contained less than the detection limit and 11% contained radioiodine ranging from the detection limit to 2,000 Bq/kg. In May, 97% contained less than the detection limit and 3% contained radioiodine ranging from the detection limit to 2,000 Bq/kg. In contrast to the radiocesium concentration, the radioiodine concentration increased from March. However, the concentration was not >2,000 Bq/kg in any sample.

The analysis of saltwater fish commenced in April. Levels with >500 Bq/kg were measured, with the highest in April, and a few had >500 Bq/kg in May and June. Radioiodine was also detected in April. Radioiodine has a short half-life and its concentration in saltwater fish decreased in May and in subsequent months. The radiocesium concentration in freshwater fish increased after May. It will be necessary to monitor these levels in detail in future. The increase in the radiocesium concentration from May was attributed to rainwater and/or groundwater flowing into rivers where radiocesium concentrated, which also increased the concentration in freshwater fish.

Raw milk was investigated at each collection centre (air-conditioned station). In March 2011, 78% of the raw milk collected contained radioactive material less than the detection limit, 21% contained radioactive material ranging from the detection limit to 500 Bq/kg, and 1% contained >500 Bq/kg. In May, 83% contained less than the detection limit and 17% contained radioactive material ranging from the detection limit to 500 Bq/kg. In June, 97% contained less than the detection limit and 3% contained radioactive material ranging from the detection limit to 500 Bq/kg. Radiocesium was not detected in raw milk in June or the subsequent months. In March 2011, 10% of the raw milk contained radioiodine less than the detection limit, 76% contained radioiodine ranging from the detection limit to 300 Bq/kg, and 14% contained >300 Bq/kg.

8.4 Samples Harvested from July 2011 to March 2012

Figure 8.3 shows the radiocesium concentrations in sample collected between July 2011 and March 2012. Radioiodine was not detected after July 2011.

8.4.1 Cereals

During the test period, 54% of wheat (*Triticum aestivum* L.) samples contained less than the detection limit and 1% contained >500 Bq/kg. Wheat is usually sown in October and harvested in July. When the nuclear power plant accident occurred in March, the wheat was growing on cultivated land and the radioactive material deposited on it directly (Tanoi et al. 2011). Therefore, the radioactive material concentration in wheat should have been slightly higher than that in other agricultural products.

It was found that 88% of rice (*Oryza sativa* L.) samples contained less than the detection limit and 11% contained radiocesium ranging from the detection limit to 100 Bq/kg. Similarly, 89% of buckwheat (*Fagopyrum esculentum* Moench) samples contained less than the detection limit, whereas 10% contained radiocesium ranging from the detection limit to 100 Bq/kg. There was a report of rice samples containing nearly 500 Bq/kg, which were outside our monitoring area. Thus, we performed a detailed reanalysis (survey = 20,387 samples) in a limited area. In the reanalysis,

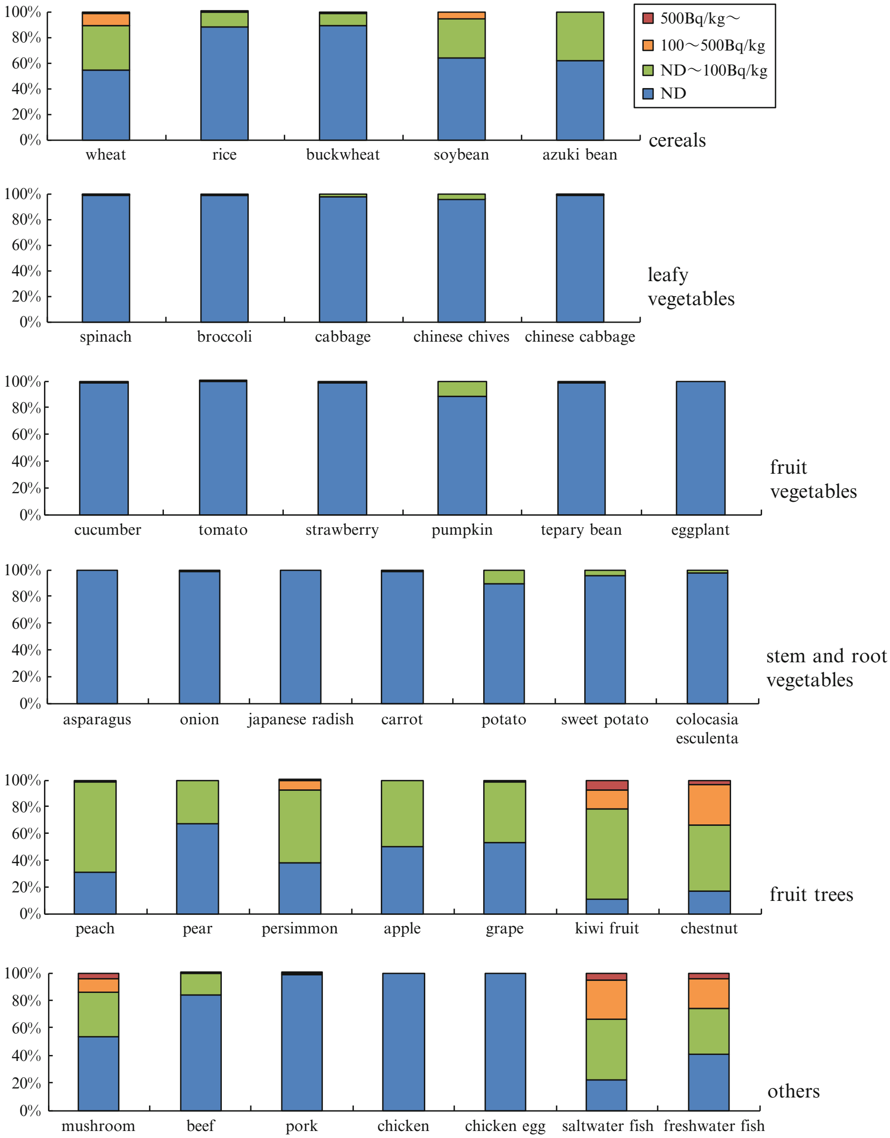

Fig. 8.3 Radiocesium concentrations in different products (from July 2011 to March 2012)

89% of rice sample contained less than the detection limit, 10% contained radiocesium ranging from the detection limit to 100 Bq/kg, and 1% contained >500 Bq/kg. The maximum concentration of radiocesium in rice was 550 Bq/kg.

We found that 64% of soybean [*Glycine max* (L.) Merr.] samples contained less than the detection limit, 30% contained radiocesium ranging from the detection limit to 100 Bq/kg, and 6% contained radiocesium ranging from 100 to 500 Bq/kg. Similarly, 61% of azuki bean [*Vigna angularis* (Wild.) Ohwi et Ohashi] samples contained less than the detection limit and 39% contained radiocesium ranging from the detection limit to 100 Bq/kg. We compared soybeans and azuki beans with other agricultural products, such as rice and vegetables which were grown after the nuclear power plant accident and their radioactive material concentrations were high. Thus, the factors determining these high levels must be addressed in future.

8.4.2 Vegetables

The radioactive material concentration of spinach and broccoli, detected from March to June, were less than the detection limit value. With the exceptions of pumpkins (*Cucurbita*) and potatoes (*Solanum tuberosum* L.), vegetables contained less than the detection limit. The vegetables harvested after July 2011 were not cultivated when the radioactive material was deposited after the nuclear accident. The radioactive material transfer from soil to vegetable was low, and thus, we considered that their radioactive material content was also low.

8.4.3 Fruit Trees

We found that 30% of peach [*Prunus persica* (L.) Batsch] samples contained less than the detection limit, 69% contained radiocesium ranging from the detection limit to 100 Bq/kg, and 1% contained radiocesium from 100 to 500 Bq/kg. The maximum concentration of radiocesium in peach was 161 Bq/kg.

We found that 68% of pear (*Pyrus pyrifolia* var. *culta*) samples contained less than the detection limit and 32% contained radiocesium ranging from the detection limit to 100 Bq/kg. The maximum concentration of radiocesium was 48 Bq/kg.

We found that 38% of persimmon (*Diospyros kaki*) samples contained less than the detection limit, 55% contained radiocesium ranging from the detection limit to 100 Bq/kg, 7% contained radiocesium ranging from 100 to 500 Bq/kg, and 1% contained >500 Bq/kg. The maximum concentration of radiocesium was 670 Bq/kg.

We found that 50% of apple (*Malus domestica*) samples contained less than the detection limit and 50% contained radiocesium ranging from the detection limit to 100 Bq/kg. The maximum concentration of radiocesium was 46 Bq/kg.

We found that 53% of grape (*Vitis*) samples contained less than the detection limit and 46% contained radiocesium ranging from the detection limit to 100 Bq/kg. The maximum concentration of radiocesium was 57 Bq/kg.

We found that 17% of chestnut (*Castanea crenata* Sieb. et Zucc.) samples contained less than the detection limit, 49% contained radiocesium ranging from the detection limit to 100 Bq/kg, 31% contained radiocesium ranging from 100 to 500 Bq/kg, and 3% contained >500 Bq/kg. The maximum concentration of radiocesium was 940 Bq/kg.

We found that 11% of kiwi fruit (*Actinidia chinensis* Planch) samples contained less than the detection limit, 68% contained radiocesium ranging from the detection limit to 100 Bq/kg, 14% contained radiocesium ranging from 100 to 500 Bq/kg, and 7% contained >500 Bq/kg. The maximum concentration of radiocesium was 510 Bq/kg.

The fruit collected from the fruit trees were not contaminated directly with radioactive material but radioactive material was deposited on the bark. Thus, the radioactive material must have translocated from the bark to the fruit, although there were differences in the level of translocation among fruit types.

8.4.4 Other Products (Mushrooms, Meat, and Fish)

We found that 53% of mushroom samples contained less than the detection limit, 32% contained radiocesium ranging from the detection limit to 100 Bq/kg, 9% contained radiocesium ranging from 100 to 500 Bq/kg, and 5% contained >500 Bq/kg. The maximum concentration of radiocesium was 28,000 Bq/kg.

The meat samples inspected included beef, chicken, and pork. We found that 84% of beef samples contained less than the detection limit and 15% contained radiocesium ranging from the detection limit to 100 Bq/kg. However, 98% of pork samples contained less than the detection limit, whereas 100% of chicken sample contained less than the detection limit. We considered that the radioactive material contamination of beef was due to the feed being contaminated with radioactive material. Therefore, we need to ensure that the rice straw and grass left outside at the time of the nuclear accident is not fed to cows or pigs. This guidance was promoted to prevent the spread of contamination to meat.

We found that 100% of the raw milk samples contained less than the detection limit from July (data not shown). Chicken eggs were also below the detection limit.

The saltwater fish and shellfish analyzed included fat greenling (*H. otakii),* righteye flounder, common skate (*Raja kenojei*), Pacific cod (*Gadus macrocephalus*), Japanese abalone [*Haliotis* (*Sulculus*) *diversicolor aquatilis*], and others. We found that 22% of the saltwater fish and shellfish samples contained less than the detection limit, 45% contained radiocesium ranging from the detection limit to 100 Bq/kg, 28% contained radiocesium ranging from 100 to 500 Bq/kg, and 6% contained >500 Bq/kg. The maximum concentration of radiocesium was 4,500 Bq/kg.

The freshwater fish we analyzed included ayu (*Plecoglossus altivelis altivelis*), raised char (*Salvelinus leucomaenisand*), seema (*O. masou masou*), and Japanese smelt (*Hypomesus nipponensis*). We found that 41% of the freshwater fish samples contained less than the detection limit, 33% contained radiocesium ranging from the detection limit to 100 Bq/kg, 22% contained radiocesium ranging from 100 to 500 Bq/kg, and 4% contained >500 Bq/kg. The maximum concentration of radiocesium was 18,700 Bq/kg.

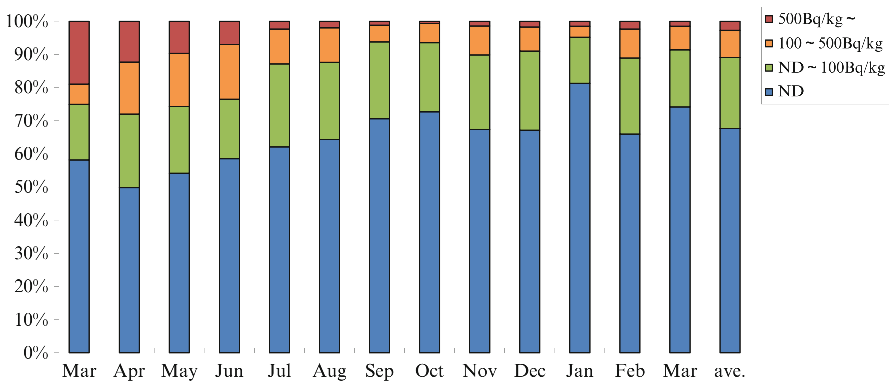

Fig. 8.4 Radiocesium concentrations in all monitoring samples

8.5 Conclusion

The radiocesium concentrations in the samples investigated from March 2011 to March 2012 are shown in Fig. 8.4. In March 2011, 19% of the samples contained >500 Bq/kg of radiocesium, followed by 12% in April, 10% in May, 7% in June, and 1–2% after July. Only 3% of the samples investigated contained >500 Bq/kg, whereas the remaining 97% contained less than the provisional guideline level of 500 Bq/kg.

The provisional levels for foods were reviewed and a new criterion of radiocesium concentration was set as 100 Bq/kg on April 1, 2012. After July 2011, 8% of the samples investigated exceeded the new criterion, i.e., >100 Bq/kg. The long-term problem of radiocesium contamination of soil has to be solved in future, particularly ^{137}Cs with a half-life of 30 years. In future, the farming of highly contaminated areas requires active decontamination practices, such as topsoil removal, deep plowing, and soil-turning tillage. Depending on the soil conditions, the application of potassium fertilizers and the addition of adsorbents may be effective methods for inhibiting any further uptake of radiocesium by plants. It is also important to understand the long-term behavior of radioactive materials based on the monitoring of contamination. Therefore, we aim to promote the revival of agriculture in Fukushima.

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Chapter 9
Changes in the Transfer of Fallout Radiocaesium from Pasture Harvested in Ibaraki Prefecture, Japan, to Cow Milk two Months After the Fukushima Daiichi Nuclear Power Plant Accident

Noboru Manabe, Tomotsugu Takahashi, Jun-You Li, Keitaro Tanoi, and Tomoko M. Nakanishi

Abstract Changes in the radioactivity of ^{131}I, ^{134}Cs, and ^{137}Cs in milk produced by cows given pasture that was contaminated with these radioactive nuclides caused by the Fukushima Daiichi nuclear power plant accident on 11 March 2011, were examined between 16 May and 26 June 2011. Pasture (Italian ryegrass) was seeded on September 2010, and cultivated in the Animal Resource Science Center of the University of Tokyo (about 140 km south-west of the power plant). Pasture was harvested 2 months after the accident and prepared for fermented grass forage (haylage). The cows examined were born and kept in the Animal Resource Science Center and were given commercial mixed feed (total mixed ration forage: TMR) that contained no radioactive ^{131}I, ^{134}Cs, or ^{137}Cs, for 2 weeks before being examined. They were given haylage and TMR (10 and 25 kg/600 kg of body weight/day, respectively) for 2 weeks, and then were given only TMR (35 kg/600 kg of body weight/day) for 2 weeks. During the examination, milk was collected twice a day and mixed in each cow. The weight and radioactivity of ^{131}I, ^{134}Cs, and ^{137}Cs of the mixed milk in each cow were measured daily. No radioactive ^{131}I was detected in either the milk or haylage. The radioactivity of ^{134}Cs and ^{137}Cs contained in the mixed feed of haylage and TMR was 380 Bq/kg (radiocesium radioactivity was represented as total concentrations of ^{134}Cs and ^{137}Cs). Radiocesium radioactivity concentrations in the milk rapidly increased to 30 Bq/kg after 4 days from the start of feeding and equilibrated to 36 Bq/kg after 12 days. After that, cows were given TMR containing no radiocesium, and radioactivity concentrations of radiocesium in the milk rapidly decreased. Two weeks after stopping radiocesium feeding, radioactivity concentrations were less than 5 Bq/kg (background level). In summary, when the

N. Manabe (✉) • T. Takahashi • J.-Y. Li
Animal Resource Science Center, Graduate School of Agricultural and Life Sciences,
The University of Tokyo, Kasama 319-0206, Japan
e-mail: amanabe@mail.ecc.u-tokyo.ac.jp

K. Tanoi • T.M. Nakanishi
Graduate School of Agricultural and Life Sciences,
The University of Tokyo, 1-1-1 Yayoi, Bunkyo-ku, Tokyo 113-8657, Japan

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cow (approximately 600 kg of body weight) was given feed with radioactive ^{134}Cs and ^{137}Cs (12,600 Bq/600 kg body weight/day), 5.71% of ^{134}Cs and ^{137}Cs was secreted into the milk (720 Bq/20 kg milk/day). Radioactivity concentrations of radiocesium in the milk were lower than new standard (50 Bq/kg) of Japan.

Keywords Cow milk • Fukushima Daiichi nuclear power plant accident • Pasture • Radiocaesium

9.1 Introduction

On 11 March 2011, the declaration of a nuclear emergency situation was issued by the Japanese Prime Minister because of the Fukushima Daiichi nuclear power plant accident that resulted from the East Japan earthquake. On 17 March 2011, the Japanese Ministry of Health, Labour, and Welfare established a provisional regulation level for cow milk that was less than 70 Bq/kg radioactive iodine and 200 Bq/kg radiocesium (^{134}Cs and ^{137}Cs). On 1 April 2012, a more stringent new standard for milk of 50 Bq/kg radiocesium was established. However, under livestock feeding management conditions in Japan, it has not been fully understood (1) how much radioactive nuclides transfer from the haylage to the milk, when dairy cows are given grass haylage prepared from pasture grown under nuclear power plant accident conditions, or (2) after that, whether radioactive nuclides rapidly disappear from the milk or not in cows are given forages without radioactive nuclides. In order to propose appropriate risk management related to the production of cow milk, which is an excellent food for infants and children who are considered to have high sensitivity to radiation, we performed the present study at the Animal Resource Science Center of The University of Tokyo (Kasama, Japan), which is located about 140 km south-west in a straight-line from the Fukushima Daiichi nuclear power plant.

9.2 Experimental Procedure

(a) Preparation of cow feed containing radioactive nuclides caused by the accident in the Fukushima Daiichi nuclear power plant: grass (Italian ryegrass, *Lolium multiflorum* Lam.) was seeded in October 2010, and cultivated in a field at the Animal Resource Science Center. After mowing between 10 and 15 May 2012 (2 months after the accident), raw grass (^{131}I: not detectable level and ^{134}Cs and ^{137}Cs: 54 and 59 Bq/kg, respectively) was dried for several days, and then packed in plastic film to prepare anaerobically fermented grass forage (haylage). Haylage contained radioactive nuclides (^{131}I: not detectable level. ^{134}Cs and ^{137}Cs: 600 and 660 Bq/kg, respectively).
(b) Cow feed without radioactive nuclides caused by the nuclear power plant accident: commercial mixed feed (total mixed ration: TMR), which was purchased from Zen-Noh Feed (Tokyo, Japan), containing no detectable ^{131}I, ^{134}Cs, or ^{37}Cs

was used. TMR contains the required levels of nutrients (energy, protein, minerals, and vitamins) needed by the cow. The raw materials of TMR were as follows: 45% maize, 29% mixture of wheat bran and rice bran, 21% mixture of soybean oil cake and rapeseed oil cake, and 5% additives (minerals and vitamins). Feed ingredients of TMR were as follows: 16% crude protein, 2.5% crude fat, 10% crude fibers, 10% crude ash, 0.8% calcium, and 0.5% phosphorus.

(c) Lactating dairy cows: lactating dairy cows born and kept in the Animal Resource Science Center were used. According to a notice by the Government of Japan (MAFF 2011), cows were raised in a closed cow barn after 20 April 2012, and were given TMR.

(d) Study design: cows were given TMR (35 kg/600 kg body weight/day) for 2 weeks before the examination (between 16 and 29 May 2011), then they were divided into two groups (control and test groups). In the control group, three cows (636 ± 34 kg body weight) were given TMR for 4 weeks. In the test group, three cows (593 ± 23 kg body weight) were given a mixture of haylage and TMR (10 and 25 kg/600 kg of body weight/ day, respectively) for 2 weeks (between 30 May and 12 June 2011), and then were given only TMR for 2 weeks (between 13 and 26 June 2011). During the examination (from 30 May to 26 June 2011), cow milk was collected, weighed twice a day, and mixed in each cow. The radioactivity of ^{131}I, ^{134}Cs, and ^{137}Cs of the mixed milk in each cow was measured daily. The body weight of each cow was measured on the 16 and 30 May, and 13 and 27 June 2011. During the study period, cows were allowed to freely drink water through automatic water supply equipment. Drinking water had no detectable radioactivity from ^{131}I, ^{134}Cs, or ^{137}Cs.

(e) Test items: during the experiment, a veterinarian diagnosed the health status of cows. On the 16 and 30 May, and 13 and 27 June 2011, blood and urine biochemical tests and haematological analyses using automatic analyzers were carried. Radioactive element concentrations in each milk sample were measured by a germanium semiconductor detector, and each nuclide was identified by gamma-ray spectrometry. ^{134}Cs and ^{137}Cs were quantified at 661.6 and 604.7 keV, respectively, and then each Bq value was calculated by the calibration of count value. Each radionuclide concentration was calculated based on the weight of each sample. The detection limit was set to three times the standard deviation of the background.

9.3 Results

(a) Body weight, feed intake, and milk yield: throughout the study period, there were no significant differences in body weight, feed intake, or milk yield (Fig. 9.1) between the two experimental groups.

(b) Health status: during the experiment, no remarkable symptoms were found in any cows from the two experimental groups. There were no significant differences in

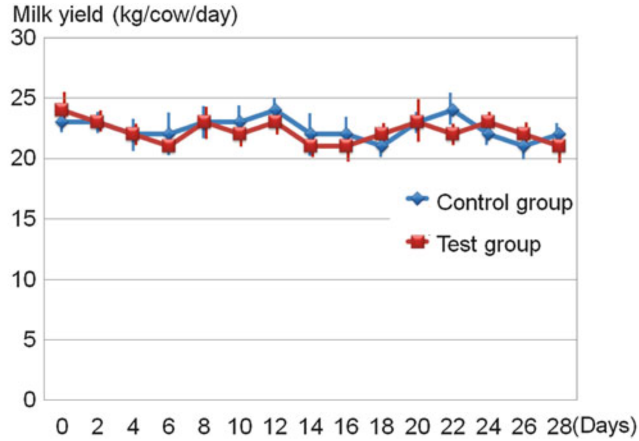

Fig. 9.1 Changes in milk yield (kg/cow/day). Milk yield was defined as the amount of twice morning and evening milking. Each point represents the mean ± standard deviation

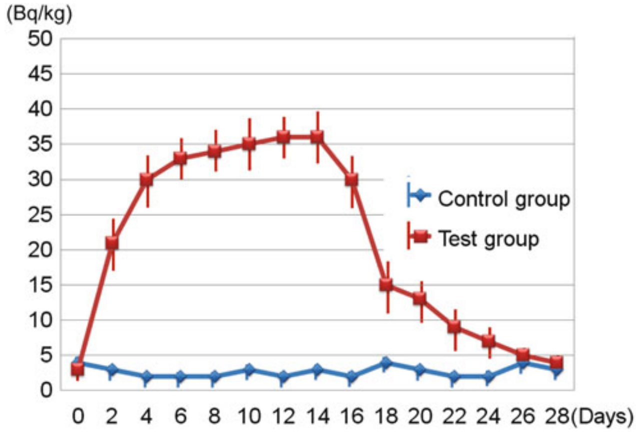

Fig. 9.2 Changes in concentrations of radiocesium in milk (Bq/kg). Each point represents the mean ± standard deviation

blood biochemical parameters, urine biochemical parameters, or haematological parameters between these two groups (data not shown). The cows used were healthy.

(c) Changes in the concentrations of radiocesium in milk: as described above, cows in the test group were given 35 kg/600 kg body weight/day of the mixture of haylage (not detectable level of ^{131}I, and 1,260 Bq/kg of radiocesium, ^{134}Cs and ^{137}Cs) for 2 weeks, and then were given only 35 kg/600 kg body weight/day of TMR for 2 weeks. As shown in Fig. 9.2, radioactivity concentrations of radiocesium in the milk rapidly increased to 30 Bq/kg after 4 days from the start of feeding, and equilibrated to 36 Bq/kg after 12 days. After stopping the feeding of forage with radiocesium, the concentration of radiocesium in the milk rapidly decreased at a rate of 2.05 Bq/kg/day. Two weeks after stopping the feeding of forage, radioactivity concentrations of radiocesium were less than 5 Bq/kg, similar to background levels.

9.4 Discussion

The present findings showed that when cows (approximately 600 kg of body weight) were given feed with radiocesium (12,600 Bq/600 kg body weight/day of ^{134}Cs and ^{137}Cs), 5.71% of ^{134}Cs and ^{137}Cs was secreted into the milk (720 Bq/20 kg milk/day) after 10–14 days from the start of feeding. These radioactivity concentrations of radiocesium in the milk were lower than Japanese Government new standards (50 Bq/kg for radiocesium). After that, cows were given feed containing no radiocesium, and radioactivity concentrations of radiocesium in the milk rapidly decreased. Two weeks after stopping the feeding of forage with radiocesium, radioactivity concentrations decreased to background levels of less than 5 Bq/kg.

The transfer coefficient (Fm) for radiocesium transfer from cow feed to milk was calculated according to the following formula: Fm (day/L) = radiocesium concentration in milk (Bq/L)/the amount of radiocesium intake in each cow (Bq/cow/day) (Sam et al. 1980; Johnson et al. 1988; Belli et al. 1993; Green and Woodman 2003; Robertson et al. 2003). In the present study, the radiocesium Fm value was 0.0029 (day/L) (36 Bq/L/12,600 Bq/cow/day) at the highest point of radiocesium concentrations in the milk. When cows were given haylage that was contaminated with radiocesium for 1 month after the Chernobyl nuclear power plant accident, the radiocesium Fm value in the milk was approximately 0.005 day/L (Voigt et al. 1989; Vreman et al. 1989; Green et al. 1994; Fabbri et al. 1994; Gastberger et al. 2001; Howard et al. 2001; Beresford et al. 2000). In Japan, when dairy cows were raised under Japanese-style management and given contaminated haylage caused by the Fukushima Daiichi nuclear power plant accident, radiocesium Fm values were between 0.0027 and 0.0064 day/L (Beresford and Howard 2011, MAFF 2011; Manabe et al. 2011; Manabe 2012; Takahashi et al. 2012). Compiled results by the International Atomic Energy Agency (IAEA) showed radiocesium Fm values of approximately 0.005 day/L (IAEA 2005, 2009, 2010). In our preliminary experiment, when cows were given radiocesium contaminated haylage caused by the Fukushima Daiichi nuclear power plant accident for 5 days, the Fm value was 0.00096 day/L (Hashimoto et al. 2011). Thus, Fm values obtained in this study were lower that those of the Chernobyl experiment and IAEA report.

The two routes of radionuclide contamination of farm animals after the Fukushima Daiichi nuclear power plant accident were inhalation and ingestion of contaminated feed and water. Beresford and Howard (2011) reported that inhalation and water intake by animals were the most important routes in the early phase of the nuclear power plant accident. However, in the Animal Resource Science Center

(140 km south-west from the Fukushima Daiichi nuclear power plant), the concentration of radionuclide contamination in the air was not at a detectable level 2 months after the accident. No radionuclide contamination in drinking water was detected. Intake **via** water is a small contributor. Hence, it is considered that the most important pathway to cows is the ingestion of radionuclide contaminated fodder in the present experiment.

The degree of absorption from the gastrointestinal tract is an important factor in determining radiocesium levels in animal tissues and milk. In the case of radiocesium, the source ingested is a major factor determining subsequent concentrations in tissues with the true absorption coefficient ranging from 0.10 to 0.80 (Howard et al. 2001). They showed that absorption levels of the radiocesium particle and soil binding radiocesium were lower than that of radiocesium incorporated within plants. In the present study, grass was grown for about 2 months after the Fukushima Daiichi nuclear power plant accident, harvested, and prepared for haylage. It has been suggested that the fermented grass forage used in the present study may have contained the radiocesium particle, which was fallout from the accident, soil binding radiocesium, and radiocesium incorporated within plants.

Once the cow has been removed from radiocesium contaminated forage, radiocesium concentrations in the milk rapidly decrease. The rate of loss of radioactive nuclides from milk is termed the biological half-life, which is defined as the time required for the radionuclide activity concentration in milk to be reduced by one half excluding physical decay. The rates of uptake and loss of radionuclides vary between animals and tissues. The biological half-life for radiocesium is associated with the metabolic turnover rate of caesium. Changes in radiocesium levels in milk after the Chernobyl nuclear power plant accident have been summarized by IAEA (2005). The rate of decline in radionuclide activity concentrations in milk of different species of dairy ruminants is rapid. The half-lives of radiocesium in milk are all in the range 0.5–3.5 days, values that are similar to our present result (approximately 2.0 days). The biological half-life of radiocesium in animals is an important factor influencing the effectiveness and practicalities of many countermeasures targeting animal derived foodstuffs, including decontamination using “Clean feeding” or Cs binders.

In the present study, we did not reveal changes in radioactive strontium (^{90}Sr), which accumulates in bones and plays a role as a carcinogenic factor, in milk. Further research on transfer kinetics and the coefficient of ^{90}Sr from forage contaminated by fallout from the Fukushima Daiichi nuclear power plant accident to the milk of cows is required. Moreover, the present findings involved the initial phase of the accident; therefore to remediate livestock management for radiocesium contamination under an “existing exposure situation” according to the ICRP (2009), further research is needed as follows: (1) improvements in pasture: to reduce radiocesium contamination in grassland, feasible and suitable surface improvements in grassland and radical improvements in meadow methods for Japanese farming practices have to be developed, (2) reductions in radiocesium intake: to prevent gut absorption by the application of radiocesium binding agents to animals, feasible and suitable agents have to be explored. The effectiveness and feasibility of candidate binding compounds, clay minerals, which adsorb caesium ions, such as bentonites, vermiculites, zeolites, added to fodder to reduce gut uptake of radiocesium by farm animals have been evaluated. The effectiveness and feasibility of the addition of hexacyanoferrate to feedstuffs have been also evaluated. The hexacyanoferrate compound, Prussian blue, is a radiocesium binder, which is added to the feed of farm animals to reduce radiocesium transfer to milk and other animal products by reducing absorption in the gut (IAEA 1997). Ammonium-hexacyanoferrate (AFCF), the commonly used form for remediation, was used extensively after the Chernobyl accident in Russia, Ukraine, and Belarus as well as in Western European countries, including Norway and Sweden, and was shown to be the most effective for animal use (Pearce 1994; Nisbet et al. 2010). Moreover, the acceptability of these strategies for Japanese farmers also needs to be investigated, and (3) Collateral safety of animal products: to produce milk without disrupting normal farming practices, "Clean feeding" management has been considered feasible and suitable. Farm animals are provided uncontaminated feed or forage with acceptable levels of radiocesium. To prevent radiocesium contamination of animal products by ensuring that feedstuffs that are too highly contaminated are not ingested by farm animals, a registration system to identify individual farm animals has to be developed.

9.5 Conclusion

Radiocesium contamination of animal products is an important potential route of internal exposure in the human food-chain after the Fukushima Daiichi nuclear power plant accident. Most farm animals are kept in closed barns in Japan. Housing farm animals reduced initial radioactive nuclide contamination of animal products in the early phase of the Fukushima Daiichi nuclear power plant accident. Moreover, in Japan, pigs and chickens were given imported feed that contained no radioactive nuclide contamination caused by the accident, a livestock management system called "Clean feeding". However, adequate amounts of grass feed, at least 30%, are essential for rearing cattle, and uncontaminated feed and/or forage containing acceptable amounts of radiocesium are needed. In conclusion, the present findings revealed that when cows received radiocesium contamination of less than 300 Bq/kg, they produced milk contaminated with less than 50 Bq/kg radiocesium.

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Chapter 10
Radiocesium Contamination of Marine Fish Muscle and Its Effective Elimination

Shugo Watabe, Hideki Ushio, and Daisuke Ikeda

Abstract High concentrations of radiocesium (^{134}Cs, ^{136}Cs, and ^{137}Cs combined) were detected in several fish species such as nibe croaker, Pacific cod, and brown hakeling, which were collected from the Pacific Ocean off the coast of the Fukushima and Ibaraki Prefectures. High levels of radiocesium accumulated in fish muscle, but the radioactivity levels of naturally occurring radioactive K in some contaminated fish exceeded the levels of ^{134}Cs and ^{137}Cs. Three washes with 0.1% NaCl solution effectively removed the radiocesium from contaminated fish meat. This can be applied in the production of surimi-based products and other processed seafood such as boiled, dried, or seasoned products.

Keywords Fish • Muscle • Radioisotope • Surimi-based products • Washing

S. Watabe (✉)
Laboratory of Marine Biochemistry, Graduate School of Agricultural and Life Sciences, The University of Tokyo, Bunkyo, Tokyo 113-8657, Japan

Department of Marine Biochemistry, Kitasato University School of Marine Biosciences, Minami, Sagamihara, Kanagawa 252-0373, Japan
e-mail: swatabe@kitasato-u.ac.jp

H. Ushio
Laboratory of Marine Biochemistry, Graduate School of Agricultural and Life Sciences, The University of Tokyo, Bunkyo, Tokyo 113-8657, Japan

D. Ikeda
Department of Marine Biochemistry, Kitasato University School of Marine Biosciences, Minami, Sagamihara, Kanagawa 252-0373, Japan

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10.1 Introduction

The Fukushima nuclear disaster spread radiation across northern Japan in March 2011. Soon after the meltdown at the Fukushima Daiichi nuclear power plants owned by the Tokyo Electric Power Company, the government evacuated people who lived within 30 km of the plants. The building exploded on 15 March, apparently fueled by hydrogen. The fallout of radioactive materials spread to land, rivers, and ocean. In addition, over 500 tons of wastewater containing radioactive material was discharged into the Pacific Ocean. Radioactivity was detected in fish caught in the coastal areas near Fukushima Daiichi. Higher concentrations of radiocesium (^{134}Cs and ^{137}Cs combined) were detected in plaice (*Paralichthys olivaceus*) caught in August (58–590 Bq/kg) compared with in April (approximately 82 Bq/kg) in Fukushima Prefecture (Fishery Research Agency 2012). Thus, the government declared that fisheries products intended for sale should not contain a level exceeding 500 Bq/kg at the market in Japan, which had not been the case since 1940 after World War II. Certain fish were found to contain radiocesium at levels of >500 Bq/kg, such as plaice and sea bass (*Lateolabrax japonicus*). Recently, the government decided to reduce the limit for radiocesium in fish sold in the market to <100 Bq/kg. This limit is extremely safe for consumption, but it has been very difficult for fisheries and related industries to operate in the coastal areas near Fukushima Daiichi because many fish are contaminated with radiocesium above this level. Therefore, it would be desirable to have methods that reduce the concentration of radiocesium in fish so that people engaged in fisheries and related industries can resume their activities and earn a living. Our study aimed to determine the effects of washing fish to remove radiocesium contamination from the muscles that can be then used to manufacture surimi-based products, which are some of the most popular and traditional forms of seafood consumed in Japan.

10.2 Detection of the Tissue Distribution of Radioactive Materials in Fish Using an Imaging Plate System

Pacific cod (*Gadus macrocephalus*), bonefish (*Pterothrissus gissu*), white croaker (*Pennahia argentata*), and nibe croaker (*Nibea albiflora*) were collected at depths of 14–180 m in the Pacific Ocean off Fukushima on August 11th and September 11th, 2011, and subjected to tissue distribution analysis using an imaging plate (IP) system (Watabe et al. 2013). The IP system was used for two-dimensional radioactivity monitoring. Various tissues were dissected from fish samples, which were placed on an IP (BAS-MS2025IP, Fuji film Co. Ltd.) and stored at −30°C for 2 weeks. Subsequently, IP was scanned using an image analyzer (FLA-5000, Fujifilm Co. Ltd., Tokyo, Japan) to detect radioactive tissues. White croaker caught in the East China Sea was used as the negative control in the IP system.

Relatively high signals were detected in the muscle, backbone (vertebral bones with neural and hemal spine bones), skin, and fins of Pacific cod (body weight = 744 g

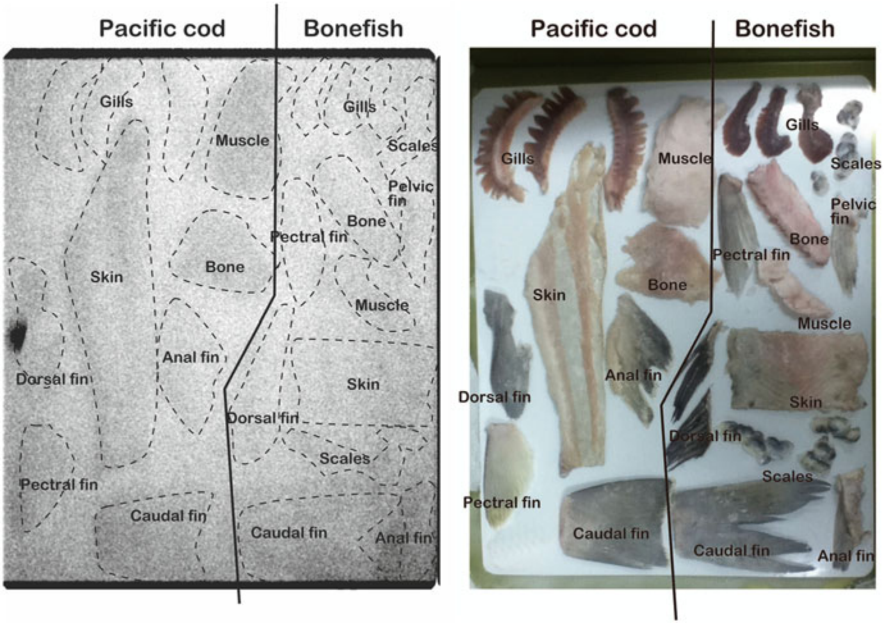

Fig. 10.1 Image plate analysis of various tissues from Pacific cod and bonefish (Watabe et al. 2013)

and total cesium radioactivity = 152 Bq/kg; Fig. 10.1). The signal distribution in the backbone and skin indicated that the positive signals in these areas were attributable to the attached muscle tissues. No obvious signal was detected in bonefish (body weight = 685 g and total Cs radioactivity = 17 Bq/kg). Radioactive substances were also detected in the muscle and fins of nibe croaker (body weight = 368 g and total cesium radioactivity = 515 Bq/kg; Fig. 10.2).

The Daigo Fukuryu Maru (fishing vessel) operating near Bikini Atoll in 1954 was affected by the nuclear fallout from a US hydrogen bomb test. Saiki et al. (1955) analyzed dorado (*Coryphaena hippurus*) caught in the Pacific Ocean and detected high radioactivity levels in the liver, kidney, spleen, and pyloric appendage and relatively high levels in the skin and gills.

It is important to note that the uncontaminated white croaker captured in the East China Sea also produced positive signals in similar tissues, which almost exceeded the signal intensity detected in the contaminated white croaker (total radioactivity of Cs = 55 Bq/kg) captured near Fukushima (Fig. 10.3; Watabe et al. 2013).

A disadvantage of the IP system is that it detects all radiations, which can only be ascribed to specific radionuclides if they are known to be present. Thus, a germanium semiconductor detector (SGD-GEM-5030P4, Ortec, Seiko EG&G Co. Ltd., Tokyo, Japan) was used to detect the radioactivity levels of ^{134}Cs and ^{137}Cs, which showed that the naturally occurring ^{40}K was present in addition to signals produced by ^{134}Cs and ^{137}Cs (Fig. 10.4; Watabe et al. 2013). Similarly high

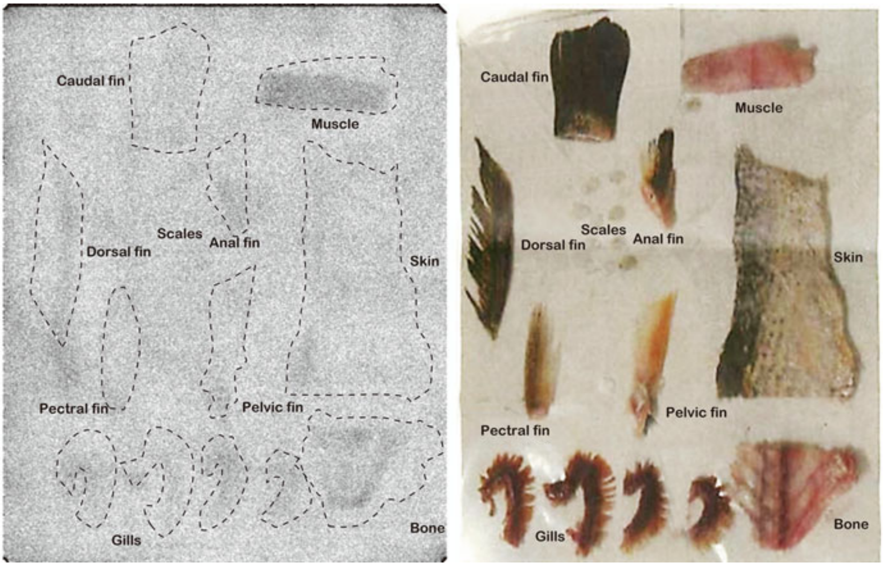

Fig. 10.2 Image plate analysis of various tissues from nibe croaker (Watabe et al. 2013)

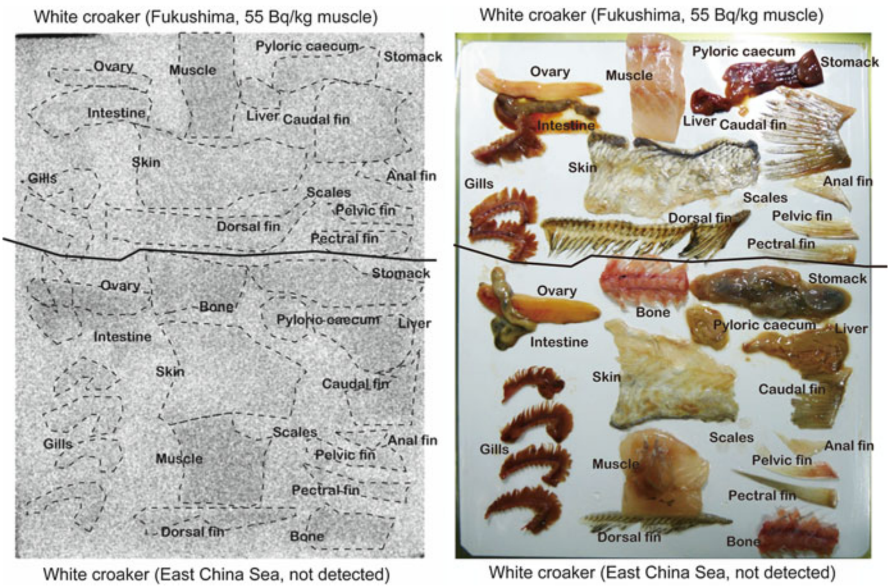

Fig. 10.3 Image plate analysis of various tissues from white croaker contaminated with radioactive materials and those from uncontaminated fish (Watabe et al. 2013)

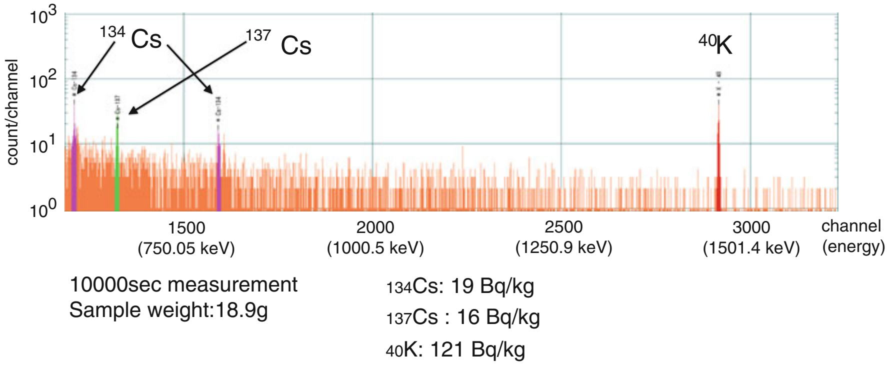

Fig. 10.4 Measurement of radionuclide contamination of Pacific cod muscle (685 g) using a germanium semiconductor detector (Watabe et al. 2013)

concentrations of ^{40}K compared with those of ^{134}Cs and ^{137}Cs have also been reported for Pacific bluefin tuna (*Thunnus orientalis*), where the latter concentrations were trivial (Madigan et al. 2012). The K ion is accumulated at high levels in the intracellular spaces of animals and plants, so radioactive ^{40}K is often present in the organ which is rich in live cells. Thus, the IP system and radiation detectors without spectroscopy may overestimate the radioactivity levels attributable to radiocesium in cell-rich food materials.

10.3 Effective Elimination of Radiocesium Contamination from Fish Meat

We tested the effects of washing fish muscle on removal of radiocesium contamination (Watabe et al. 2013).

In order to determine the most efficient washing method for removing radioactive cesium from fish meat, we determined the effects of different volumes of washing solutions on the residual cesium radioactivity in fish meat. The surimi production process often includes washing steps that use relatively low concentrations of salt solution; therefore, washing procedures using 0.1% NaCl solution were employed in our study. The ordinary muscle of nibe croaker (body weight = 241–368 g and body length = 27.8–30.0 cm) was cut into small pieces and added to various volumes of 0.1% (w/v) NaCl solution (Watabe et al. 2013). The muscle pieces were stirred in the salt solution at 15°C for 5 min. After centrifugation at 2,500×*g* for 10 min, the pellets were subjected to further washing procedures in 0.1% NaCl solution under the same conditions. The residual levels of radioactive cesium (approximately 1/3rd of the original level) reached a plateau after being washed with three volumes of wash solution (Table 10.1), which suggested that three volumes of wash solution is adequate for the removal of cesium from nibe meat. Saiki et al. (1955) also reported that washing the meat of yellowfin tuna (*Thunnus albacores*) in an equal volume of

Table 10.1 Effects of washing volumes on the residual radiocesium level in nibe muscle (cited from Watabe et al. 2013)

Volume	^{134}Cs (Bq/kg)	^{137}Cs (Bq/kg)	Total Cs (Bq/kg)	Residual Cs (%)
Meat	93.7	127	220.7	100
3	38.3	47.2	85.5	38.8
4	29.8	41.6	71.4	32.2
5	37.8	46.7	84.5	38.5
9	25.8	36	61.8	31.6

Table 10.2 Effects of washing times on the residual radiocesium level in Pacific cod muscle (cited from Watabe et al. 2013)

Washing	^{134}Cs (Bq/kg)	^{137}Cs (Bq/kg)	Total Cs (Bq/kg)	Residual Cs (%)
0	–	–	163	100
1	29.5	35.7	65.2	40.3
1	26.3	39.9	66.2	41
1	37.8	46.7	84.5	38.5
1 average				41.5
2	24.2	24.3	48.5	30
2	20.7	23.2[a]	43.9	27.1
2 average				28.6
3	13.9	23.5	37.4	23.1
3	15.1	21.3	36.4	22.4
3	18.8	15.7	24.5	21.4
3 average				22.3

[a]Calculated based on values lower than the detection limit

0.5% ethylenediaminetetraacetic acid (EDTA) solution reduced the residual radioactivity levels to 1/3rd.

As shown in Table 10.2, the residual radioactive cesium level reached a plateau after three washes (Watabe et al. 2013). A similar trend was also observed with Pacific cod using three washing volume (body weight = 473 and 744 g, body length = 35.0 and 40.0 cm; Table 10.2); therefore, the most efficient washing volume may be independent of the fish species.

We also evaluated the effects of the size of meat samples on the washing efficiency. Nibe croaker (body weight = 241–368 g and body length = 27.8–30.0 cm) meat was cut into small pieces and washed with three volumes of 0.1% (w/v) NaCl solution (Watabe et al. 2013). The pieces were stirred in the salt solution at 15°C for 5 min and used as group A samples. Another portion was homogenized using a Waring blender (Ace AM-8, Nissei, Tokyo, Japan) at 8,000 rpm for 5 min and used as group B samples. After centrifugation of both groups at 2,500×*g* for 10 min, the pellets were subjected to further washing procedures in 0.1% NaCl solution under the same conditions and used to monitor the radioactivity levels. The samples from groups A and B that received “n” washes were defined as A-n and B-n, respectively. Group B samples that were homogenized using a Waring blender showed cesium

Table 10.3 Effects of homogenization on the residual radiocesium level in nibe muscle (cited from Watabe et al. 2013)

Sample	^{134}Cs (Bq/kg)	^{137}Cs (Bq/kg)	Total Cs (Bq/kg)	Residual Cs (%)
Meat	151	183	334	100
A-1	32.3	44.3	76.6	23.8
A-2	28.6	40.8	69.4	21.7
A-3	26.1	38.8	64.9	20.3
B-1	38.5	47.3	85.8	27.1
B-2	16.8[a]	24.8	41.6	14.4
B-3	5.8[a]	12.3	18.1	5.9

[a]Calculated based on values lower than the detection limit

elimination with a high efficiency (a final residual radioactivity of approximately 5%; Table 10.3), probably because of the larger surface area of the homogenized meat. This may be an effective approach for eliminating radioactive Cs. However, extensive washing of small pieces of meat may also remove the flavor attributable to amino acids, nucleotides, and other small compounds present in surimi, thereby yielding a tasteless product. Thus, appropriate conditions should be determined for a balance between risk reduction and food quality.

Washing fish muscle contaminated with radioactive cesium may be a very effective approach for the production of surimi-based products. This method may also be applicable to the production of other processed seafood such as boiled, dried, or seasoned products. These trials are currently in progress in our research.

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Chapter 11
Excretion of Cesium Through Potassium Transport Pathway in the Gills of a Marine Teleost

Toyoji Kaneko, Fumiya Furukawa, and Soichi Watanabe

Abstract We used a newly developed technique that insolubilizes and visualizes K^+ excreted from the gills to demonstrate that gill mitochondria-rich (MR) cells are responsible for K^+ excretion in seawater-acclimated Mozambique tilapia. To achieve a better understanding of the molecular mechanism of K^+ excretion from the gills, we identified the cDNA sequences of some candidate molecules that may be involved in K^+ transport. Of these candidate molecules, only a renal outer medullary K^+ channel (ROMK) exhibited markedly upregulated mRNA levels in response to a high external K^+ concentration. We found that ROMK expressed in the apical opening of MR cells was the main molecular pathway responsible for K^+ excretion. Using the insolubilization technique, we further demonstrated the excretion of Cs^+ and Rb^+, which are known to be biochemical analogs of K^+, through the K^+ transport pathway in the gills of seawater tilapia. The activation of ROMK in the gill MR cells induced by K^+ supplementation could eliminate unnecessary biochemical analogs of K^+ more rapidly, possibly serving as an effective countermeasure against radiocesium in aquaculture.

Keywords Cesium • Mitochondria-rich cell • Mozambique tilapia • Potassium • Renal outer medullary K^+ channel • Rubidium

Abbreviations

Cs Cesium
Rb Rubidium

T. Kaneko (✉) • F. Furukawa • S. Watanabe
Graduate School of Agricultural and Life Sciences, The University of Tokyo,
1-1-1 Yayoi, Bunkyo, Tokyo 113-8657, Japan
e-mail: kaneko31@marine.fs.a.u-tokyo.ac.jp

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11.1 Introduction

The recent nuclear accident at the Fukushima Daiichi nuclear power plant in Japan released massive amounts of radioactive materials into the environment including radiocesium (^{134}Cs and ^{137}Cs). Cesium belongs to the alkali metal group and shares chemical properties with K and Rb; therefore, it is regarded as a biochemical analog of K (Relman 1956). Radiocesium is considered to behave like K in living organisms after being absorbed into the body because of its similar biochemical properties. Similar to terrestrial animals, aquatic animals are at a high risk of radioactive contamination because radioactive pollutants enter rivers and the ocean. In particular, marine teleosts are vulnerable to the absorption of higher levels of minerals dissolved in seawater because they drink a large amount of ambient seawater to compensate for their osmotic water loss (Evans et al. 2005; Marshall and Grosell 2006). Assuming that Cs behaves exactly like K, it is critical to clarify the excretory pathway of K in marine teleosts. Although the precise molecular mechanisms for the regulation of plasma Na^+ and Cl^-, which are the major ions in the plasma, have been elucidated in several stenohaline and euryhaline teleosts (Marshall and Grosell 2006), less information is available on the regulatory mechanisms of K^+.

Our recent findings indicate that excess K^+ is excreted from the gills in marine teleosts (Furukawa et al. 2012a), as is the case with Na^+ and Cl^- secretion. The gills are also responsible for eliminating unnecessary Cs^+ and Rb^+, which are biochemical analogs of K^+, from the body fluid, presumably through the K^+ excretion pathway in the gills (Furukawa et al. 2012b). In this chapter, we review recent advances in the understanding of K^+ regulation in teleost fish and provide basic information related to possible countermeasures for radiocesium in aquaculture.

11.2 Osmoregulation in Teleost Fishes

11.2.1 Body Fluid Osmolality

Body fluid osmolality is an essential prerequisite for vertebrate species, including teleosts, in order to maintain various chemical and physical parameters of the body fluids within certain physiological ranges, which ensures the normal activities of the cells that constitute the body. One of the most important factors that define the characteristics of body fluids is the osmotic pressure, or osmolality, which is determined by the total concentration of solutes (osmolytes) in the body fluid, mostly inorganic electrolytes. Because Na^+ and Cl^- are the major electrolytes in the blood plasma, the regulation of both Na^+ and Cl^- is critical for osmoregulation. Another important factor is regulation of the water balance because water acts as a solvent for osmolytes. Thus, vertebrates regulate their body fluid osmolality by controlling their ion concentration and water content.

11.2.2 Passive Ion and Water Movements in Teleosts

Teleost species that inhabit aquatic environments experience different osmotic challenges compared with terrestrial vertebrates. Aquatic animals such as teleosts possess gills, which are permeable to ions and water to some extent, as their respiratory organs. Ion and water movements depend on concentration and osmotic gradients, respectively, between the body fluid and the external aquatic environment. This is not the case with terrestrial vertebrates, where gas exchange occurs through the air in the lungs. Teleost fish have developed excellent osmoregulatory mechanisms that maintain their ion and water balance in a wide environmental salinity range.

11.2.3 Freshwater and Seawater Adaptation

As with most other vertebrate species, teleosts maintain the osmolality of their body fluids at a constant level of approximately 1/3rd of seawater osmolality. Osmoregulation in adult teleosts is mainly the result of integrated ion and water transport activities in the gills, kidneys, and intestine (Evans et al. 2005; Marshall and Grosell 2006). Freshwater teleosts experience water load and salt loss through their permeable body surfaces, most of which occur through their gill epithelia. To overcome these problems, they discharge excess water by producing dilute urine in their kidneys and they absorb ions through their gill epithelia. In contrast, marine teleosts must cope with water loss and their salt load. The water loss is compensated by drinking seawater and absorbing water in the intestine, whereas the excess ions are actively excreted from the gills and kidneys. Therefore, in adult fish, the gills, kidneys, and intestine are important osmoregulatory organs that create ionic and osmotic gradients between the body fluids and the external environment. Of the ion-transporting epithelial cells found in these osmoregulatory organs, the mitochondria-rich (MR) cells, or chloride cells, found in the gill epithelia are the main sites of ion secretion and absorption, which are important for seawater and freshwater adaptation, respectively (Kaneko et al. 2008). MR cells are also involved in acid–base regulation and Ca^{2+} uptake and thus play crucial roles in the adaptation to various osmotic and ionic aquatic environments.

11.2.4 MR Cells

MR cells are characterized by the presence of numerous mitochondria in their cytoplasm, which allows detection of MR cells using mitochondria-specific fluorescent probes, such as DASPEI, Rhodamine 123, and MitoTrackers, with a fluorescence or confocal laser scanning microscope (Kaneko et al. 2008). The cells are also characterized by an extensive tubular system in the cytoplasm. This tubular system is continuous with the basolateral membrane, which presents a large surface area for the localization of ion

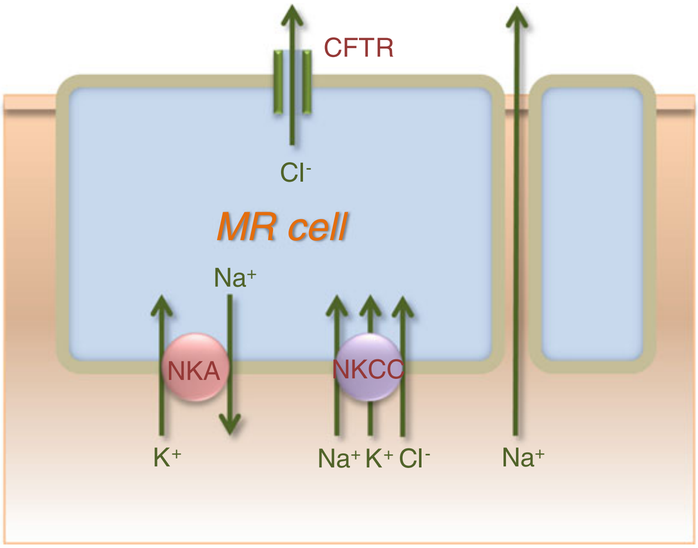

Fig. 11.1 Molecular mechanisms of Na^+ and Cl^- secretion from mitochondria-rich (MR) cells in seawater fish, involving the cooperative actions of three major ion transporters: Na^+/K^+-ATPase (NKA), $Na^+/K^+/2Cl^-$ cotransporter (NKCC), and cystic fibrosis transmembrane conductance regulator (CFTR) Cl^- channel

transporting proteins. Of these, Na^+/K^+-ATPase is a key enzyme with ion transporting functions in MR cells, which creates ionic and electronic gradients and provides the major driving force for ion secretion and absorption in MR cells (McCormick 1995).

11.2.5 Molecular Mechanisms Underlying the Ion Transporting Functions of MR Cells

The current accepted model of Na^+ and Cl^- secretion from MR cells in seawater fish (Fig. 11.1) involves the cooperative action of three major ion transporters: Na^+/K^+-ATPase, $Na^+/K^+/2Cl^-$ cotransporter (NKCC), and cystic fibrosis transmembrane conductance regulator (CFTR) Cl^- channel (Hirose et al. 2003; Evans et al. 2005). The basolaterally located Na^+/K^+-ATPase causes low intracellular Na^+ concentration and a high negative charge within the cell. This Na^+ gradient is used to transport Na^+, K^+, and $2Cl^-$ into the cell through the basolateral NKCC. Next, Cl^- exits the cells down on an electrical gradient through an apical Cl^- channel, whereas Na^+ is transported back outside the cells by Na^+/K^+-ATPase before being secreted through the paracellular pathway formed between MR and adjacent accessory cells.

However, the mechanisms underlying ion uptake in MR cells of freshwater fish are still unclear and controversial, although several different models have been proposed for ion absorption by MR cells (Hirose et al. 2003). The difficulty lies in the fact that different species may possess different mechanisms and that each species may have more than one type of ion-absorbing MR cell. For example in Mozambique tilapia (*Oreochromis mossambicus*), two distinct types of MR cells have been observed in freshwater-acclimated fish, i.e., MR cells with apically expressed Na^+/H^+ exchanger 3 and those with apically expressed Na^+/Cl^- cotransporter (Inokuchi et al. 2008).

11.2.6 Possible K^+ Secretion from the Gills

While the molecular mechanisms of Na^+, Cl^-, and acid–base regulation and those of nitrogen metabolite excretion in fish MR cells have become increasingly clear (Hwang et al. 2011), the regulatory mechanisms of K^+ in fish is yet to be identified. In general, K^+ is a major monovalent cation in vertebrate animals (Their 1986). Due to the action of Na^+/K^+-ATPase on the cell membrane, a large proportion of K^+ is confined within cells, whereas the plasma K^+ concentration is maintained at a relatively low level. Terrestrial animals regulate their plasma K^+ levels within physiological ranges by modulating renal K^+ excretion in the urine. In the mammalian kidney, the renal outer medullary potassium channel (ROMK or Kir 1.1), located in the apical membrane of tubular epithelial cells from the thick ascending limb through the outer medullary collecting duct, is the main route for K^+ secretion to the lumen (Hebert et al. 2005). Other proteins involved in K^+ handling are also expressed in the mammalian kidney, such as the K large conductance Ca^{2+}-activated channel, subfamily M (Maxi-K or BK), and K^+–Cl^- cotransporters (KCC1, KCC3, and KCC4) (Wang and Giebisch 2009).

Seawater contains more than twice as much K^+ as the typical plasma in vertebrate animals. However, seawater-inhabiting fish produce a small amount of K^+-poor isosmotic urine, which is insufficient to maintain an appropriate internal K^+ level. Thus, seawater fish are considered to have an extra-renal mechanism that eliminates any excess K^+ from their body fluids. In this context, we recently demonstrated the excretion of K^+ from the gills and elucidated its molecular mechanism in seawater-acclimated Mozambique tilapia (Furukawa et al. 2012a).

11.3 K^+ Secretion from MR Cells

11.3.1 Visualization of K^+ Secretion from Gill MR Cells

Using a newly developed technique that insolubilized and visualized K^+ excreted from the gills, we demonstrated that gill MR cells were responsible for K^+ excretion in seawater-acclimated Mozambique tilapia (Furukawa et al. 2012a). Mozambique tilapia is one of the most suitable fish species for studies on osmoregulation because this euryhaline tilapia is adaptable not only to a wide range of salinity from freshwater to seawater but also to extremely low-ion water and even concentrated seawater.

To determine the site of K^+ excretion, K^+ was reacted with sodium tetraphenylborate (TPB) to form insoluble precipitates of K-TPB. The gills of seawater-acclimated tilapia were dissected out and washed using 0.025% $KMnO_4$ in PBS for 1 min to remove mucus from the filament surface. After washing with mannitol solution (320 mM mannitol, 1.8 mM $CaCl_2$), the gill filaments were placed in a reaction solution containing 30 mM Na-TPB, 265 mM mannitol, 1.8 mM $CaCl_2$, and 50 mM HEPES (TPB solution). After 30 s, the TPB solution was removed and the filaments were washed using mannitol solution. To label the MR cells, the gill filaments were immersed for 15 min in a solution containing 0.05 mM rhodamine 123, 2 mM HEPES, 2.7 mM KCl, 0.4 mM $MgCl_2$, 158 mM NaCl, and 1.8 mM $CaCl_2$. The samples were then observed using a confocal laser scanning microscope. The TPB-treated gill filaments were also analyzed by energy dispersive X-ray spectrometry (EDS) to confirm the presence of K in the precipitates observed in the apical openings of MR cells. After fixation in 4% paraformaldehyde in 0.1 M phosphate buffer (pH 7.4) for 24 h, the gill filaments were washed with distilled water and snap-frozen in liquid N_2. The frozen filaments were dried in a freeze-dryer. The dried filaments were mounted on specimen stubs, coated with Au in an ion coater, and analyzed using a scanning electron microscope (SEM) equipped with an EDS system.

When the gills were observed by differential interference contrast (DIC) microscopy, precipitates of possible K-TPB were visible as particles on the gills (Fig. 11.2a). The particles were localized in the apical openings of MR cells, which were confirmed by merging the DIC image with the corresponding fluorescent image of MR cells (Fig. 11.2b). In the SEM observations, the precipitates were visible as amorphous structures attached to the surface of the gill filaments (Fig. 11.2c). The EDS analysis showed that the K signals were closely associated with the precipitates (Fig. 11.2d). These findings clearly indicated that K^+ was secreted from gill MR cells in seawater-acclimated tilapia.

11.3.2 Molecular Mechanism of K^+ Secretion

To achieve a better understanding of the molecular mechanism of K^+ excretion from the gills, we identified the cDNA sequences of the candidate molecules that could be involved in K^+ handling in the Mozambique tilapia gills, such as the renal outer medullary K^+ channel (ROMK), K large conductance Ca^{2+}-activated channel, subfamily M (Maxi-K), and K^+–Cl^- cotransporters (KCC1, KCC2, and KCC4). The tissue distribution patterns of ROMK, Maxi-K, KCC1, KCC2, and KCC4 mRNA expression were analyzed by RT-PCR in freshwater- and seawater-acclimated tilapia. All of them were confirmed to be expressed in the gills, with the exception of KCC2, which was exclusively expressed in the brain.

To further determine the primary K^+ channel or transporter involved in K^+ secretion in the branchial MR cells, we compared the expression levels of the candidate

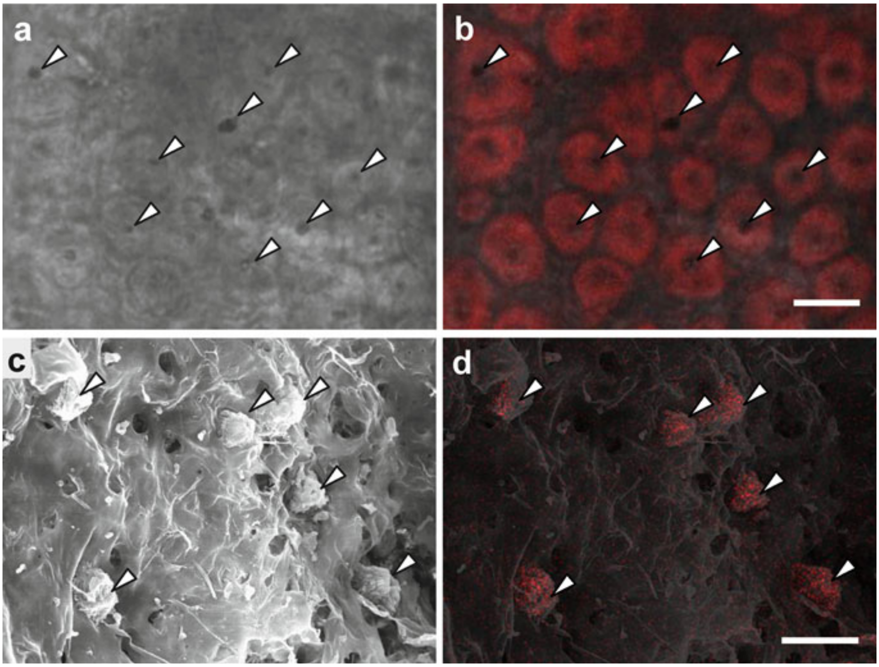

Fig. 11.2 Detection of K^+ excreted by the gills of seawater-acclimated Mozambique tilapia. A transmitted differential interference contrast (DIC) image of the precipitates (*arrowheads*) formed on the gill surface (**a**) and a DIC image merged with a panfocal fluorescent micrograph showing mitochondria-rich cells (*red*) in the same field (**b**). A scanning electron micrograph of the precipitate (*arrowheads*) on the gill surface (**c**), and the same image merged with a mapping image of potassium obtained using energy dispersive X-ray spectrometry (**d**). Bars, 20 μm (modified from Furukawa et al. 2012a)

molecules in the gills of Mozambique tilapia acclimated to environments with different K^+ concentrations. The tilapia were acclimated to freshwater (K^+, <0.1 mM), seawater (8 mM), and artificial seawater containing moderate (8 mM) and high (17 mM) concentrations of K^+ for 1 week. Of the candidate molecules, ROMK exhibited marked upregulation of mRNA levels in response to a high external K^+ concentration (Fig. 11.3a), whereas the expression levels of Maxi-K, KCC1, and KCC4 did not change significantly in response to the environmental K^+. These findings suggest that ROMK is the key molecule involved in K^+ secretion from MR cells.

In addition to ROMK, NKCC1a tended to be highly expressed in high K^+ environments (Fig. 11.3b). NKCC1a was expressed in the basolateral membrane of seawater-type ion-secretory MR cells (Fig. 11.1). Although K^+ enters MR cells primarily through the basolateral Na^+/K^+-ATPase, the upregulation of NKCC1a in tilapia acclimated to high K^+ conditions may contribute to additional K^+ entry into MR cells.

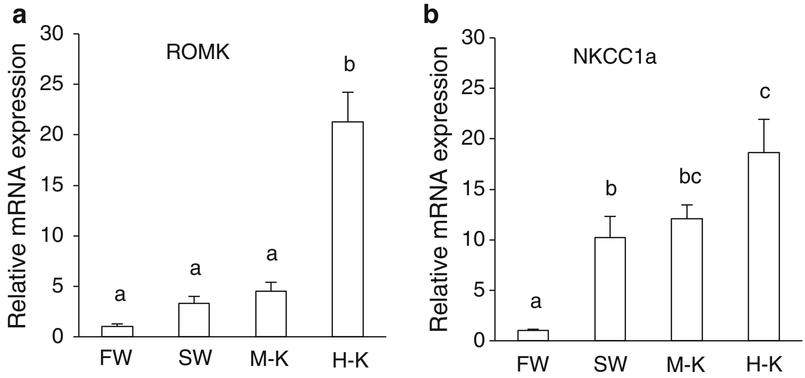

Fig. 11.3 Expression levels of ROMK (**a**) and NKCC1a (**b**) mRNAs in the gills of tilapia acclimated to freshwater (FW, $[K^+] < 0.1$ mM), seawater (SW, 8 mM), and artificial SW with moderate (M-K, 8 mM) and high (H-K, 17 mM) concentrations of K^+ for 1 week. Data are expressed as mean ± SE ($n = 6$). Different letters indicate significant differences at $p < 0.05$ (modified from Furukawa et al. 2012a)

11.3.3 Confirmation of K^+ Secretion Through ROMK

The K^+ secretory function of ROMK was further confirmed by exposing the gills to TPB solution containing Ba^{2+}, a non-selective blocker of ROMK and Maxi-K, or iberiotoxin (IBTX), a specific blocker of Maxi-K. In the control, K-TPB precipitates were evident on the surfaces of the gills. Treatment with 5 mM Ba^{2+} inhibited the formation of K-TPB precipitates, whereas 200 nM IBTX did not. These results support the conclusion that K^+ excretion occurs mainly through ROMK.

To localize ROMK protein in the gill MR cells, the gill filaments of Mozambique tilapia acclimated to environments containing different K^+ concentrations were double-stained using an antiserum raised against tilapia ROMK and anti-Na^+/K^+-ATPase for detecting MR cells. Immunofluorescence microscopy showed that ROMK was localized in the apical openings of Na^+/K^+-ATPase-immunoreactive MR cells (Fig. 11.4) and that the ROMK immunoreaction was most intense in fish acclimated to environments with a high K^+ concentration (Fig. 11.4d). In contrast, the immunoreaction was weak in fish acclimated to freshwater (Fig. 11.4a).

Ordinary cells contain approximately 140 mM K^+ in their cytoplasm, but MR cells are likely to be richer in K^+ due to the active basolateral uptake of K^+ through Na^+/K^+-ATPase and NKCC1a. Thus, the concentration gradient across the apical membrane of MR cells favors K^+ excretion through ROMK to the external environment. The strong ROMK immunoreaction of MR cells in high K^+-acclimated fish indicates a higher density of ROMK in the apical openings of MR cells, suggesting the elimination of excess K^+ at a higher rate.

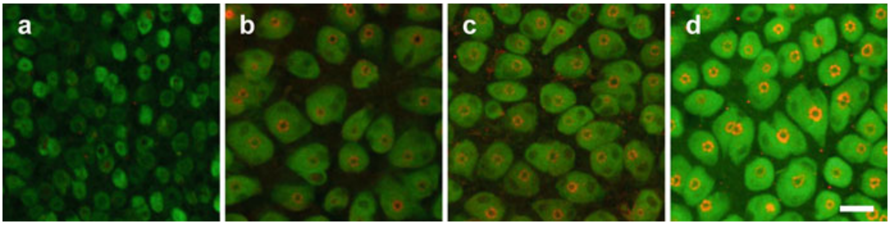

Fig. 11.4 Immunofluorescence micrographs of gills stained with anti-Na^+–K^--ATPase (NKA, *green*) and anti-ROMK (*red*) in tilapia acclimated to freshwater (**a**), seawater (**b**), and artificial seawater with moderate (**c**) and high concentrations (**d**) of K^+. Bar, 20 μm (modified from Furukawa et al. 2012a)

11.4 Cs^+ and Rb^+ Secretion from MR Cells

11.4.1 Cs^+ and Rb^+ as Biochemical Analogs of K^+

Small amounts of Cs^+ and Rb^+, which are generally considered to be biochemical analogs of K^+, are detectable in fishes and seawater, although their concentrations are much lower than that of K^+. The distributions of Cs^+ and Rb^+ have also been studied in fish, where these ions are mainly located in the muscle (Peters et al. 1999). Although the elimination route of Cs^+ and Rb^+ is still unclear, the K^+ excretion pathway in the branchial epithelium is a strong candidate for the elimination of these ions.

In spite of little physiological impact of non-radioactive Cs^+ in fish, the radiocesium (^{137}Cs and ^{134}Cs) fallout generated by the recent nuclear disaster at the Fukushima Daiichi nuclear power plant in Japan caused extensive damage to the surrounding environment and living organisms. Marine teleosts were also contaminated by radiocesium. Thus, it is very important to know whether marine teleosts can secrete Cs^+ through the K^+ transport pathway in the gill MR cells.

11.4.2 Detection of Cs^+ and Rb^+ Excreted from the Gills

To detect Cs^+ and Rb^+ possibly excreted from the gills, we used the K^+ insolubilization technique with some modifications because Na-TPB reacts not only with K^+ but also with Cs^+ and Rb^+ to form insoluble precipitates. A gill arch in seawater-acclimated tilapia was cut at the ventral end and infused intra-arterially for >1 min with 1 ml of balanced salts solution (BSS; 140 mM Na^+, 146 mM Cl^-, 2.95 mM K^+, 1.5 mM Ca^{2+}, 1.29 mM Mg^{2+}, 5 mM HEPES, pH 7.5), where K^+ was replaced with the same amount of Cs^+ (Cs-BSS) or Rb^+ (Rb-BSS). After replacing the blood with Cs-BSS or Rb-BSS, the gills were dissected out and incubated in BSS for 10 min to allow the infused Cs^+ or Rb^+ to spread. The gills were then treated with TPB solution, in the same manner as described previously, and subjected to SEM observation

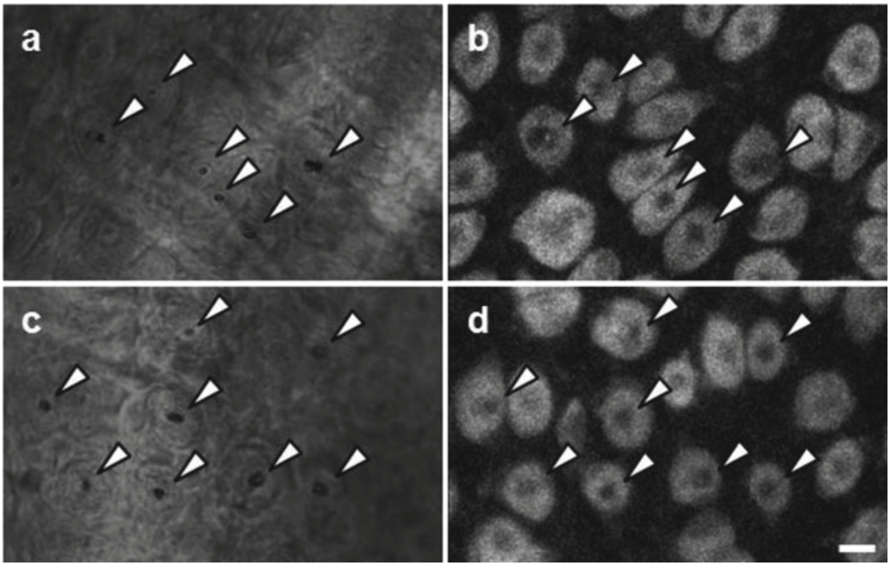

Fig. 11.5 Transmitted differential interference contrast images of the precipitates (*arrowheads*) on gills infused with Rb^+ (**a**) and Cs^+ (**c**), and those merged with panfocal fluorescent micrographs showing mitochondria-rich (MR) cells (**b**, **d**). The precipitates are shown on MR cells stained with rhodamine 123 (**b**, **d**) on the gill epithelia. Bar, 10 μm (from Furukawa et al. 2012b)

and EDS analysis. To simultaneously detect ion excretion and MR cells, some gills were infused with Cs-BSS or Rb-BSS containing 50 μM rhodamine 123 and examined using a confocal laser scanning microscope.

After treatment with TPB solution, the precipitates were observed as black particles on the openings of MR cells (Fig. 11.5), in a similar manner to the previous observations (Fig. 11.2a, b). The gill surface areas with and without precipitates were subjected to EDS analysis to obtain their X-ray energy spectra (Fig. 11.6). X-ray energy peaks characteristic of Cs were detected in the areas of Cs-BSS-infused gills with precipitates (Fig. 11.6e, f). Similarly, an X-ray energy peak that was characteristic of Rb was detected in the areas of Rb-BSS-infused gills with precipitates (Fig. 11.6a, b). These precipitates also showed a sharp X-ray energy peak characteristic of K. In contrast, there were no clear X-ray energy peaks for K, Cs, or Rb in the areas devoid of the precipitates, irrespective of whether the samples were infused with Cs-BSS or Rb-BSS (Fig. 11.6c, d, g, h).

The distribution patterns of K, Cs, and Rb on the lines across the precipitates were also examined by EDS line-scanning analysis (Fig. 11.7). In the Rb-BSS-infused gills, there was a clear correspondence between the distribution patterns of K and Rb, and both elements were specifically detected in the precipitate (Fig. 11.7a, c, e). Similarly, the characteristic X-ray spectra of K and Cs were specifically detected in the precipitate in Cs-BSS-infused gills (Fig. 11.7b, d, f). Overall, these results suggest that Rb^+ and Cs^+ were excreted through the K^+ transport pathway in the gill MR cells.

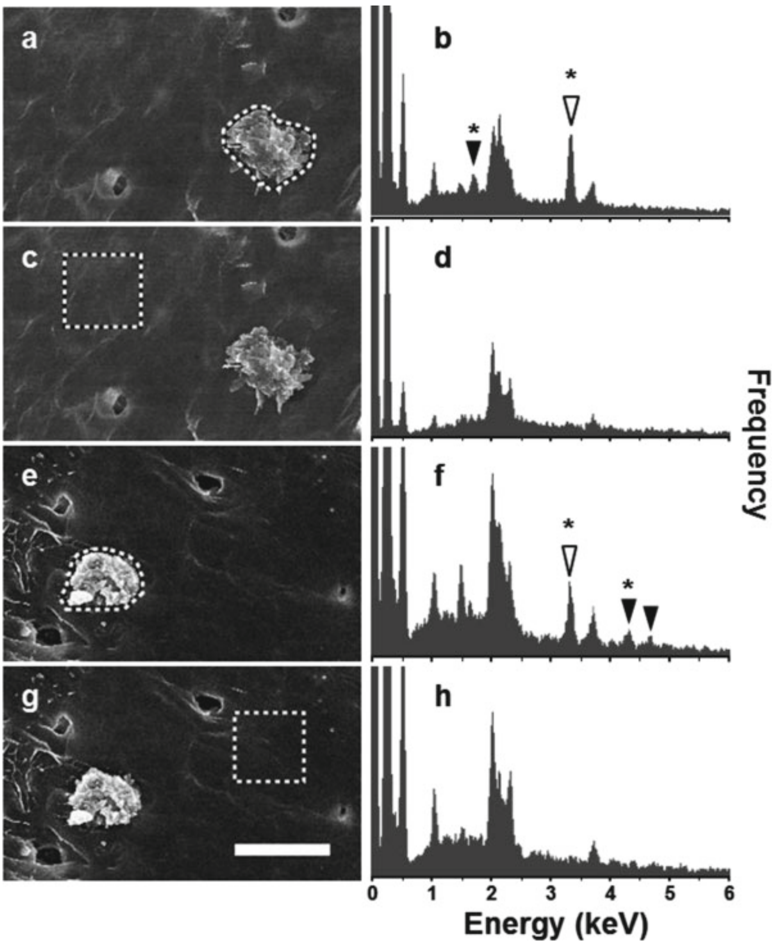

Fig. 11.6 Scanning electron micrographs (**a**, **c**, **e**, **g**) and energy dispersive X-ray spectrometry (EDS) profiles (**b**, **d**, **f**, **h**) of gills infused with Rb^+ (**a–d**) and Cs^+ (**e–h**). The *amorphous* and *square* lines (**a**, **c**, **e**, **g**) indicate the areas subjected to EDS analysis (**b**, **d**, **f**, **h**). *White arrowheads* indicate a representative X-ray energy peak of K. The *black arrowhead* in panel **b** indicates an energy peak of Rb, whereas the *black arrowheads* in panel **f** indicate those of Cs. Bar, 10 μm (from Furukawa et al. 2012b)

11.5 Perspectives

Our findings clearly elucidated the branchial Cs^+ and Rb^+ eliminating pathways through the ROMK-based K^+ excretion mechanism of MR cells in seawater-acclimated Mozambique tilapia (Fig. 11.8). The radioactive Cs fallout generated by the recent nuclear disaster at the Fukushima Daiichi nuclear power plant in Japan is an extremely serious environmental issue that needs to be addressed urgently. The present study provides important information about the possible elimination

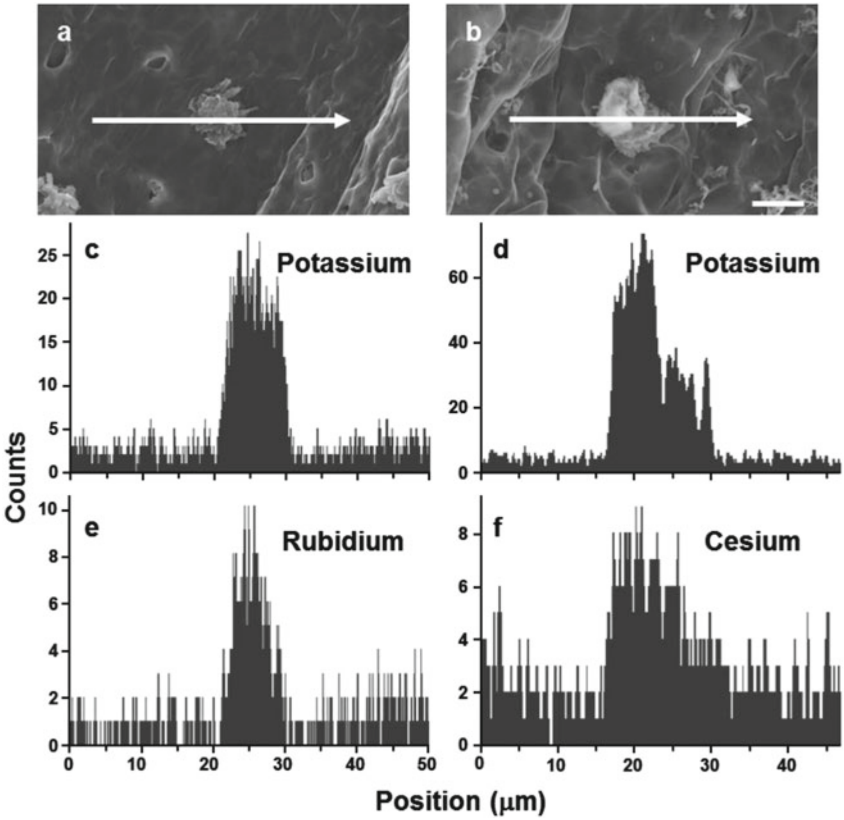

Fig. 11.7 Line-scanning element analyses of gills infused with Rb^+ (**a**, **c**, **e**) and Cs^+ (**b**, **d**, **f**). Analyses were performed on the arrows crossing the precipitates in panels (**a**) and (**b**) at 3.31 keV for K (**c**, **d**), 1.69 keV for Rb (**e**), and 4.29 keV for Cs (**f**). Bar, 10 μm (from Furukawa et al. 2012b)

route of Cs in marine teleosts, which clearly enhances our understanding of the metabolism of radiocesium in teleosts. The excretion of biochemical analogs of K^+ probably accounts for the shorter biological half-life of ^{137}Cs in marine teleosts than freshwater teleosts (Kasamatsu 1999). In euryhaline Mozambique tilapia, ROMK is expressed in the gills in seawater and freshwater, although its expression level is lower in freshwater than in seawater (Furukawa et al. 2012a), suggesting that freshwater teleosts may also possess the ability to excrete Cs^+ through their MR cells. The activation of ROMK in MR cells induced by K^+ supplementation could eliminate unnecessary biochemical analogs of K^+ more rapidly, possibly acting as an effective countermeasure for radiocesium in aquaculture.

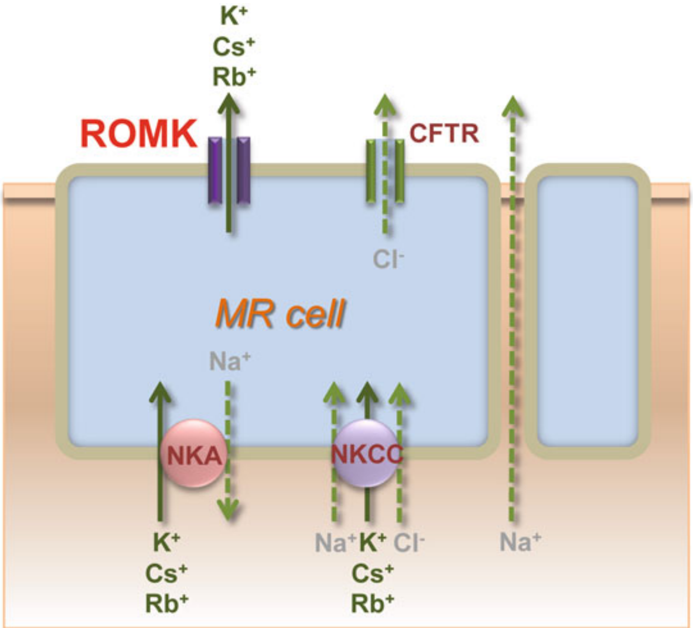

Fig. 11.8 Molecular mechanisms of K^+, Cs^+, and Rb^+ secretion from mitochondria-rich (MR) cells in seawater fish, involving the cooperative actions of three major ion transporters: Na^+/K^+-ATPase (NKA), $Na^+/K^+/2Cl^-$ cotransporter (NKCC), and renal outer medullary K^+ channel (ROMK)

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Chapter 12
Contamination of Wild Animals: Effects on Wildlife in High Radioactivity Areas of the Agricultural and Forest Landscape

Ken Ishida

Abstract This study presents new findings related to radioactivity measurements in birds and other wild animals. High levels of radioactive contamination were found in the feathers of birds after the accident and most of this radioactivity was not washed out. However, there was little contamination in the new feathers produced by the same species after approximately 1 year. We discuss the possible effects of radioactivity, particularly cascade effects among wildlife in the Satoyama ecosystem. Satoyama consists of well-managed agricultural fields, grasslands, and several types of forests at a small-scale topography level. We describe our long-term monitoring approach.

Keywords Bush warbler • Cascade effect • Long term monitoring • Radioactive effect • Wildlife

12.1 Introduction

The highly radioactive plume that originated from Fukushima Daiichi nuclear power reactors 1–3 in mid-March 2011 moved toward the northwest and fell with the rain, contaminating an area of the northern Abukuma highlands in Fukushima Prefecture (Fig. 12.1). The dose rates at 1 cm above the ground were 20 μSv/h, with more in some hotspots >30 km from the Fukushima Daiichi nuclear power plant (F1-NPP). The season this occurred was before the shooting of plants in the spring or the breeding of most birds and animals. The areas affected contained paddy fields, farmlands, and natural and artificial forests in the highland areas, where the

K. Ishida (✉)
Graduate School of Agricultural and Life Sciences, The University of Tokyo,
1-1-1 Yayoi, Bunkyo-ku, Tokyo 113-8657, Japan
e-mail: ishiken@es.a.u-tokyo.ac.jp

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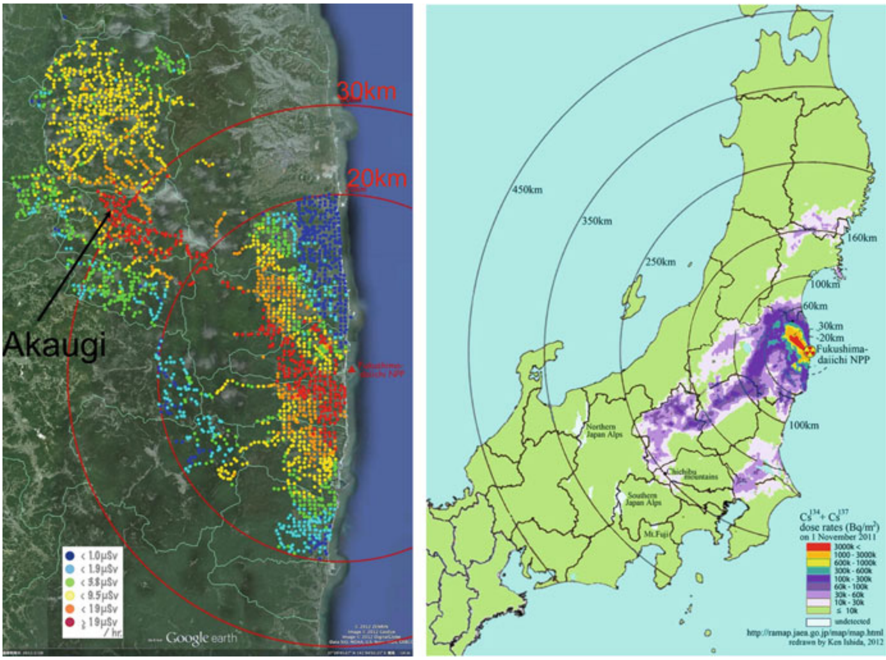

Fig. 12.1 *Left*, the dose rate at 1 cm above the ground surface in a high radioactive contamination area around and northwest of F1-NPP. The dose rates (μSv/h) were measured by the survey consortium (TEPCO team) during July 4th and August 20th, 2011. *Right*, estimated total ^{134}Cs and ^{137}Cs (Bq/m^2) accumulation on the ground surface, based on the airborne monitoring survey conducted by MEXT from August to October 2011 (Ministry of the Education, Culture Sports, Science and Technology—Japan, November 11, 2011). All the estimated values are fitted to the assumed ones, which were at November 1st, 2011. Redrawn by the author

biodiversity is high. These areas contain more than 15 terrestrial mammals, including the macaque monkey, approximately 150 bird species, many insects, other animals, plants, and mushrooms. All residents were evacuated from this area. Thus, the wildlife was left with little or no human interferences, such as through agriculture, hunting, fishing, harvest, or automobiles.

In this study, I report the situation for birds and some mammals that inhabited the area 1.5 years after the accident. We need to understand that the condition is dynamic and the situation is changing constantly.

12.2 Landscape of the Abukuma Highlands and Its Biodiversity

The Abukuma Mountains cover an area of approximately 170 km from south to north and 40 km from east to west, and they are surrounded by a 10-km wide narrow plain on the Pacific coast to the east (Hamadori area), the Abukuma River plain to

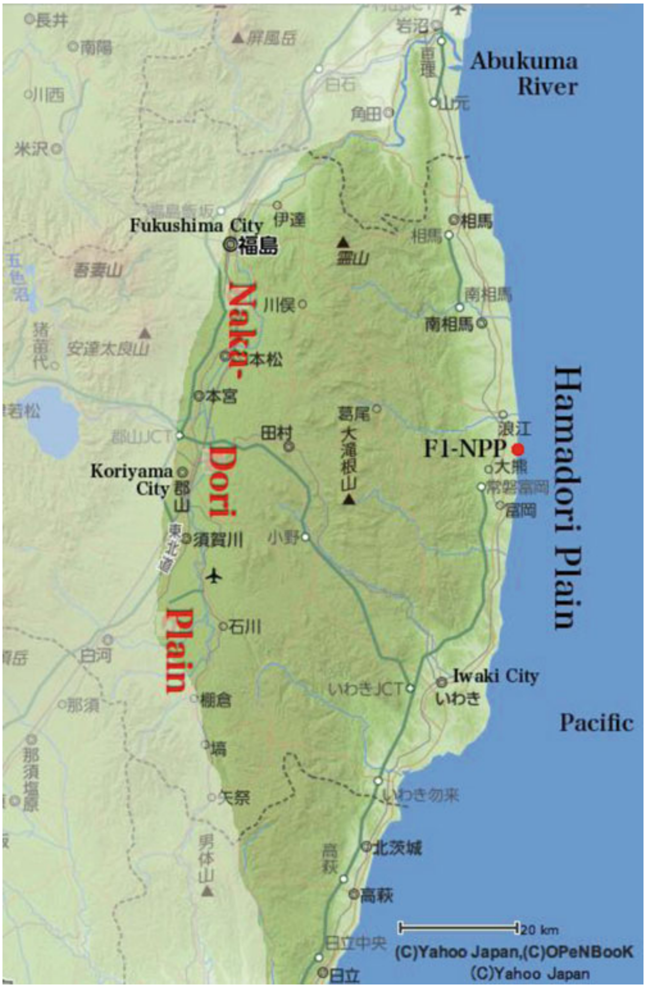

Fig. 12.2 Map of the Abukuma Mountains, the surrounding plains, and rivers. F1-NPP is located on the Pacific coast. A mountainous area covered with natural and artificial forests, paddy fields, and farmland is close to F1-NPP, which contains an abundance of wildlife

the west (Nakadori area) and the north, and the Kuji River valley to the south (Fig. 12.2). F1-NPP is located in the central east region of the Pacific coast on the Hamadori plain. Immediately behind the Hamadori plain, there are small valleys with steep streams that reach up to an altitude of approximately 400 m. There are few houses and little human activity on these slopes. The Abukuma highlands are gentle hills at an altitude of approximately 600 m that reach up to 1,192 m and they contain a mixture of paddy fields, farmlands, secondary deciduous forest, and evergreen conifer plantations. The dominant natural tree species are *Fagus japonica*, *Carpinus tschonoskii*, *Zelkova serrata*, *Aesculus turbinata*, *Pterocarya rhoifolia*, *Acer mono*, and *Abies firma*. The main plantation species are *Cryptomeria japonica*

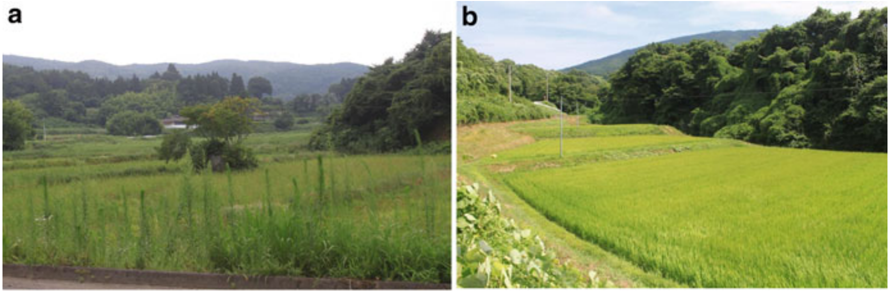

Fig. 12.3 Abandoned (*left*, Kawamata village) and cultivated (*right*, Nihonmatsu town) paddy fields in the Abukuma highlands. They were also cultivated on the slopes (*right*). The images were captured in August 2012. These locations were close to each other and approximately 30 km west of F1-NPP

and *Chamaecyparis obtusa*. Many small streams and ponds (Fig. 12.3, *left*) are scattered among the agricultural fields and forests. Thus, the ecosystem structure is heterogeneous and there are plenty of niches for wildlife. This highly human-affected natural landscape is known as Satoyama in Japanese.[1]

The western Abukuma Mountains have gentle slopes and there are human residences surrounded by a Satoyama-like landscape with many small sections of paddy fields. There are streams in abundance and the small hills are covered with natural forest. Most of the western area escaped high radiation exposure; therefore, human activity has continued in this area (Fig. 12.3, *right*).

12.3 The Bird Community in the Northern Abukuma Highlands

From 1972 to 2003, Tozawa (2005) recorded 195 bird species in the northeast corner of Abukuma mountains and the connecting plain area (old Haramachi city). In a 20 km by 20 km area of the northern Abukuma highland, I recorded 52 species during 56 separate 5-min counts in May 2012 and 42 species in 38 separate 5-min counts in June 2012. The dominant species were *Cettia diphone*, *Hypsipetes amourotis*, *Emberiza cioides*, *Cuculus policephalus*, *Corvus macrorhynchos*, *Parus major*, *Passer montanus*, *Phasianus versicolor*, *Streptopelia orientalis*, and *Carduelis sinica* (Table 12.1).

The distribution of the highly dominant bush warbler (*C. diphone*) and brown-eared bulbul (*H. amourotis*) was not related (negatively or positively) to the radiation levels (Fig. 12.4). Their distributions during mid-July 2011, based on 3-min point counts in my early surveys after the accident, resembled the results obtained in 2012. When I first visited the area in 2011, the weather was not ideal for

[1]In Japanese, Sato means an area with human residences and activity, whereas Yama refers to hills or mountains covered with forest.

Table 12.1 The ten dominant bird species and their detection rates. Birds were detected during 5-min count surveys in an approximately 20 km × 20 km area in the northern Abukuma highlands of Fukushima prefecture in May and June 2012

Species	Rate per 56 counts from April 28 to May 4	Rate per 38 counts from June 13 to June 16
Cettia diphone	46/56 (82%)	36/38 (95%)
Hipsipetes amourotis	37 (66%)	32 (84%)
Emberiza cioides	34 (61%)	24 (63%)
Parus major	23 (41%)	18 (47%)
Phasianus versicolor	20 (36%)	17 (45%)
Corvus macrorhynchos	13 (23%)	18 (47%)
Passer montanus	20 (36%)	7 (18%)
Streptopelia orientalis	15 (27%)	11 (29%)
Carduelis sinica	8 (14%)	13 (34%)
Cuculus policephalus	0	20 (53%)

Large parts of this area were contaminated by radiation

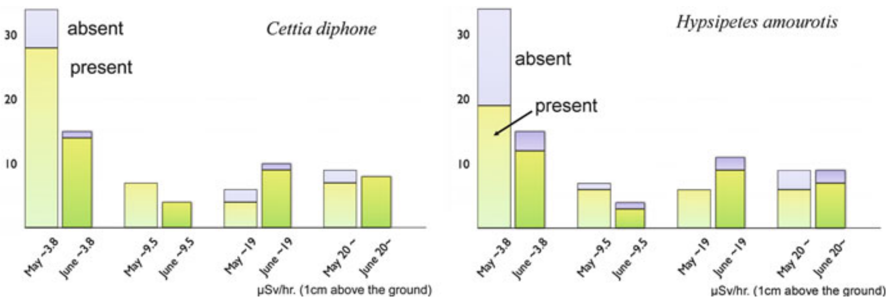

Fig. 12.4 Relationship between observation frequency and radiation levels in May and June 2012 (based on 94 separate 5-min counts), in the bush warbler (*left*) and the brown-eared bulbul (*right*)

studying the breeding bird community. The weather during summer 2011 was clearer and hotter than summer 2012 (Meteorological Agency, Japan), and I recorded only 26 bird species during 4 days. However, I recorded and caught bush warblers in the area that received the highest radiation dose, where I could enter and survey without any public permission in July and August 2011. My results are inconsistent with the conclusions published about birds in the Fukushima Daiichi area (Møller et al. 2012). It is possible that the situation was different in the two seasons of 2011 and 2012. However, the environmental cline from the center of the highlands to the west and northwest is clear; therefore, design of the bird count survey and their analyses by Møller et al. were inappropriate for discriminating the effects of radioactivity and other environmental factors. The ecosystem of the Abukuma highlands is quite complex and we need to accumulate more data to understand the wildlife activities and ecology of this area. I have been conducting automatic animal sound recording surveys since the summer of 2011. However, my analysis of the sound recordings is not yet complete.

During early May 2012, there were no oriental cuckoos (*Cululus poliocephalus*), whereas they were observed 20 times in 38 count surveys (53%) during mid-June 2012. It was not common in July or August 2012. This species is a parasite of the most dominant species, the bush warbler, and its calls are easy to detect from a distance. The presence of this parasite indicated the successful breeding of the warbler.

The top predators, the birds of prey *Buteo japonicus* and *Strix uralensis*, were common in the survey area, although their density was lower than the smaller bird species, whereas the number detected in the point count surveys during the daytime was very low. The abandoned paddy fields became dense grass bush because of the lack of agricultural management and they will soon become a secondary forest. This type of vegetation succession will not be profitable for the two main raptors, which mainly prey on small terrestrial mammals found in open grassland. This change will also possibly not be good for another raptor, *Butastur indicus*, which is common in the Satoyama landscape where it preys on frogs, snakes, and other animals.

The invasive alien *Garrulax canorus* was observed 9 times during 57 counts (21%) in May and 10 times during 38 counts (26%) in June. It prefers young forest and I observed it mainly around the plantation stands in the northern Abukuma highlands. The effects of this species on native species have yet to be examined.

12.4 Bush Warbler and Its Contamination with Radioactivity

In 2011, I captured four adult male bush warblers in mid-August using a mist net from the highly contaminated (>20 μSv/h) area of the Abukuma highland (Fig. 12.5). The rectrices (tail feathers) were collected and placed on imaging plates (IP) for 3 days. This yielded clear radioactive images of the feather shapes (Fig. 12.6, feather no. Dh and Di). However, the feathers collected in the Chichibu Mountains, western Saitama prefecture, approximately 250 km southwest of F1-NPP (Fig 12.1 right), and the feathers from a young female captured in an area with <2 μSv/h in October produced no images on the same IP (Fig. 12.6). The nuclear species and radioactivity levels of the four male feathers were measured using a germanium semiconductor detector. High doses of ^{134}Cs and ^{137}Cs were detected in all four, whereas ^{110m}Ag was detected in eight rectrices from male D (Fig. 12.7, Table 12.2). One rectrix from the individual D was cleaned with Kimwipes and ethanol, followed by ultrasonic cleaning for 20 min, but the contamination was only partially reduced (thick dots in Fig. 12.6, feather no. Di). We detected no highly contaminated points on the feathers (*thick dots* in Fig. 12.5, feather no. Dh and Di) during microscopic observations. Thus, very small particles with variable radioactivity levels appeared to be attached to the outside of the feather barbs and barbules. The feathers grew in 2010 during late summer, therefore, we hypothesized that this was not due to internal contamination of the feathers. The rectrices from one adult male collected in July 2012, two adult males and one adult female, and two juvenile males in August 2012, produced

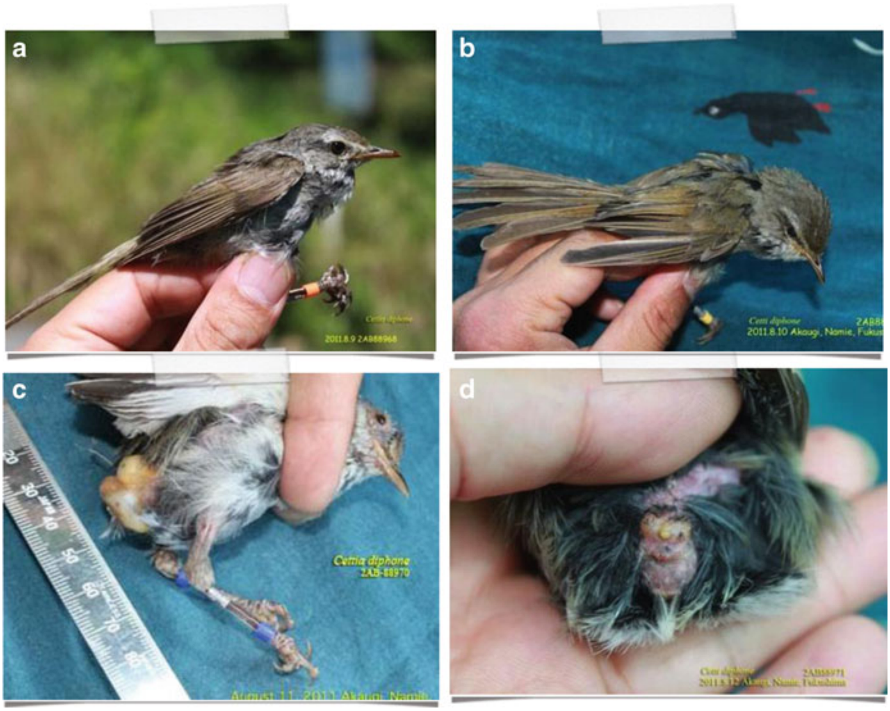

Fig. 12.5 Bush warbler males captured in highly contaminated areas of the Abukuma highlands. I caught these individuals in August 2011, when they were starting to molt

much weaker images after 3–5 days contact with IP. As indicated by Tanoi (2013), radioactivity was deposited on the leaves and ground after the accident, whereas newly grown leaves showed little or no contamination later in spring. Thus, birds that entered this area during the spring of 2012 appeared to have little contamination on their feathers. Birds that produced new feathers during late summer 2011 in this area should have some contamination both on the outside and from the inside.

The bush warbler usually migrates between its breeding and wintering sites. The distance ranges from 50 km east to 90 km south, e.g., the Chichibu Mountains in central Japan (K. Ishida, unpublished data). It is reasonable to assume that the population in Fukushima has almost the same migration habits as the Chichibu population because we can observe this warbler during the winter in the Hamadori and Nakadori areas. I observed a bush warbler during mid-March 2012 in the Abukuma highlands, although we had some snow accumulation at that time. Continuous automatic recording from winter through spring at 11 sites in this area, indicated that the warbler appeared (began to produce sounds) in mid-March in 2012. The differences in the temperature and snow accumulation between March 2011 and 2012 were

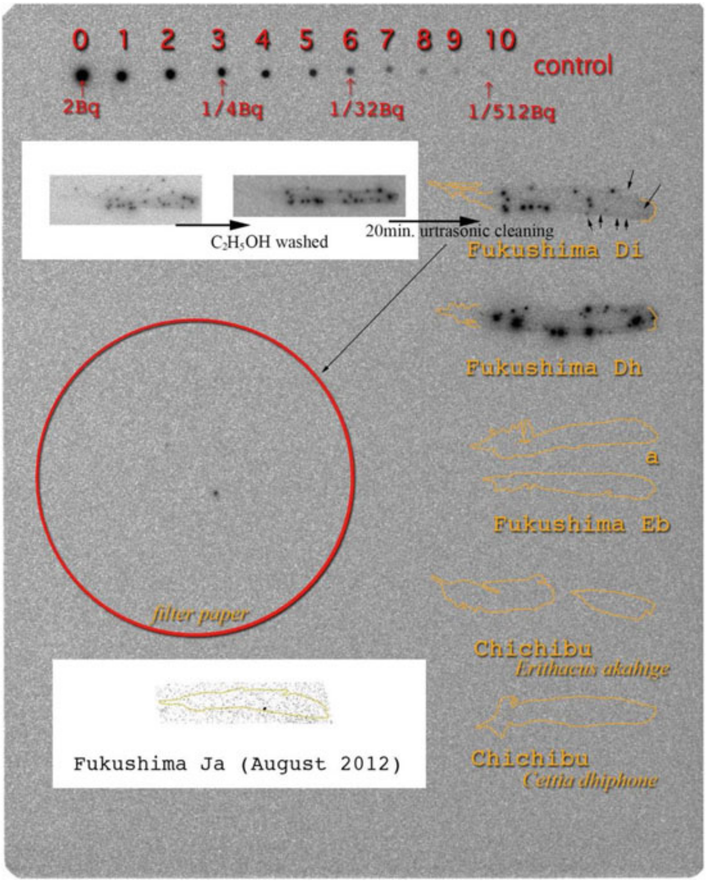

Fig. 12.6 IP figures of the bush warbler feathers, after 3 days of contact, D-h and D-i; Abukuma mountains, August 2011, E; Abukuma Mountains, October 2011, Ja; Abukuma Mountains, August 2012, Chichibu mountains, 250 km southwest of F1-NPP, August 2011 and October 2011. For D-i, the image shows a feather that was cleaned using Kimwipes and ethanol, followed by an ultrasonic cleaner. The Abukuma—August-2011 feather produced an abundance of contamination *dots*. The *red circle* indicates the paper filter contact image after percolation with the ultrasonic cleaning water of feather D-i. The outline of D-h and D-i feathers are clear and it indicates that the whole feather was highly contaminated. The *thick dots* indicate intensive (very high) radioactive contamination. The 11 control *dots* at the *top* indicate 2-Bq radiation and those, which are reduced sequentially twofold (from 2 to 1/512 Bq) from *left* to the *right*

small. The first song of the bush warblers in mid-March was almost "perfectly" performed at this highland. These observations suggest that the males caught in mid-August had bred in the area with high radioactivity contamination for 5 months. However, we cannot ascertain whether they were present when the F1-NPP accident occurred on March 11, 2011.

My results and the habits of the bush warbler population in Fukushima suggest that examinations of feather contamination estimate the level of radiation exposure of individual birds.

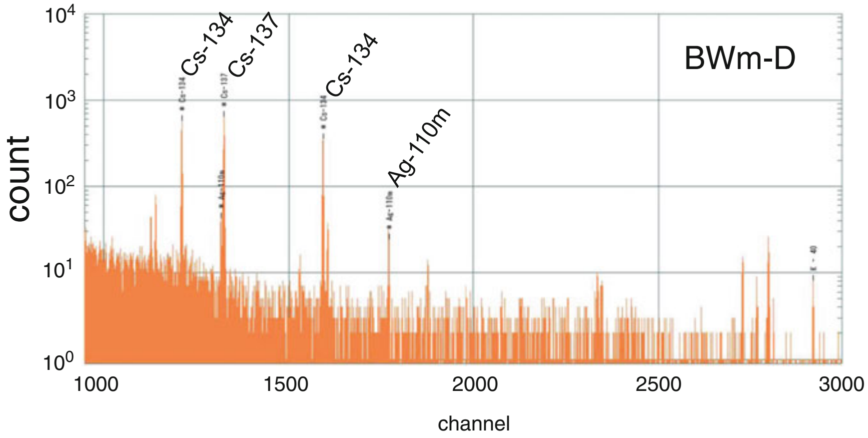

Fig. 12.7 The radioactivity profiles measured using a germanium semiconductor detector using eight rectrices from the individual D. ^{134}Cs, ^{137}Cs, and ^{110m}Ag were identified and the radioactivity was 12.5, 17.3, and 1.4 Bq, respectively. The total weight of the eight feathers was approximately 56 mg (Table 12.2)

Table 12.2 The contamination of feathers

Individual	No. of feathers	Total weight	Radioactivity[a] ^{134}Cs	Radioactivity[a] ^{137}Cs	Radioactivity[a] ^{110m}Ag	Date of capture
A	2	0.015 g	0.2 Bq	0.2 Bq	N.D.	August 9
B	2	0.014 g	2.5 Bq	3.6 Bq	N.D.	August 10
C	4	0.026 g	1.2 Bq	1.8 Bq	N.D.	August 11
D	8	0.056 g	12.5 Bq	17.3 Bq	1.4 Bq	August 12

[a]The experiments were conducted by Dr. K. Tanoi from the University of Tokyo. The detection period was 0.7–3.0 h

12.5 Effects of Radioactivity on Bush Warbler and Boar

One of the four bush warbler males had a “lesion looks very much like its due to avian poxvirus (dry chronic form)” (Lucio John Filippich, personal communication) around its cloaca (Fig. 12.5c). Almost the same appearance had been observed on the two males of the same bird species at Wakayama Prefecture, western Japan, in 2006 summer (Kumashiro et al., unpublished data). The lesions were not likely tumors but may be abscesses or dilatation of cutaneous glands, while histological examination was not performed (Hiroyuki Nakayama, personal communication). The same individual had *Leucocytozoon* protozoa in its blood, whereas the other three males did not (Koichi Murata et al., unpublished data). In the Chichibu mountains, blood parasite protozoa including *Leucocytozoon* spp. were found in 6/58 bush warblers (Imura et al. 2012). We are processing the plasma samples of Bush warblers to examine their stress levels and behavior (Wingfield et al. 1995).

Table 12.3 Total ^{134}Cs and ^{137}Cs levels in wild boar hind limb muscle

Radioactivity level (Bq/kg)	No. of individuals Fukushima prefecture	No. of individuals other prefectures	Date of capture
14,600[a]	1		September 5th, 2011
13,300	1		December 25th, 2011
8,000<	2		
4,000–8,000	5	4	
2000–4,000	30	4	
1,000–2,000	59	8	
500–1,000	32	14	
250–500	17	22	
100–250	6	20	

Boars were hunted in Fukushima, Miyagi, Tochigi, and Ibaraki Prefectures between May 2011 and March 2012

The data is published in Japanese at the following URLs:

http://wwwcms.pref.fukushima.jp/download/1/shizen23-inosisi.pdf

http://www.pref.miyagi.jp/sizenhogo/seibutu/honyurui/housyaseibussitukensa/housyaseikensa231205.pdf

http://www.pref.tochigi.lg.jp/kinkyu/d04/houshanou_choujuu.html

http://savechild.net/wp-content/uploads/2011/09/syokuhin.gif

[a]33,000 Bq/kg was reported from an individual captured on October 29, 2012 in Fukushima

Wild boars hunted in Fukushima and neighboring prefectures had various levels of ^{134}Cs and ^{137}Cs in their muscle (Table 12.3). The two highest levels were 14,600 and 13,300 Bq/kg, whereas more than half of those captured in Fukushima prefecture had >500 Bq/kg, as did many in other neighboring prefectures. The boar forages for animals and plants by digging through the litter on the ground and in the soil, which were highly contaminated with ^{134}Cs and ^{137}Cs. It is possible that the boar consumed radioactive materials directly from the soil and through food. The earthworms were also highly contaminated with radioactivity in the area (Hasegawa et al., unpublished data), which resembled the food web effect of radioactivity in the woodcock in Norway 5 years after the Chernobyl accident (Kålås et al. 1994). A copper pheasant (*Syrmaticus soemmerringii*) hunted at Fukushima in November had 735 Bq/kg, whereas 5/10 had >200 Bq/kg total radiocesium in their muscle (Fukushima Prefecture). This endemic Japanese pheasant feeds on ferns in winter, which are also known to concentrate radioactivity like mushrooms. Because we have not yet little evidence of the direct effects of radioactivity on wildlife at Fukushima, we will continue to observe and collect samples.

12.6 Factors that Affected the Wildlife

The accident occurred on March 11, before plants had begun to produce their spring shoots and birds had begun breeding. There was still some snow accumulation in the highlands of Fukushima. Thus, plant and animal cells were inactive at the time of the highest radioactive exposure. Therefore, the direct and decisive effects of the radiation from F1-NPP were low at that time. No forest death caused by the F1-NPP accident was observed, even in forests very close to it.

Radioactivity levels of 20 or 80 μSv/h can be detected around the highly contaminated ecosystem in the northwest area of F1-NPP, particularly close to the ground and in the litter. These levels are not sufficiently high to have obvious and direct results on the wildlife (Kryshev et al. 2005). The wildlife ecology is affected by various factors such as predators, pathogens, parasites, competitors, resource shortage, and low temperature. I caught healthy looking youngsters of bush warblers and their probable parents at a place with approximately 5 μSv/h dose rate (1 m above the ground) on August 14th, 2012. Several other individuals caught in this area also looked healthy and I could record their normal songs even at a point approximately 10 km northwest of F1-NPP of $>$40 μSv/h. In the northern Abukuma highlands of Fukushima, the lack of human activity is also an important factor that affects the wildlife. It has negative and positive effects depending on the ecology of each wild plant or animal. Thus, long-term and wide-range monitoring is required to understand the effects of the F1-accident and to distinguish the effects of radiation.

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Chapter 13
Remediation of Paddy Soil Contaminated by Radiocesium in Iitate Village in Fukushima Prefecture

Masaru Mizoguchi

Abstract Paddy soil freezes during winter in Iitate village. The frozen soil is as hard as asphalt, and local farmers can easily peel a plate of frozen soil using an agricultural machine. The radiation level in the ground surface was demonstrated to reduce from 1.28 to 0.16 μSv/h by stripping the frozen soil in field experiments. In this chapter, two remediation methods have been introduced and the collaboration between researchers, volunteers, and farmers in Iitate Village has been discussed.

Keywords Decontamination • Frozen soil • Rotary weeding method • Stripping method

13.1 Introduction

The radiocesium released from the Fukushima Daiichi nuclear power plant accumulates mainly within the top 5 cm of the soil surface. However, the state of the ground surface differs depending on the land use; therefore, radiocesium is distributed heterogeneously in space and depth. Thus, the efficient decontamination of contaminated soil demands the stripping of topsoil, which depends on the state of the farmland. The group known as the "Resurrection of Fukushima" has been independently developing soil decontamination methods in collaboration with the Agriculture Committee of Iitate village, although the Ministry of the Environment and the Ministry of Agriculture, Forestry, and Fisheries are also investigating various

M. Mizoguchi (✉)
Graduate School of Agricultural and Life Sciences, The University of Tokyo,
1-1-1 Yayoi, Bunkyo-ku, Tokyo 113-8657, Japan
e-mail: amizo@mail.ecc.u-tokyo.ac.jp

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decontamination methods. In this chapter, two methods developed by the group are described. In addition, the collaboration between researchers, volunteers, and farmers to decontaminate farmland has been discussed.

13.2 The "Resurrection of Fukushima" Group

This group is a volunteer organization, which aims to reconstruct the lives and the industries of the area that were severely damaged by the Great East Japan Earthquake and the accident at the Fukushima Daiichi nuclear power plant. The group has been conducting activities in the disaster-affected areas with a central focus on the Fukushima Prefecture. The group established a base of activities in Iitate village in June 2011, and it has advanced various projects aimed at the resurrection/reconstitution of the area while finding the best way of working with the affected people.

The major projects that are now under development are (1) conducting thorough radiation measurements and creating radiation dose survey maps; (2) decontamination demonstration experiments in houses, agricultural land, and forests; (3) care of affected people by teams of professionals including medical doctors; (4) publicizing the real lives of people in the area affected by the nuclear power plant accident through information communication technologies. The activities and results of the projects are available for public inspection on their current website (Resurrection of Fukushima 2012a).

13.3 Distribution of Paddy Soil Contaminated by Radiocesium in Iitate Village

There are many small paddy fields between the forests that comprise 74% of the Iitate village. Therefore, the soil and weather (such as the temperature and wind speed) in the paddy fields of the village differ depending on their location. On the other hand, because farmers in this district generally plow the paddy before winter, the paddy surfaces are uneven rather than flat. Figure 13.1 shows the distribution of radiocesium in paddy soil in 19 districts of Iitate village, which were measured by the group between January and March 2012 (Resurrection of Fukushima 2012b). Although radiocesium has been reported to accumulate near the ground surface (Shiozawa et al. 2011), Fig. 13.1 shows that the distribution of radiocesium was different in each place and it reached deeper levels in some places. This is because the surface state was different in each paddy field. In some places, summer grass was flourishing (Fig. 13.2) because farmers did not use their paddy in 2011, whereas wild boars had been digging holes in another paddy (Fig. 13.3). For efficient decontamination, it is necessary to create a pollution map and to ensure that the surface of the paddy is flat before decontamination.

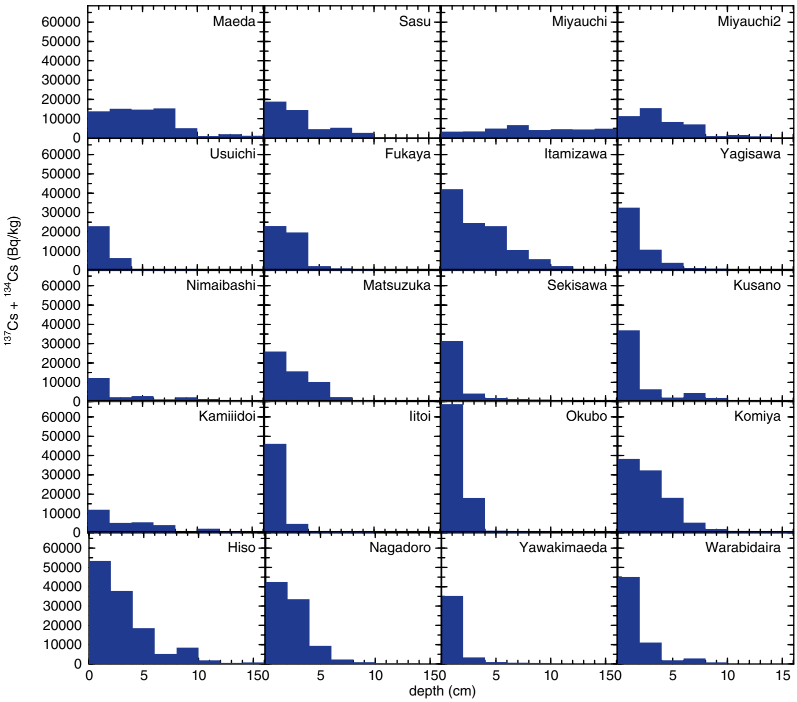

Fig. 13.1 Distribution of radiocesium in paddy soil from 19 regions in Iitate village (Resurrection of Fukushima 2012b)

Fig. 13.2 Summer grass growing on a paddy

Fig. 13.3 Paddy destructed by wild boar

13.4 Decontamination by Stripping Frozen Soil

The soil freezes during the winter in Iitate village. Figure 13.4 shows the changes in the soil temperature and soil moisture level (volumetric water content) at depths of 5, 10, and 15 cm. The accuracies of the soil temperature and moisture measurements were 0.1°C and 0.03 m^3/m^3, respectively. The soil temperatures at a depth of 5 cm reached 0°C, whereas the soil moisture level at the same depth fell to 0.2 m^3/m^3 at 6:00 on January 1st, 2012. This indicated that the soil froze to a depth of at least 5 cm on January 1st, 2012.

The frozen soil is as hard as asphalt and farmers could easily strip the frozen soil using a backhoe, which farmers normally possess (Fig. 13.5). In field experiments, we confirmed that the level of radiation in the ground surface reduced from 1.28 to 0.16 μSv/h on stripping the frozen paddy soil (Fig. 13.6). The radioactivity in the unfrozen soil after stripping was 2,670 Bq/kg, whereas that in the stripped frozen soil was 23,760 Bq/kg. This dramatic reduction in the radioactivity level indicated that the radiocesium in the topsoil could be removed by stripping the

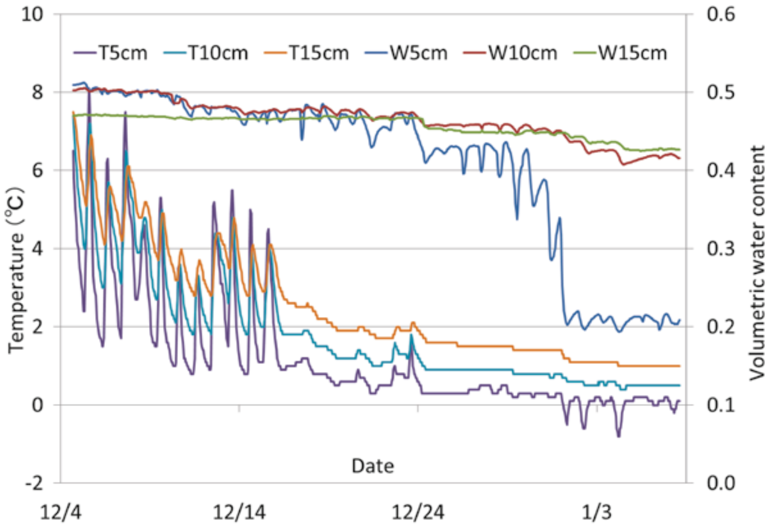

Fig. 13.4 Changes in the soil temperature and soil moisture level at depths of 5, 10, and 15 cm in a paddy in Iitate village

Fig. 13.5 Stripping the frozen soil using a backhoe by a local farmer on January 8th, 2012

Fig. 13.6 The frozen soil stripped using a backhoe formed a plate-shaped mass

frozen soil. This simple method exploits a natural process. Therefore, it was possible to reduce the risk exposure for workers engaged in decontamination work. We also confirmed that the radiation dose could be reduced further if the frozen soil is removed carefully without turning it upside down.

The difficulty with this method was predicting the timing of stripping frozen soil. This was because more fertile soil was lost if a thicker layer of frozen soil was stripped. Therefore, it was necessary to predict the optimum thickness of frozen soil before stripping. This was achieved using a hand-made freezing depth gauge. Figure 13.7 shows a test tube containing an aqueous solution of methylene blue that was buried in the field. The thickness of the frozen soil matched the thickness of the clear ice. It was also possible to estimate the thickness of the frozen soil from the air temperature in the field. However, because this estimation method is complex, it has been described in the appendix.

In Iitate village, the topsoil on some farmlands contains radiocesium mixed with the subsoil. Thus, the topsoil stripping method cannot be used for these farmlands. However, if the clay particles that fixed radiocesium are accumulated by puddling the paddy soil before winter, radiocesium can be removed effectively by only stripping the frozen soil during winter.

The soil moisture moves into the frozen soil during the freezing process on farmlands. Therefore, the frozen soil stripping method can also be used with relatively dry soil. However, the frozen soil on pasture land is not hard because of its low bulk density. In this case, we must use a heavy machine to compress the topsoil before winter to increase the efficiency of stripping frozen soil.

Fig. 13.7 A hand-made freezing depth gauge containing an aqueous solution of methylene blue

13.5 Decontamination Method Using a Rotary Weeding Machine

Rotary weeding machines (intertillage weeding machine) are used in Japan for weeding after planting. In April 2012, we irrigated a paddy with water to a depth of approximately 5 cm and agitated the soil surface using a rotary weeding machine. The muddy water was swept out into a drainage ditch using a tennis court brush (Fig. 13.8). We repeated this operation thrice and measured the amount of radiocesium in soil. Figure 13.9 shows the radiocesium profiles before/after rotary weeding. The results showed that approximately 80% of the radiocesium was removed from the paddy using this method.

The problems that remained were the efficiency of the work and the treatment of the muddy water. This experiment was performed manually. However, given the vast area of farmland that needs to be decontaminated, we will require a machine to improve the efficiency of work. In the muddy water treatment, we swept the muddy water into a drainage ditch that measured 1 m deep and 50 cm wide, which was dug around the paddy field. After 3 months, the muddy water infiltrated into the soil and remained as a dry clay layer on the surface of the ditch. Figure 13.10 shows the concentration of radiocesium in the soil surrounding the drainage ditch (Mizoguchi 2012). The radiocesium had penetrated to a maximum depth of 6–7 cm

Fig. 13.8 The rotary weeding method tested by a volunteer group (Resurrection of Fukushima 2012c)

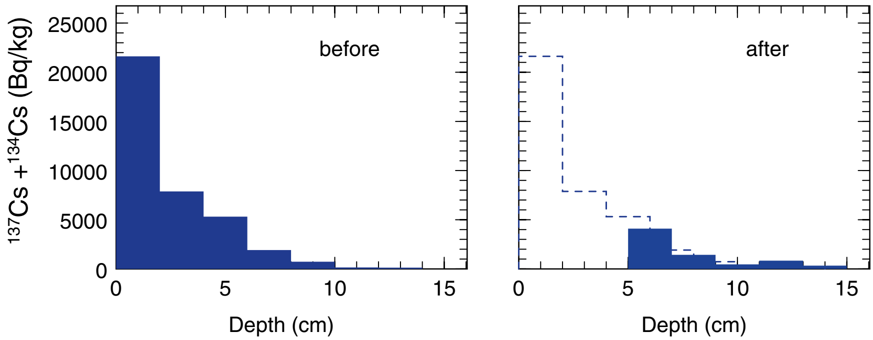

Fig. 13.9 The radiocesium profiles before/after rotary weeding (Resurrection of Fukushima 2012c)

in the soils at the bottom and on the walls of the ditch. Thus, the drainage ditch had a filtration capacity (Yahata 1980) and it effectively trapped clay particles that contained radiocesium.

Another possible muddy water treatment is to gather muddy water in a downstream paddy and keep the paddy flooded to attenuate the radiation dose from the ground surface. The frozen soil can be stripped in winter after the natural infiltration of water just before winter.

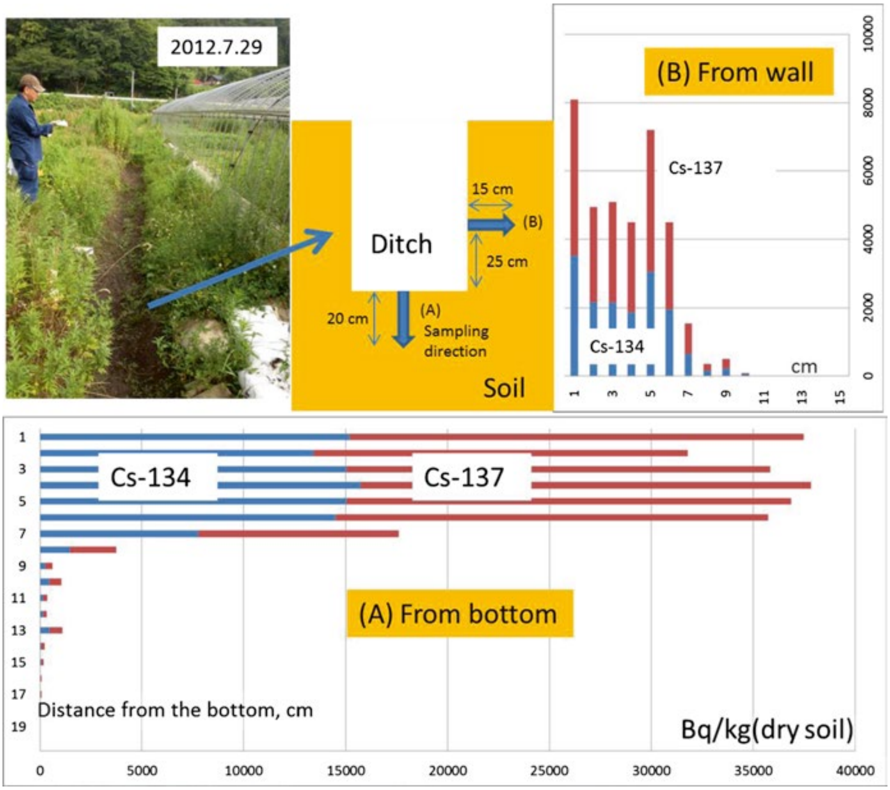

Fig. 13.10 Concentration of radiocesium in the soil surrounding the drainage ditch (Mizoguchi 2012)

13.6 Radiation Dose Reduction Method Based on Soil Cover

The treatment of the removed contaminated soil is a big challenge. It is difficult to find a waste disposal site in Iitate village. Even if one can be located, the capacity of the disposal site is insufficient. There is also a risk of environmental contamination with radiocesium while carrying the waste soil to the waste disposal site. Thus, it might be practical to bury the contaminated soil in a hole in a corner of the paddy field. A laboratory experiment showed that a 50-cm thick layer of soil could reduce the radiation dose to approximately 1/100 (Miyazaki 2012).

However, the problem with this method will be preventing the leakage of radiocesium from contaminated soil, which depended on soil type, soil moisture, and groundwater level. If there is sand or gravel in the lower layer of the paddy, it is necessary to dig a trench that is shallower than the layer. Assuming that we need to bury the top 5–10 cm layer of the surface soil at a 1-m depth, the burial area have to be approximately 10% of the paddy field area.

Radiocesium is fixed by weathered micas, which are clay minerals found in the typical soils distributed in the Iitate village area. After radiocesium is fixed by these clay minerals, it moves in the soil together with the clay particles, which are trapped by the filtration capacity of the soil. The radiocesium penetrated into a 6–7 cm layer of soil around the drainage ditch (Fig. 13.10). Thus, we will be able to reduce the radiation dose from the ground surface by covering the ditch with fresh soil.

From the perspective of soil physics, the best method is to bury the waste soil containing radiocesium in a hole at the corner of the paddy field. However, in order to convince the neighboring residents about the safety of this method of waste soil treatment, it is necessary to develop a method for monitoring the radiation in the soil around the waste soil. It is also necessary to demonstrate that the clay particles never flow into the downstream basin using a simulation based on the theory of soil physics in a condition of variable groundwater levels.

13.7 Environmental Monitoring in the Village

After decontaminating the paddy field, we need to evaluate the effectiveness of this decontamination procedure. In this evaluation, it is necessary to continuously monitor the soil radiation at important points (hotspots). In particular, it is important to observe the relationship between the radiation dose and the weather, particularly the precipitation and wind. It is also important to observe the relationship between precipitation and runoff/turbidity of rivers. However, radiation meters are too expensive for the general public. Thus, there is a need for an economical radiation dose sensor that provides relatively good value. Fortunately, a volunteer group has recently developed a Pocket Radiation Sensor in response to this need (the nonprofit project "radiation-watch.org," 2011).

I have also been developing a field monitoring system (FMS) for quasi-real-time data collection in a remote agricultural field in Asia (Mizoguchi 2011). After adding this new radiation sensor to FMS, in situ soil monitoring was initiated in Iitate Village. Figure 13.11 shows a comparison of the radiation measurements and images captured in a garden, a forest, and a deforested area from the autumn to the winter. The radiation levels fluctuated daily, particularly in the house, but they declined after January 20 when snow started to cover the ground. This showed that snow prevented radiation being emitted from the ground. In addition, we also collected a great deal of valuable radiation data using FMS. The preliminary comparisons of the radiation and meteorological data showed that the radiation levels tended to be higher on fine days with low humidity.

13.8 Conclusion: The Roles of Researchers in Society

Decontamination of farmland is the biggest challenge faced in agricultural engineering. There are differences in our positions in the nation and in organizations, but researchers are required to make a rapid response with many responsibilities. Radioactivity is an invisible opponent. Therefore, the risk of exposing young people in the Iitate village fields cannot be taken. However, senior volunteers in their 70s are working vigorously

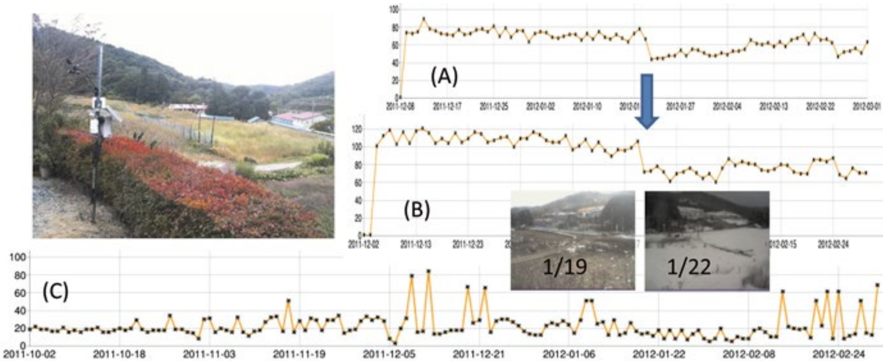

Fig. 13.11 Comparison of the radiation levels in three FMS sites: a garden, a forest, and a deforested area in Iitate village

in the fields each weekend. Their activities are changing the roles of researchers in our society. Beyond the confines of our specific disciplines, we must confront common challenges such as the decontamination of farmland with flexibility, depending on the situation. At present, we must question the identity of researchers in Japan.

Acknowledgments All field experiments were conducted with the cooperation of Mr. Muneo Kanno, Agriculture Committee in Iitate village, and members of the "Resurrection of Fukushima" (Representative: Dr Yoichi Tao). I express my sincere gratitude to them for their contribution. In addition, I thank the agricultural engineering team for reconstructing Fukushima at the University of Tokyo (Representative: Prof. Naritaka Kubo) and the Early return to village project at the Meiji University (Representative: Prof. Kosuke Noborio) for their assistance. Each FMS system was donated by Decagon Devices, Inc., AINEX Co., Ltd, and X-Ability Co., Ltd. through The Japanese Society of Irrigation, Drainage, and Rural Engineering (JSIDRE).

Appendix: Equation for Estimating the Frozen Soil Depth Based on the Air Temperature

Assuming that the temperature of the freezing front is a constant 0°C and there is a linear change from the ground surface to the freezing front, the heat balance of the freezing front can be expressed using the following differential equation:

$$k\frac{T}{x} = \theta L\frac{dx}{dt}$$

where T = temperature at the ground surface, x = the frost depth, k = the thermal conductivity of frozen soil, θ = volumetric water content of frozen soil, L = latent heat of freezing, and t = time. In a case without θ, this is known as the Stephan equation. This equation can be solved easily by separating the variables, where the frost depth is expressed using the following equation.

$$\frac{k}{\theta L}\int Tdt = \frac{x^2}{2}$$

$$x = \sqrt{\frac{2k}{\theta L}\int T dt}$$

If we introduce the freezing index F, which is defined as $F = \int T dt$, the frost depth x is given as follows:

$$x = \sqrt{\frac{2k}{\theta L} F}$$

where k and θ are physical properties of the frozen soil, and L is a constant. Therefore, the frost depth can be estimated as the square root of the freezing index F.

Theoretically, it is necessary to obtain F based on the ground surface temperature T. However, if we use the air temperature as a substitute for the ground surface temperature, the frost depth can be estimated from the air temperature data alone in the field using an empirical parameter α.

$$x = \alpha\sqrt{F}$$

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Chapter 14
Distribution of Radiocesium from the Radioactive Fallout in Fruit Trees

Daisuke Takata

Abstract We report the impact of radiocesium released by the Fukushima Daiichi nuclear power plant accident on fruit trees in the vicinity. Specifically, we discuss our findings related to the absorption and translocation of radiocesium in peach (*Prunus persica* L. Batsch) trees because peaches are a major crop in Fukushima Prefecture.

At the time of the nuclear accident, the majority of deciduous fruit tree species had no foliage and had not entered the flowering stage. This was also the case for peach trees, although we confirmed at harvest time that radiocesium had migrated to new plant parts. One possible explanation for this is that the presence of radiocesium in the new leaves and fruits, which was not present at the time of the nuclear accident, was affected by direct radiocesium deposition on the existing above-ground plant parts and this may have been absorbed by the tree. In the year of the accident, the root uptake of radiocesium deposited on the soil contributed very little to the overall contamination compared with absorption by the trees through the above-ground plant parts. The magnitude of radiocesium translocation from the old to new plant parts was significantly different between the year of the accident and the year after the accident. We report the findings since the Fukushima Daiichi nuclear power plant accident and speculate the pathways of radiocesium entry and release in peach trees.

Keywords Bark • Contamination • Fruit tree • Peach • Radiocesium

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D. Takata (✉)
Institute for Sustainable Agro-ecosystem Services, Graduate School of Agricultural and Life Science, The University of Tokyo, 1-1-1 Midori-cho, Nishi-tokyo-shi, Tokyo, Japan
e-mail: takata@isas.a.u-tokyo.ac.jp

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14.1 Introduction

There are numerous reports of the effects of radionuclides on annual crops, which are related to the impact of fallout from atmospheric nuclear tests conducted since 1950s (Mitsui and Tensyo 1958), reports of the Chernobyl nuclear power plant accident aftermath (IAEA 2010), and various experiments conducted under controlled conditions involving the direct application of radionuclides to plants (Ralls et al. 1971). There are also reports related to the transfer of radionuclides from the soil to perennial fruit trees, i.e., reports that examine the transfer factors (TF) for radionuclides, but are few in number. Furthermore, tree fruits are often lumped together with fruits and vegetables, such as tomatoes and strawberries (IAEA 2010). Many investigations of fruits have dealt with the translocation of radionuclides from the soil to plant parts e.g., the study of grape leaves (Zehnder et al. 1995).

Annual peach production in Japan is approximately 170,000 tons, the majority of which is intended for fresh consumption. Given the high demand for peaches for table consumption and as gifts, they fetch relatively high prices. Peaches are among the major commodities produced in Fukushima Prefecture, where the annual peach production was valued at ten billion JPY and accounted for approximately 1/5th of the total peach production in Japan in 2010. However, there have only been a few attempts to understand the migration of radionuclides in peach trees. Therefore, there is an urgent need to clarify the impact of radionuclides on peach trees.

Following the Fukushima Daiichi nuclear power plant accident, surveys and cultivation experiments have been conducted to investigate the migration of radiocesium in peaches and other fruit trees in Fukushima and Tokyo. Based on information derived from these investigations, the factors related to radiocesium translocation and pathways of absorption by fruit trees in the year of the nuclear accident have been reported in this paper in addition to brief descriptions of ongoing investigations.

14.2 Pathways of Radiocesium Absorption by Fruit Trees

There are two apparent pathways of radiocesium entry into plants, i.e., uptake of radiocesium deposited on the soil through the roots and direct absorption of radiocesium deposited on the above-ground plant parts. For direct absorption, it is necessary to distinguish the deposition of radiocesium on existing plant parts, such as branches and the main trunk that were present at the time of the Fukushima Daiichi nuclear power plant accident, from the deposition of radiocesium in new leaves, shoots, and fruit that emerged after the accident.

14.2.1 Uptake by Roots

The Chernobyl nuclear power plant accident occurred when leaves had sprouted on many tree species. There has been much discussion about the direct absorption of radiocesium by the new plant parts with deposition. However, the Fukushima Daiichi nuclear power plant accident occurred when the majority of deciduous fruit trees, with the exception of plums and a few other early blooming species, had not yet flowered or produced foliage. Thus, it is expected that the pathways and levels of radiocesium absorbed by the plants in Fukushima will differ from those in Chernobyl. As expected, in the 2011 crop of plums, which had already finished flowering at the time of the accident, radiocesium was detected at levels exceeding the tentative allowable standards. These elevated levels were due to the direct airborne deposition of radiocesium on the plum flowers. However, radiocesium was also detected in fruits, leaves, and other new plant parts of fruit species such as peach and persimmon that had not produced foliage at the time of the accident. Moreover, field crops and tree species were found where the levels detected exceeded predictions based on TF using data collected before the accident. According to data collected before ploughing in May 2011, 96% of the radiocesium found in soil at a depth of 0–5 cm below the soil surface was radioactive after the accident (Shiozawa et al. 2011). While it has been demonstrated that radiocesium is absorbed by structures on the root surfaces (Eichert et al. 1998), with the exception of shallow-rooted species such as fig, the majority of the root mass of fruit trees is found at depths >5 cm. Given that the radiocesium had not yet migrated to deeper soil strata during the year when the nuclear accident occurred, it is possible that there was little uptake of radiocesium through the roots.

14.2.1.1 Is It Possible to Estimate the Amount of Radiocesium Absorbed from the Soil by Fruit Trees in the First Year After the Nuclear Accident?

The presence or absence and the contribution of soil radiocesium uptake by fruit trees following the Fukushima Daiichi nuclear power plant accident was estimated using trees that were grown under conditions where there was no radionuclide fallout on the soil. Takata et al. (2012c) also conducted a different experiment where the soil surface of container-grown "Akatsuki" peach trees had been covered 1 month before the accident (Fig. 14.1). After the accident, these plants were quickly incorporated into an experiment that aimed at estimating the uptake of radiocesium from the soil.

We used eight 5-year-old "Akatsuki" peach trees, which were grown outdoors under restricted root zone conditions in containers placed in an experimental field. Each pot contained 36 l of an 8:5 mixture of loamy soil and leaf mold. The soil surface of four of the trees was covered with weed control fabric since February 11, 2011.

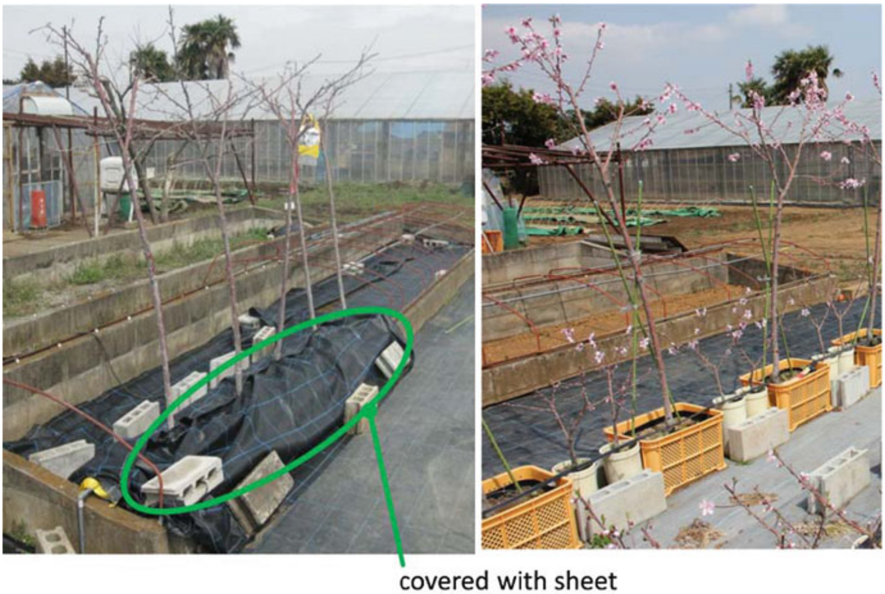

Fig. 14.1 Peach trees in containers with covering the soil surface. *Left*: covered (February 2011), *right*: without covered (March 2011)

Table 14.1 Concentration of ^{134}Cs, ^{137}Cs and ^{40}K in soil planted "Akatsuki" peach tree in Tokyo, respectively (from Takata et al. 2012c)

		Radioisotopes(Bq/kg-dry weight)		
Condition	Depth	^{134}Cs	^{137}Cs	^{40}K
Non-covered	0–5 cm	75.7 ± 6.2	87.4 ± 7.0	247.6 ± 83.6
	5–20 cm	20.0 ± 1.4	26.7 ± 1.7	220.6 ± 32.8
Covered	0–5 cm	15.2 ± 1.4	11.7 ± 1.4	220.4 ± 18.0
	5–20 cm	6.7 >[a]	7.8 >[a]	228.6 ± 13.6

[a]Indicates detection limit value

The remaining four trees, designated as the test samples, were cultivated without covering the soil surface. Peaches were harvested and the trees were dissected in July. The tree parts were separated into fruits, leaves, shoots, old branches (1–3 years old), main trunks (4 years old), and roots. Fruit were separated into the pericarp, pulp, and stones/seeds. The old branches and main trunks were separated into the exterior parts (bark), including the cambium, and the interior parts (wood). The container soil was sampled at 0–5 and 5–20 cm. After washing, all dissected samples were oven-dried and weighed, and the amount of radiocesium in each plant part was assessed.

In the containers in which the soil was covered, the radiocesium level in the surface soil layer was approximately 1/7th of that in the uncovered soil and below the detection limit in the deeper soil (Table 14.1). We attributed the detection of radiocesium in the surface soil of the covered containers to the relatively porous cover used to cover the soil because of the low flood tolerance of peaches. We also detected radiocesium with an

Table 14.2 Concentration of ^{134}Cs, ^{137}Cs and ^{40}K in container grown "Akatsuki" peach tree in Tokyo, respectively (from Takata et al. 2012c)

		Radioisotopes(Bq/kg-dry weight)		
Condition	Part	^{134}Cs	^{137}Cs	^{40}K
Non-covered	Mature fruit			
	– Skin	32.3 ± 3.8	52.7 ± 4.5	556.0 ± 32.9
	– Pulp	12.1 ± 3.5	15.7 ± 3.2	515.4 ± 56.2
	– Stone with seed	5.5 >[a]	6.7 ± 2.1	331.0 ± 26.3
	Shoot	22.0 ± 2.9	23.4 ± 3.2	415.2 ± 50.7
	Leaf	31.5 ± 2.9	40.9 ± 3.4	908.3 ± 93.3
	Branch			
	– Bark	832.9 ± 14.6	998.6 ± 14.1	201.3 ± 100.8
	– Wood	14.2 ± 7.3	17.4 ± 2.5	131.5 ± 33.0
	Trunk			
	– Bark	321.8 ± 19.7	389.8 ± 11.7	184.9 ± 46.0
	– Wood	12.8 ± 7.2	15.5 ± 6.5	143.8 ± 22.0
	Root	2.3 >[a]	4.1 >[a]	116.1 ± 10.1
Covered	Mature fruit			
	– Skin	37.2 ± 5.6	44.5 ± 6.2	756.4 ± 74.3
	– Pulp	12.6 ± 1.7	14.0 ± 1.9	474.8 ± 31.9
	– Stone with seed	6.6 >[a]	9.4 ± 2.2	315.4 ± 32.9
	Shoot	27.7 ± 5.9	37.9 ± 7.2	618.9 ± 102.4
	Leaf	41.6 ± 7.7	54.4 ± 9.1	1148.7 ± 117.3
	Branch			
	– Bark	761.1 ± 15.9	1047.8 ± 19.3	214.1 ± 43.3
	– Wood	12.2 ± 3.4	9.8 ± 3.4	100.7 ± 36.2
	Trunk			
	– Bark	319.8 ± 28.9	379.0 ± 19.1	180.8 ± 21.0
	– Wood	10.7 ± 2.8	14.9 ± 0.8	144.9 ± 16.6
	Root	2.6 >[a]	2.9 >[a]	117.3 ± 39.5

[a]Indicates detection limit value

unknown isotopic composition in surface soil from another part of the experimental field covered with the same fabric, which led us to believe that radiocesium could pass through the covering material, albeit in small quantities. Next, we examined the concentration of radiocesium in different plant parts (Table 14.2). Because we did not observe any differences in the radiocesium concentrations in any of the above-ground plant parts between trees grown in covered and uncovered containers, we concluded that the amount of radiocesium absorbed by the trees through the above-ground plant parts greatly exceeded that absorbed from the soil, at least in the year of the accident. In addition, we did not detect radiocesium in the roots of trees grown in covered or uncovered containers. Thus, we concluded that it was unlikely that the root uptake of radiocesium deposited on the soil surface had a significant role under natural conditions. In addition, the results clearly indicated that the majority of the radiocesium detected in trees was absorbed through the above-ground tree parts during the year of the accident.

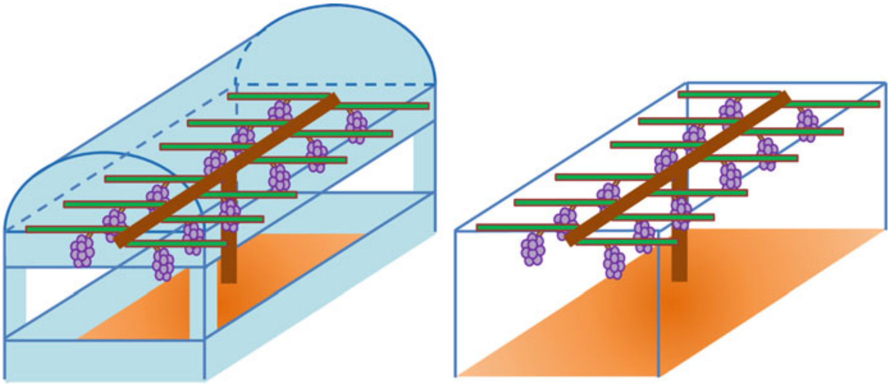

Fig. 14.2 Cultivation environment in Fukushima and Tokyo and concentration of $^{134+137}$Cs in soil and grapes berry

14.2.1.2 Field-Dependent Differences, After Accounting for Differences in the Cultivation Environments

Next, we attempted to estimate the uptake of radiocesium from the soil based on the characteristics of different fields. We measured the concentration of radiocesium in various parts of "Kyoho" grape trees (*Vitis* spp.) cultivated in Fukushima and Tokyo. It is reasonable to assume that under the same cultivation conditions, the concentration of radiocesium in fruit would be higher in trees grown in Fukushima than those grown in Tokyo because of the higher airborne radiocesium dose rates in Fukushima. However, it would be difficult to conclude that the remaining differences in the concentration were solely attributable to differences in the soil. Therefore, we compared grapes grown in a plastic-covered greenhouse in a field in Fukushima with grapes grown in an open field in Tokyo (Fig. 14.2). In the covered field, grapes were grown in open-sided greenhouses, where the tops were covered with plastic film but the sides were open. The soil itself was not covered with plastic or fabric, but the concentration of radiocesium in the surface layer of the soil inside the greenhouse was approximately 3% of that in the adjacent uncovered areas (Fig. 14.2, bottom). The concentration of radiocesium in the surface soil was approximately three times that of the surface soil in the Tokyo field (256.8 vs 82.7). The concentration of radiocesium was higher in the fruits grown in Tokyo than those grown in Fukushima. The radiocesium concentration in the other above-ground plant parts was higher for grape plants grown in the uncovered field in Tokyo or the same for grape plants grown in both fields, which did not reflect the difference in the radiocesium concentration of the soil. These results suggest that above-ground sources accounted for more of the radiocesium absorption by grape plants than below-ground sources at least in the year of the accident.

These experiments suggested that differences in the soil radiocesium concentration did not affect the concentration of radiocesium in various tree parts 5 months after the accident. Thus, the proportion of radiocesium absorbed by the trees through the roots was substantially lower than the proportion absorbed directly by plant parts such as the bark, which were in direct contact with radiocesium that adhered to the surface, at least in the year of the accident. Only a small proportion of radiocesium was translocated from above-ground plant parts to below-ground plant parts in the 5 months after the accident. Thus, it is difficult to estimate the absorption of radiocesium by below-ground plant parts in natural conditions, i.e., TF, at least in the year of the accident. It should be noted that the TF of plants cultivated in natural conditions are not strictly an assessment of radiocesium absorption from the soil alone, but rather represent apparent TF that also include the absorption of radiocesium deposited directly on the bark and other plant parts. Thus, in the case of radiocesium detected in the fruit of peach trees on which leaves had not sprouted at the time of the accident, there is a need to investigate the possibility, described in greater detail below, that the radiocesium was absorbed through the bark or was absorbed after being deposited by the wind or rain on leaves and fruits after sprouting.

14.2.2 Translocation of Radiocesium from Existing Above-Ground Plant Parts

14.2.2.1 Radiocesium Concentration in the Above-Ground Plant Parts

As explained in Sect. 14.2.1, there was a need to investigate the possibility of radiocesium absorption by the above-ground tree parts. First, we present a brief summary of an investigation that clarified the relative distribution of radiocesium in the above-ground tree parts (Takata et al. 2012b). The samples comprised three "Chiyohime" peach trees, which were grown in an experimental field (soil type: andosol) in the grounds of the University of Tokyo. On the day of harvest (June 22), we collected fruits, shoots, and leaves from bearing branches, shoots and leaves from succulent shoots, and 3-year-old branches (secondary scaffold limbs). Fruits were separated into three parts: the pericarp, pulp, and stones/seeds. In addition to harvesting the cover crop from 50 × 50-cm plots established approximately 75 cm south of the crowns of the sampled trees, we collected soil samples from the same plots at various depths. We also collected peach tree roots (2–5 mm in diameter) from the same plots. We used soil samples collected from the same field on September 16, 2010, as reference samples. We also separated the succulent shoots, 3-year-old branches, and roots into exterior parts (bark), including the cambium, and interior parts (wood).

When the radionuclide concentrations in the shoots and fruits were low, the ^{134}Cs and ^{137}Cs concentrations in the bark of 3-year-old branches were approximately 25 times of those observed in the pulp at 759.8 and 811.6 Bq/kg dry weight, respectively. These concentrations were also substantially higher than those in the soil and the cover crop. Thus, it is unlikely that soil-to-plant migration was the only process involved. Meteorological data for the experimental field indicated that rainfall occurred from March 21 to March 24, and it is believed that radiocesium was deposited during this period. However, the peach trees had not flowered or sprouted at this point and the flower buds still measured 3 mm. Thus, it is difficult to imagine that the radionuclides detected in the leaves and fruits in this investigation originated solely from the intense fallout that occurred in the brief period immediately after the nuclear accident. The bark:wood radiocesium ratio varied greatly from 30:1 to 200:1 in the 3-year-old branches, particularly high radiocesium levels were detected in the bark of trees grown in Tokyo, which is a low-dose area, and Fukushima (Takata et al. 2012a, b, d, e, 2013). These results suggested that there was a need to carefully investigate the translocation of radiocesium to the bark.

There also tended to be lower partitioning of radiocesium to the fruits in high-contamination areas than in the low-contamination areas. This result suggest that, unlike potassium, which is abundant in the soil and is actively translocated to new above-ground plant parts, cesium is not translocated in large quantities or that the radiocesium was deposited directly on the leaves and fruits after sprouting.

14.2.2.2 Translocation from the Bark

We conducted an experiment using peach trees to study the inward translocation of radiocesium from above-ground plant parts, particularly to the interior of the bark layer. Using an imaging plate, we visualized the distribution of radionuclides within the main trunks of the peach trees collected in January 2012, in Date-shi, Fukushima Prefecture (Takata et al. 2012f). The images shown in Fig. 14.3 are composites of the developed imaging plates placed over regular photographs of the samples. The black spots represent areas where radionuclides were detected. It should be noted that these were 6-year-old trees; thus, the outer bark was relatively smooth and lacked the roughness that is typical of the outer bark. Furthermore, all the samples were washed before imaging. Radionuclides were detected only in the outermost layer (epidermis) of the peach tree bark and not in the layer immediately below the epidermis. Using a germanium semiconductor detector, we determined that the concentration of naturally occurring ^{40}K did not differ in the epidermis and lower layers, implying that the different image intensities were the result of radionuclides that originated from the nuclear accident, including radiocesium. Furthermore, we used a germanium semiconductor detector to measure the radiocesium concentration in the epidermis and the layer immediately below the epidermis, which yielded the same result and showed that extremely high concentrations of radiocesium were present in the epidermis, i.e., ≥50 times that in the lower layers (Takata et al. 2012d, f). Based on these observations, we considered that radiocesium deposited on the trees

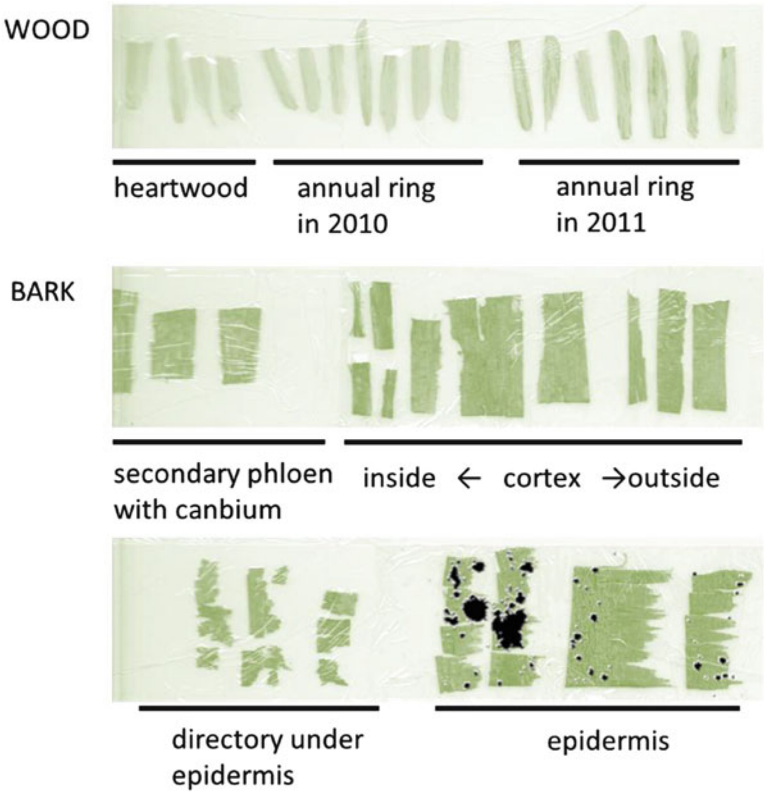

Fig. 14.3 Imaging plate of trunk in "Akatsuki" peach (Date Fukushima)

was unevenly distributed in extremely high-concentration patches within the thin outermost layer of the bark. An analogous experiment in August 2011 using imaging plates and germanium detection with peach trees collected from Higashi-shirakawa-gun in Fukushima Prefecture, which was a low-contamination area, produced similar results, albeit with different concentration ranges (Takata et al. 2013). However, these results do not prove that the radiocesium in the bark was translocated to internal structures such as phloem or xylem vessels. It is possible that the radiocesium was strongly bound to the epidermis itself and was not translocated in significant quantities to the water-carrying phloem located deeper in the tree. Thus, the potential and magnitude of the inward translocation of radiocesium in the epidermis is still unknown.

We can only speculate the pathways through which radiocesium migrates inward from the bark, but it is possible that it is somehow translocated through lenticels on the bark to the phloem or wood (Tanoi et al. 2012). After analyzing the imaging plates, we located radiocesium in the vicinity of lenticels (Fig. 14.4). Based on these results, we suggest that radiocesium was translocated inward through lenticels on the bark. However, lenticels are generally considered to be *structures for gas*

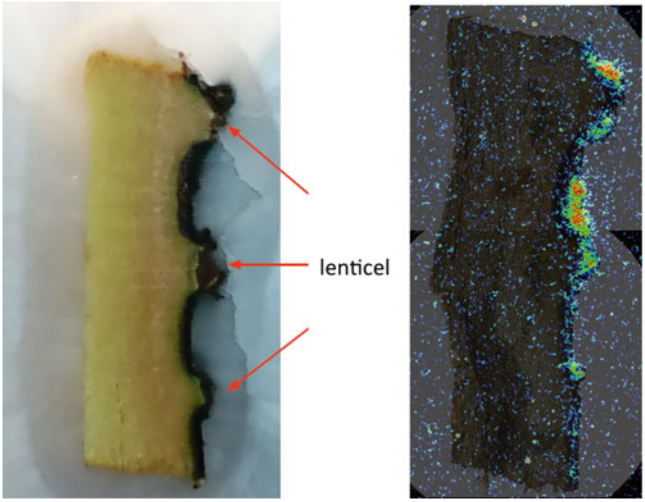

Fig. 14.4 Imaging plate of lenticel in "Akatsuki" peach (grown in Fukushima). The photo and image was obtained by Atsushi Hirose (University of Tokyo)

exchange and they exist on the bark in the form of cork. Thus, because they are relatively impermeable to water, they would not be expected to provide an effective pathway for radiocesium translocation in their typical state. Bearing this in mind, we generated imaging plates of the structures directly beneath the lenticels using peach trees collected in Fukushima as samples. We divided the lenticels into two groups—those with and without cracked centers. However, we were only able to image radionuclides in the structures below the former group of lenticels (unpublished data). The fact that radionuclides were only detected beneath cracked lenticels suggests that radionuclides were absorbed in these locations. The size of lenticels on peach tree bark varied substantially with branch age, but a greater number of lenticels with cracked centers were observed on older branches and these may be the locations where radiocesium was absorbed and translocated internally. In addition, physical injury to the branches did not only occur in the lenticels. Using imaging plates, we detected intense imaging in areas that had been scarred by pruning. In these areas, the surface was rough, which made it easier for dust accumulation, which may contain radiocesium. Thus, it is quite possible that these areas served as points for radiocesium absorption and subsequent inward translocation.

It has been reported that the concentration of radiocesium in bark decreased from summer to winter and that this was related to the sloughing away of the epidermal layer of the bark as the tree trunks expanded (Takata et al. 2012f). In ongoing experiments using washed peach trees, we have confirmed that the bark concentration of radiocesium did not change substantially between the winter and spring of 2012 (Takata et al. 2012a). This suggests that radiocesium strongly bound to the bark itself did not translocate inward during this time period. However, the concentration of radiocesium in the wood decreased from winter to spring. Reduction in the

Table 14.3 Concentration of ^{134}Cs and ^{137}Cs in "Akatsuki" peach trees grown in Tokyo with and without washing (from Takata et al. 2012b and additional data)

		Radioisotopes(Bq/kg-dry weight)	
	Part	^{134}Cs	^{137}Cs
Washed	Shoot	16.3 ± 1.2	21.7 ± 1.3
	Leaf	76.4 ± 11.0	71.7 ± 11.2
	Pericarp	117.7 ± 12.0	95.9 ± 11.7
	Bark (3-year old branch)	759.8 ± 36.0	811.6 ± 36.8
Without washing	Shoot	22.9 ± 1.7	30.8 ± 2.9
	Leaf	96.0 ± 8.5	107.8 ± 10.7
	Pericarp	133.8 ± 18.5	139.7 ± 17.5
	Bark (3-year old branch)	1044.6 ± 45.8	1287.8 ± 36.8

radiocesium concentration of wood during the 2012 fruit-growth period appeared to be the result of not only dilution caused by the expansion of the wood but also the translocation of radiocesium to new plant parts. We are currently analyzing the results of these investigations (Takata et al. 2012a).

14.2.3 Translocation from New Above-Ground Plant Parts

Table 14.3 shows the radiocesium concentrations of the above-ground parts of washed and unwashed peach trees grown in Tokyo. The isotopic composition of radiocesium on the surface of the 3-year-old branches is unknown, but the concentration of radiocesium decreased with washing. Similarly, washing resulted in a decrease in the radiocesium concentration in the pericarps, leaves, and other new plant parts that did not exist at the time of the nuclear accident. A similar trial in Fukushima involving the measurement of radiocesium in washed and unwashed plant parts yielded similar results. It was demonstrated that irrespective of the air dose rate, it was possible to remove radiocesium from the tree surface by washing it with water. These results led to the following two speculations. First, readily mobile radiocesium continues to be present on the surface of old branches. Second, it is possible that this radiocesium was redeposited on leaves and fruits. In the case of this NPP accident, the fallout occurred only around the mid of March, 2011. However, it was possible that the radiocesium in soils or trees scattered around the fields and redeposited on leaves and fruits. It is well known that leaves take up ionic elements thorough their stomata (Eichert et al. 1998) and is reasonable to suppose that this is also the case for radiocesium. It is also possible that radiocesium was deposited by the wind and rain on the surfaces of new fruits, shoots, and leaves and was subsequently absorbed into these plant parts.

A comparison of the radiocesium concentrations in the pulp and pericarps of peaches produced in (Takata et al. 2012b) the year of the nuclear accident (2011) and 2012 (Takata et al. 2012a) showed that the ratio of the pericarp radiocesium concentration relative to the pulp radiocesium concentration was higher in 2011 than in 2012.

Fig. 14.5 Removing the outer layers of bark using high-pressure water sprays to peach (*upper*) and persimmon (*bottom*) trees. Photo: Mamoru Sato and Kazuhiro Abe (Fukushima Prefectural Fruit Research Center)

Based on these results, we suggest that greater quantities of radiocesium were deposited on the surfaces of fruits in 2011, thereby creating conditions where radiocesium could be more readily absorbed into the pericarp itself. Thus, it is possible that secondary deposition on new plant parts occurred for at least the first few months following the accident. A comparison of the radiocesium concentrations of the leaves growing on succulent shoots and the shoots of bearing branches (Takata et al. 2012b, 2013) showed that the radiocesium concentration in the leaves of vigorously growing succulent shoots was lower than that in the leaves of shoots on bearing branches, which ceased elongating early in the fruit growth period. There are two potential explanations for these results. First, it is possible that the redeposition of radiocesium on new leaves occurred up to the point when the shoots on the bearing branches stopped elongating (ostensibly in June). While this is difficult to verify, it is possible that the radiocesium existed as a readily soluble ion and that it was redeposited on leaves and fruits by rain. A second possibility is that, unlike potassium, radiocesium undergoes differential fractionation within peach trees, depending on factors such as the plant part age and growth conditions. Potassium and cesium appear to have similar behaviors in terms of root uptake (Ehlken and Kirchner 2002), but the two elements may be translocated differently in the above-ground plant parts. For example, it has been shown that cesium, unlike potassium, tends to accumulate in the older leaves in rice (Tsumura et al. 1984). Thus, with respect to the distribution of radiocesium, it was necessary to carefully examine the differences in redeposition and translocation.

In the winter of 2011, in addition to the stricter than usual enforcement of removing the outer layers of bark, farmers employed high-pressure water sprays to wash the surfaces of the peach trees in Fukushima Prefecture (Fig. 14.5). As described above, radiocesium attached to the surface can be removed by washing, which prevents it from acting as a contaminant source for redeposition onto leaves and fruits by the wind and rain. In this sense, removing the outer layer of bark and the high-pressure washing in Fukushima during the winter of 2011 were important for removing radiocesium from the tree surfaces and reducing the risk of redeposition on new plant parts in 2012.

14.3 Release of Radiocesium from Trees

In addition to the absorption of radiocesium by trees, we must also consider the release of radiocesium from trees. This included removal through the sloughing away of the bark, the harvesting of fruits, the falling of leaves, and release into the soil through the sloughing away or senescence of roots, as well as artificial removal by pruning. The decontamination of tree surfaces by high-pressure washing should also be included in the broader sense. To evaluate the quantity or proportion of radiocesium released or removed, it was first necessary to understand how much radiocesium was translocated to each part of the tree. We investigated the distribution of radiocesium within trees and determined various factors related to the release of radiocesium from trees based on these results.

14.3.1 Distribution of Radiocesium Within Peach Trees 5 Months After the Nuclear Accident

The proportion of radiocesium in fruits and leaves that were defoliated (harvested) before the winter were investigated in the experiments described below (Takata et al. 2012d). The experimental samples comprised three 5-year-old "Akatsuki" peach trees grown in an experimental field in the grounds of the University of Tokyo. Fruits were harvested on July 25, 2011, after reaching a harvestable state. The trees were subsequently excavated on August 9 and separated into leaves, shoots, 1–3-year-old branches, main trunks, rootstock, and roots. The roots were further separated according to diameter into fine roots of <2 mm, intermediate roots

Table 14.4 Concentration of ^{134}Cs and ^{137}Cs in "Akatsuki" peach tree grown in Tokyo (from Takata et al. 2012d)

Position	^{134}Cs Bq/kg-dry weight	^{137}Cs Bq/kg-dry weight
Bearing branch		
Shoot	12.6>[a]	15.4>
Leaf	42.7 ± 4.3	44.8 ± 5.0
Mature fruit		
Pericarp	44.2 ± 5.4	50.4 ± 6.0
Pulp	27.0 ± 4.4	39.3 ± 4.5
Stone with seed	6.3>	7.5 ± 2.4
3-Year-old branch		
Succulent shoot		
– Bark	11.6 ± 3.7	14.4>
– Wood	9.4>	10.4>
– Leaf	17.4 ± 3.5	16.3 ± 4.0
Bud(without washing)	333.8 ± 20.6	419.1 ± 23.6
1,2-Year-old branch		
– Bark	235.5 ± 9.6	284.2 ± 11.1
– Wood	13.1 ± 3.6	19.5 ± 4.5
3-Year-old branch		
– Bark	355.0 ± 13.0	434.6 ± 15.5
– Wood	11.5 ± 2.7	12.8 ± 3.0
Trunk		
Bark		
– Epidermis	245.5 ± 16.2	290.4 ± 18.3
– Directory under epidermis	6.5 ± 1.3	4.7 ± 1.5
– Cortex	5.8 ± 1.8	10.1 ± 1.9
– Secondary phloem with cambium	19.4 ± 3.2	28.8 ± 3.9
Wood		
– Sapwood	19.7 ± 3.3	19.4 ± 3.8
– Heartwood	7.0>	8.8>

[a]Under detection limit. Value means detection limit

of 2–10 mm, and large roots of 10–50 mm. In the trees used in this experiment, the 3-year-old branches corresponded to primary scaffold limbs, whereas the 1- and 2-year-old branches corresponded to secondary scaffold limbs and lateral branches. The shoots and 1–3-year-old branches were further separated into bark and wood. The bark of the main trunk was separated into four parts: the epidermis, the layer immediately below the epidermis, the cortex, and the secondary phloem and cambium. The wood was separated into two parts: the sapwood containing xylem and the heartwood with closed-off xylem. We also calculated the total radiocesium adsorption (Bq equivalent) for each plant part by multiplying the radiocesium concentration by the total dry weight of each plant part.

The radiocesium concentrations of the above-ground tree parts are presented in Table 14.4. In the old branches, the radiocesium concentration was substantially higher in the bark than in the wood. The radiocesium concentration was lower in the

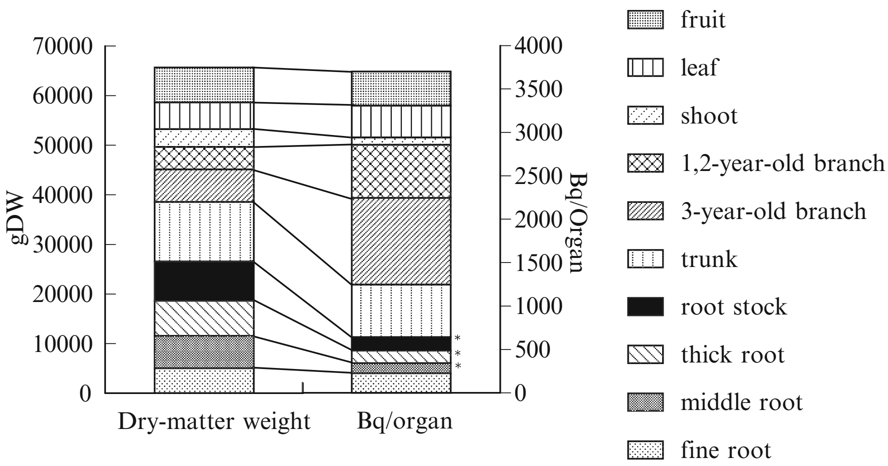

Fig. 14.6 Dry matter weight and radiocesium content for every organ in “Akatsuki” peach tree. *Under detection limit. Value means detection limit × dry weight. From Takata et al. (2012d)

main trunks than in the 3-year-old branches. Compared with the main trunks, which grew perpendicular to the ground, the 1–3-year-old branches, corresponding to the primary and secondary scaffold limbs in the sample trees, grew in a more exposed and open manner. It is possible that this difference in geometry allowed the deposited radiocesium to accumulate more readily on the surface of the 1–3-year-old branches compared with the surface of the main trunks, which accounts for the differences in the radiocesium concentration. In the bark of the main trunk, the radiocesium concentration was higher in the outermost epidermal layer than in any other part of the bark. In fact, the radiocesium concentrations in the layer immediately below the epidermis and in the cortex were only approximately 2% of that in the epidermis. It appeared that an extremely high proportion of the radiocesium was absorbed by the 0.09–0.20 mm thick epidermis of the main trunk. The radiocesium concentration in the secondary phloem was approximately three times that in the cortex. It is possible that this was because of absorption through the bark and translocation from leaves and other plant parts.

We calculated the total absorption of radiocesium by the different plant parts by multiplying the radiocesium concentration with the total dry weight for each plant part (Fig. 14.6). Because the radiocesium measured in all but the fine roots was below the detection limit, we tentatively assigned the detection limit to these roots. Because the detection limit was used as a proxy to calculate the total radiocesium content of the roots, the estimate represented the maximum potential concentration. Even with this upper estimate, the radiocesium content of the roots was lower than that of the above-ground parts. It was evident that the majority of the radiocesium in the peach trees was present in the above-ground parts at the time of the harvest. The radiocesium content of fruits and leaves that would not be retained until the next year accounted for approximately 20% of the total radiocesium content of the trees.

We conducted a similar destructive sampling in Fukushima in January 2012, and we are currently analyzing the results (data unpublished). To summarize the results, despite differences in the magnitude of concentration and differences related to the presence or absence of leaves and fruits, no difference in the relative distribution of radiocesium within the trees were observed between locations. We are also in the process of determining whether it will be possible to predict the radiocesium concentration in fruits using these results.

14.3.2 Release of Radiocesium from Above-Ground Tree Parts

Approximately 80% of the radiocesium that was absorbed in the 5 months between the nuclear accident and the destructive sampling was expected to be retained until the next year. It has been demonstrated that the radiocesium concentration in leaves tends to decrease throughout the fruit growth period and it falls dramatically during the leaf fall period (Takata et al. 2012f). We must also consider the re-initiation of shoot elongation in the fall and the amount of radiocesium that is newly absorbed by the trees. Furthermore, with regard to the radiocesium content of branches, we must consider the removal by pruning. A study comparing peach tree pruning practices in Japan (Takata et al. 2008) showed that under conventional cultivation practices, 56.6% of peach tree branches were removed depending on the length during the winter pruning period. It is not possible to accurately calculate the mass of material removed because the above information was expressed as a ratio of the length; however, we estimated that approximately 30% of the total radiocesium content (Bq equivalent) of trees was removed by branch pruning. Given that the radiocesium concentration was higher in the primary and secondary scaffold limbs, which grow more horizontally than the vertically growing main trunks (Takata et al. 2012d), the active removal of secondary scaffold limbs during the winter pruning period and nurturing the succulent shoots that emerged after the nuclear accident as bearing branches in subsequent years or otherwise renewing the lateral branches, may be effective from the viewpoint of tree decontamination.

Calculating the amount of tree material lost or removed by pruning, wood expansion, and other methods, makes it possible to estimate the amount of radiocesium released by the trees over time. Examining the radiocesium concentration in peach trees in 2012 showed that in contrast to 2011, only a small percentage of the radiocesium had translocated from the old plant parts to new plant parts. Furthermore, the radiocesium concentration in the bark did not change over the fruit growth period, whereas the concentration in the wood decreased dramatically over the same time period (Takata et al. 2012f). The different distributions of radiocesium in the new plant parts in the 2 years may reflect the direct absorption of high quantities of radiocesium from the surface fruits and leaves in 2011.

14.3.3 The Release of Radiocesium into the Soil Through the Roots

The peak growth of peach roots occurs in spring and fall. At the time of harvest, the radiocesium concentration of roots was low. Given that radiocesium was only present in the surface layer of the soil at the time, we assume that it would have been unlikely for deep-rooted tree species to take up significant quantities of radiocesium from the soil. Furthermore, because spring root growth utilizes nutrients stored during the previous year, we believe that there was little translocation of radiocesium from the above-ground to below-ground plant parts. However, it is possible that the radiocesium accumulated in the trees was translocated to below-ground parts along with carbohydrates assimilated in the above-ground plant parts to form new roots during the fall growth. Not all the new roots of peach trees lignify and become permanent parts of the tree. A large proportion of roots senesce and are sloughed away. As roots senesce and decompose, the radiocesium contained therein is released into the soil. Thus, it is necessary to consider the transfer of radiocesium from old to new roots and the translocation of radiocesium from above-ground plant parts to below-ground plant parts.

In January 2012, we excavated peach trees grown in high dosages areas and conducted a cultivation experiment. Roots of <2 mm in diameter were removed and the remaining root mass was washed. The trees were then replanted in contaminant-free soil purchased before the nuclear accident (Fig. 14.7). Although the results of this experiment are still under analysis, we have made the following observations related to the roots. The radiocesium concentration was higher in the new peach roots than the fruits and other new above-ground plant parts. Because the trees were grown in an environment where there was no soil-to-plant transfer, we can conclude that the radiocesium contained in the new roots was translocated from older plant parts. Furthermore, radiocesium was detected in the soil that was still attached to the roots. Radiocesium may be transferred to the soil through the senescence and sloughing away of new roots, or the detachment and sloughing away of the bark of older roots due to root expansion. Thus, it is possible that radiocesium was transferred to the deeper soil in loc.l orchards through the roots. However, the total mass of new roots was lower than that of the new above-ground plant parts and thus, the total amount of radiocesium translocated to the new roots was relatively small. Therefore, it is necessary to look carefully at the quantity of radiocesium that is actually translocated. Consistent with these results, an investigation of container-grown grapes (Zehnder et al. 1995) demonstrated that radiocesium applied to leaves translocated rapidly to the below-ground parts. Thus, when considering fruit trees and the orchards where they are grown, it is important to include the uptake of radiocesium from the soil and the environment and the release of radiocesium into the soil.

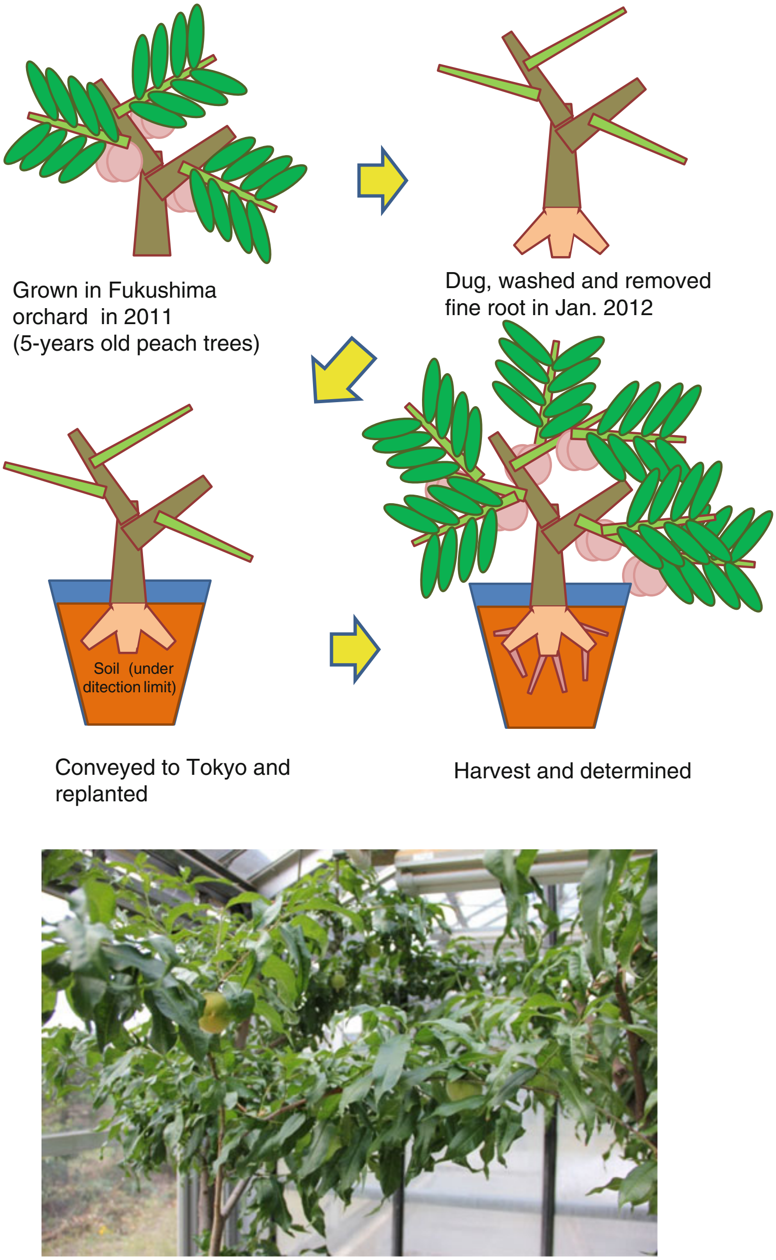

Fig. 14.7 Experimental illustration (*upper*) and photo (*bottom*) in translocation from old organ to new organ in "Akatsuki" peach trees

14.4 Conclusions

Compared with other crops, there have been relatively few studies related to the migration of radiocesium in fruit trees. The same is true of experiments related to the cultivation of fruit trees. Limiting factors include the size of trees, the effort required to conduct such experiments, the duration of an experiment, considering that it may take several years for trees to become harvestable, and the difficulty in repeating experiments because of such time factors. Potted test results were sometimes different from those of orchard-based experiments. Unlike annual crops, fruit trees have well-established root systems; thus, the actual analysis of responses to the nuclear accident, including the felling or replanting of trees, were hindered by significant obstacles, particularly the loss of production due to the non-harvestable period following replanting. If the soil has to be decontaminated in conjunction with replanting, it is necessary to consider the amount of labor and effort required for such remediation, as well as the disposal of the contaminated soil and tree material. In the case of peach trees, it may also be unwise to replant all the trees at one time given the potential problem of replant soil sickness. Thus, for fruit trees that are perennial, it is necessary to clarify where the radiocesium absorbed in 2011 accumulated in the trees and how this radiocesium was subsequently translocated. However, this analysis also requires a certain amount of time. If the trees are not to be replanted, there is an even greater urgency for this clarification. Although radiocesium is bound strongly by the soil, it does not mean that it is not taken up at all by the roots. Furthermore, it is possible that the radiocesium released by roots, particularly that released in the vicinity of the rhizosphere of fruit trees, is reabsorbed by the trees. If no remedial action is taken, it is expected that the magnitude of radiocesium migration in the soil and the contribution of soil radiocesium to overall tree contamination will increase gradually with time.

In fruit trees and other plant species, among the potential sources of radiocesium contamination of new plant parts include soil radiocesium, radiocesium accumulated within the plants in the year of the nuclear accident, radiocesium resuspended in the surface layer of the soil, and radiocesium redeposited on new plant parts after its surface adhesion to the branches. For plants grown in contaminated soil or contaminated environments, it is difficult to determine the contribution of each of the above-mentioned sources to the radiocesium detected in new plant parts. We are currently conducting experiments using various approaches including transferring trees that were previously grown under contaminated to non-contaminated environments. We plan to report the results of these experiments in the near future.

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Chapter 15
Mushrooms: Radioactive Contamination of Widespread Mushrooms in Japan

Toshihiro Yamada

Abstract High concentrations of radionuclide contamination of wild mushrooms occurred near the Fukushima Daiichi nuclear power plant and along the path of radioactive plumes. This is a serious problem for the forest ecosystem and commercial wild mushroom production. Radioactivity in wild mushrooms collected from low-level contamination forest areas throughout Japan was measured approximately 6 months after the accident. In general, the radioactivity in mushrooms did not exceed those in the neighboring forest litter. However, further accumulation of ^{137}Cs is expected in mushrooms; therefore, continuous monitoring is necessary even in low-contamination areas. We also found that residual ^{137}Cs radioactivity due to nuclear weapons tests, mainly in 1950s, besides fallout from the Fukushima nuclear accident still remained in soil and was accumulated by mushrooms.

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

Abbreviations

Cs	Cesium
DW	Dry weight
FW	Fresh weight

T. Yamada (✉)
The University of Tokyo Chiba Forest, Graduate School of Agricultural and Life Sciences, The University of Tokyo, 770 Amatsu, Kamogawa, Chiba 299-5503, Japan
e-mail: yamari@uf.a.u-tokyo.ac.jp

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I	Iodine
K	Potassium
NPP	Nuclear power plant
NWT	Nuclear weapons test
TF	Transfer factor

15.1 Introduction

Fungi are one of the major and important components of forest ecosystems. 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, including crops, domestic animals, and wood products, through its circulation in the forest ecosystem. From 1950s to 1960s, a high uptake of ^{137}Cs, derived from atmospheric nuclear weapons tests (NWT), by mushrooms was also noticed in Japan (Muramatsu and Yoshida 1997; Sugiyama et al. 1994; Yoshida and Muramatsu 1996).

Radionuclides released by the accident at the Fukushima Daiichi nuclear power plant (F1-NPP) deposited over a wide area of East Japan. Radiocesium concentrations exceeding the safety threshold were detected in mushrooms hundreds of kilometers from F1-NPP. The Japanese interim limit for imported foods was 370 Bq/kg fresh weight (FW) of radiocesium after the Chernobyl nuclear accident in 1986 (Sugiyama et al. 1994). The interim limit after the Fukushima nuclear accident was set to 500 Bq/kg FW. However, a stricter limit of 100 Bq/kg FW was introduced in April 2012. Therefore, reducing the radiocesium contamination of food has become more important.

Mushrooms have been reported to accumulate radiocesium (Byrne 1988; Kammerer et al. 1994; Mascanzoni 1987; Muramatsu et al. 1991; Sugiyama et al. 1990, 1994). For example, the transfer factors (TF) for radiocesium in mushrooms were reported to be 2.6–21 in several culture tests (Muramatsu et al. 1991; Ban-nai et al. 1994). However, the radiocesium activity ratio in mushrooms relative to the soil in a field study were rather low and the ratio was often <1 (Heinrich 1992). Different fungal species also exhibit widely varying degrees of ^{137}Cs contamination; however, saprotrophic mushrooms tend to have lower TF of ^{137}Cs than the symbiotic mycorrhizal fungi (Heinrich 1992; Sugiyama et al. 1993). Cesium shows a similar behavior to potassium. TF of ^{137}Cs and potassium in fungi is very high compared with those in vegetables (Miyake et al. 2008; Sugiyama et al. 1990). Not all mushrooms accumulate cesium at the same levels as potassium. For example, TF of cesium was lower than that of potassium in *Lentinula edodes* (Sugiyama et al. 1990). A reason for radiocesium contamination to be high in mushrooms appears to be because of potassium richness (Seeger 1978). However, fungi also absorb cesium more specifically compared with potassium (Kuwahara et al. 1998), particularly radiocesium (Tsukada et al. 1998). Further research is necessary to reveal the absorption and accumulation processes.

The radioactivity in mushrooms is an appropriate index for determining radioactive contamination of forest ecosystems. Thus, data from high and low-contamination areas are required to understand the overall situation. The University of Tokyo has seven research forests, 250–660 km from F1-NPP. Inspections of radioactivity in food conducted by local governments and survey on low- contamination areas in the University of Tokyo Forests revealed the state of radioactive contamination of mushrooms.

15.2 High-Level Contamination

A radioactive plume of short distance but high contamination level travelled northwest from F1-NPP (Fig. 15.1). Two major long distance plumes south and southwest bound were also observed. Thus, radioactive nuclides were dispersed throughout a wide area of East Japan, and high-level contamination was detected in wild mushrooms, particularly those in the vicinity of F1-NPP and along the plume paths.

Similar to mushrooms cultivated on wood logs, such as shiitake mushroom (*L. edodes*), the shipment of wild mushrooms was also halted in 41 out of a total of 59 cities, towns, and villages on July 27, 2012, in Fukushima Prefecture (area 13,782 km^2) (http://www.mhlw.go.jp/stf/houdou/2r9852000002gufy-att/2r9852000002gult.pdf). Outside Fukushima Prefecture, wild mushrooms and log-cultivated mushroom shipments are restricted in some areas, particularly those along the plume paths.

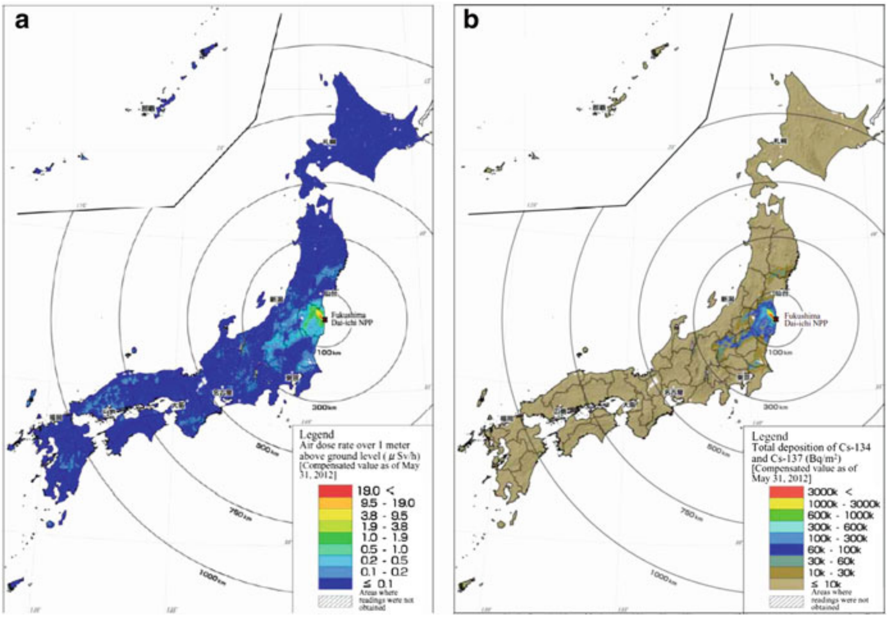

Fig. 15.1 Distribution map of γ-ray air dose rate (μSv/h) 1 m above ground level (**a**) and total deposition of ^{134}Cs and ^{137}Cs (Bq/m^2) on the ground surface (**b**). Reproduced from the results of airborne monitoring surveys conducted by Ministry of Education, Culture, Sports, Science and Technology, Japan, http://radioactivity.mext.go.jp/en/contents/6000/5188/24/203_e_0727_14.pdf

Table 15.1 Radionuclide contamination of mushrooms and lichens immediately after the Fukushima nuclear accident (modified from Kakishima et al. 2011)

	Moisture content (%)[a]	^{131}I (Bq/kg FW)	^{137}Cs (Bq/kg FW)	^{134}Cs (Bq/kg FW)
Mushrooms				
Russula sp. (M)	94	ND	130	ND
Astraeus hygrometricus (M)	86	928	1,683	1,512
Morchella esculenta (M/S)	74	36	78	ND
Psathyrella sp.	45	67	529	530
Psathyrella sp.	8	184	255	278
Schizophyllum commune (S)	73	2,301	5,719	5,506
Cryptoporus volvatus (S)	–	ND	ND	ND
Daedaleopsis tricolor (S)	91	1,631	868	812
Lichens				
Dirinaria applanata	7	3,438	3,558	3,219
Phaeophyscia sp.	5	8,436	12,641	12,413

(*M*) mycorrhizal fungus, (*S*) saprophyte
[a]Moisture content (%) = 100 × (fresh weight − dry weight)/fresh weight

Shortly after the accident, extremely high concentrations of radiocesium were detected in wild mushrooms collected on April 26, 2011, from the University of Tsukuba campus (Table 15.1, Kakishima et al. 2011), which is 170 km from F1-NPP. The University of Tsukuba is located in an area where the radionuclide deposition is rather low. However, a high concentration of radionuclides, including ^{131}I, ^{134}Cs, and ^{137}Cs, was detected in three mushroom species. Of them, two species (*Daedaleopsis tricolor* and *Schizophyllum commune*) were saprophytes that grow on dead trees. This high contamination level appeared to be mainly due to the direct deposition on long-lived fruiting bodies. Lichens showed high radioactivity. Direct deposition may have contributed to the majority of the contamination considering the lifespan of lichens. ^{134}Cs was not detected in 2 species of mushrooms (*Morchella esculenta* and *Russula* sp.), which had relatively lower radioactivity. This suggests that the contamination was derived from NWT or the Chernobyl accident. This assumption has been discussed later.

The radioactivity of wild mushrooms was investigated as a part of a food survey in Fukushima Prefecture (http://www.new-fukushima.jp/monitoring/result.php). A high level of radioactive contamination was observed along the plume paths. For example, the maximum concentration of radiocesium (28,000 Bq/kg FW) was detected in *Lactarius volemus* in September 2011, approximately 70 km from F1-NPP. However, a radioactivity concentration of 40 Bq/kg FW was also recorded for the same mushroom species at the same time in the same town. Thus, the radioactivity varied considerably. In the autumn of 2011, high radiocesium concentrations were recorded in the mycorrhizal fungi *Lactarius lividatus* (maximum = 19,900 Bq/kg FW, approximately 20 km) and *Tricholoma matsutake* (maximum = 3,300 Bq/kg FW, approximately 60 km) apart from *L. volemus* as well as in the saprophytic fungus *Grifola frondosa* (maximum = 2,800 Bq/kg FW, approximately 20–30 km). Large differences in radioactivities were observed depending on the types of mushrooms.

However, apart from the immediate vicinity of F1-NPP and the plume paths, the contamination concentrations in wild mushrooms in the autumn of 2011 were relatively low in Fukushima Prefecture. For example, radiocesium was undetectable in the following mushrooms collected from August to October, 2011 30–40 km from NPP: *Entoloma sarcopum* (Mycorrhizal fungi), *Lyophyllum fumosum* (M), *Ramaria botrytis* (M), and *Lyophyllum decastes* (Saprophyte). Rather low concentrations of radiocesium (19–26 Bq/kg FW) were detected in *L. volemus* (M), *Leucopaxillus giganteus* (S), *Laetiporus sulphureus* (S), and *Armillaria mellea* (S).

The earliest measurement of shiitake mushrooms in open-field log cultivation was conducted on April 3, 2011, which detected ^{131}I at 3,100 Bq/kg, ^{134}Cs at 450 Bq/kg, and ^{137}Cs at 440 Bq/kg FW. The highest concentration was recorded on April 10, i.e., ^{131}I at 12,000 Bq/kg, ^{134}Cs at 6,400 Bq/kg, and ^{137}Cs at 6,600 Bq/kg FW. Individual fruiting bodies of shiitake mushrooms were usually harvested within 1 week of their emergence from the log so that there was no possibility of direct deposition on the fruiting bodies. Rapid absorption of radionuclides from the logs was a possibility.

15.3 Low-Level Contamination

The University of Tokyo Forests comprise seven research forests located in East Japan. Mushrooms appeared in the autumn of 2011 and their possible substrates, i.e., the O horizon (organic layer, which included the litter layer), the A horizon (mineral layer and accumulated organic matter), and the Ch horizon (mineral layer with organic matter, which is little affected by pedogenic processes) of the soil, or mushroom logs were collected from six research forests listed below (Figs. 15.2 and 15.3).

UTCF: The University of Tokyo Chichibu Forest, Saitama Prefecture, 250 km from the F1-NPP
UTCBF: The University of Tokyo Chiba Forest, Chiba Prefecture, 260 km
FTRI: Forest Therapy Research Institute, Yamanashi Prefecture, 300 km
ARI: Arboricultural Research Institute, Shizuoka Prefecture, 360 km
ERI: Ecohydrology Research Institute, Aichi Prefecture, 420 km
UTHF: The University of Tokyo Hokkaido Forest, Hokkaido Prefecture, 660 km

Samples were collected from October to November 2011, which included symbiotic mycorrhizal fungi and saprophytes, and the concentrations of ^{134}Cs and ^{137}Cs were determined.

Radiocesium contamination of mushrooms was observed in all the University of Tokyo forests examined, except in ERI (Aichi) (Table 15.2). Although no radiocesium was detected in the mushrooms collected from ERI, a very small amount of ^{137}Cs was detected in the A horizon of soil from ERI. This contamination may be due to atmospheric NWTs. Radiocesium was detected in many mushroom and soil samples from UTCF (Chichibu), UTCBF (Chiba), and FTRI (Fuji).

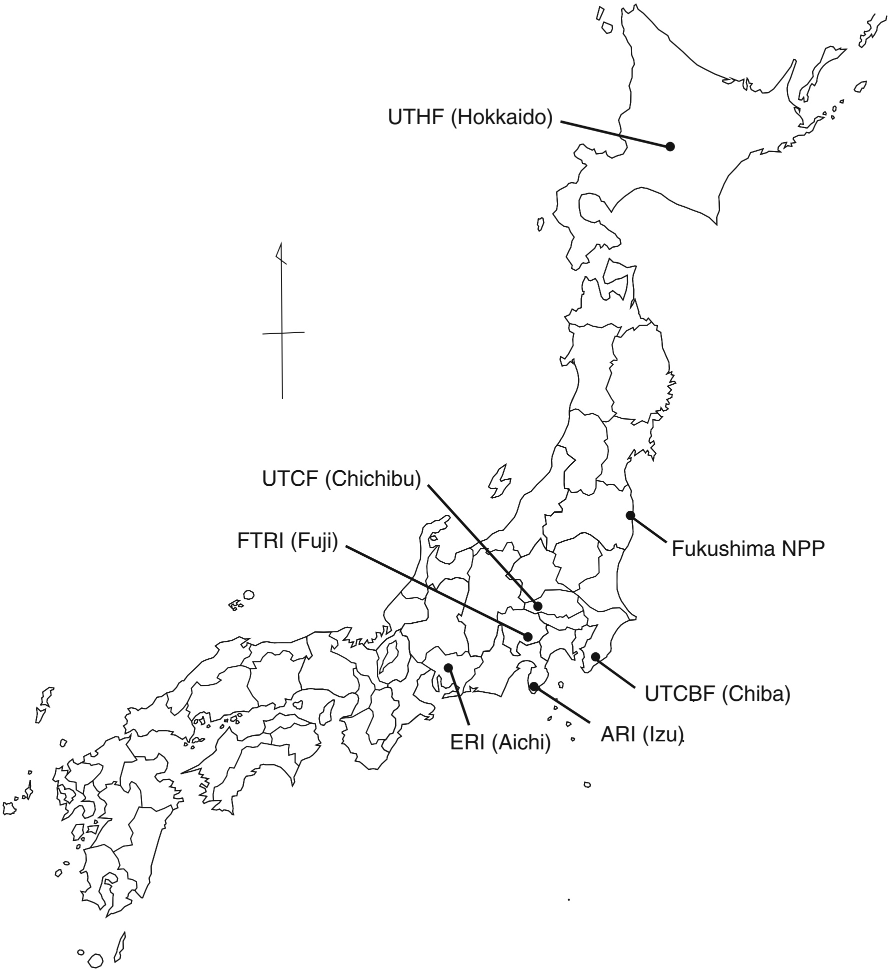

Fig. 15.2 Locations of the University of Tokyo Forests from where samples were obtained. *F1-NPP* Fukushima Daiichi nuclear power plant, *UTCF* The University of Tokyo Chichibu Forest (Chichibu), *UTCBF* The University of Tokyo Chiba Forest (Chiba), *FTRI* Forest Therapy Research Institute (Fuji), *ARI* Arboricultural Research Institute (Izu), *ERI* Ecohydrology Research Institute (Aichi), *UTHF* The University of Tokyo Hokkaido Forest (Hokkaido)

UTCF is located in a moderately contaminated area where the gamma ray dose rate exceeded 0.1 μSv/h (Fig. 15.1) at some sampling sites (Table 15.3) because it is at the tip of the southwest bound plume path, despite the long distance (250 km) from F1-NPP. The dose rates in other research forests were <0.06 μSv/h, which was within the normal range before the accident. The radioactive contamination level was low in UTCBF (Chiba), which is located outside the plume path, although its distance from F1-NPP was similar to UTCF. The contributions due to fallout from F1-NPP alone were evaluated by subtracting the natural background dose rates

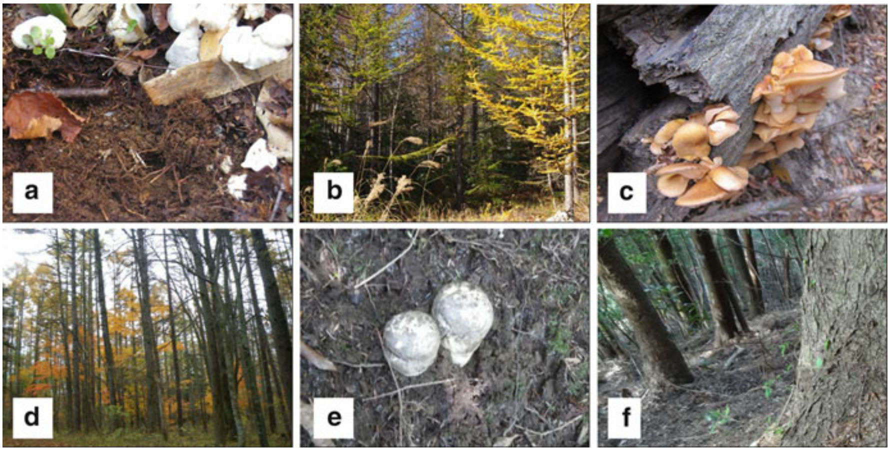

Fig. 15.3 Examples of collected mushrooms and forest where mushrooms were collected. (**a**) *Lyophyllum connatum* mushrooms, O and A horizon of soil under the mushrooms in UTHF; (**b**) deciduous forest where *L. connatum* mushrooms were collected; (**c**) *Panellus serotinus* mushrooms and wood in UTCF; (**d**) deciduous forest in FTRI where mushrooms of *Suillus grevillei* and *Pleurotus ostreatus* were collected; (**e**) *Catathelasma imperiale* mushrooms in UTCBF; (**f**) evergreen forest where *C. imperiale* mushrooms were collected. (**a**, **b**) Photo by K. Iguchi, UTHF; (**c**) photo by K. Takatoku, UTCF; (**d**) photo by H. Saito, FTRI; (**e**) photo by T. Tsukagoshi, UTCBF

Table 15.2 Radiocesium contamination of mushrooms in the University of Tokyo Forests

Location	Site number and corresponding mushroom	Concentration[a] [Bq/kg DW (Bq/kg FW)]	
		^{134}Cs	^{137}Cs
UTCF (Chichibu)	1 *Bondarzewia berkeleyi* (S)	102 (21)	154 (32)
	2 *Tricholoma saponaceum* (M)	162 (35)	233 (50)
	3 *Panellus serotinus* (S)	492 (68)	706 (97)
	4 *Trametes versicolor* (S)	67 (58)	89 (77)
UTCBF (Chiba)	1 *Catathelasma imperiale* (M)	ND	77 (16)
	2 *Catathelasma imperiale* (M)	ND	ND
FTRI (Fuji)	1 *Suillus grevillei* (M)	137 (6)	714 (34)
	2 *Suillus grevillei* (M)	ND	680 (32)
	Pholiota lubrica (S/M)	1,070 (98)	1,630 (150)
	3 *Pholiota lubrica* (S/M)	1,840 (108)	2,440 (143)
	4 *Pleurotus ostreatus* (S)	ND	ND
ARI (Izu)	1 *Lentinula edodes* (S)	53 (3)	78 (4)
	2 *Lentinula edodes* (S)	ND	68 (7)
ERI (Aichi)	1 *Macrolepiota procera* (S)	ND	ND
	2 *Armillaria mellea* (S)	ND	ND
UTHF (Hokkaido)	1 *Lyophyllum connatum* (M)	ND	62 (6)
	2 *Suillus grevillei* (M)	ND	31 (1)

(*M*) mycorrhizal fungus, (*S*) saprophyte

[a]ND: not detected = concentration less than the detection limit

Table 15.3 Radiation dose rate at the mushroom collection sites

Location and month of measurement	Site number and corresponding mushroom	Dose rate (μSv/h)	
		10 cm above ground	1 m above ground
UTCF (Chichibu) February 2012	1 *Bondarzewia berkeleyi*	0.109	0.082
	2 *Tricholoma saponaceum*	0.064	0.050
	3 *Panellus serotinus*	0.110	0.114
	4 *Trametes versicolor*	0.083	0.090
UTCBF (Chiba) January to April 2012	1 *Catathelasma imperial*	0.042	0.033
	2 *Catathelasma imperial*	0.040	0.032
FTRI (Fuji) May 2012	1 *Suillus grevillei*	0.022	0.024
	2 *Pleurotus ostreatus*	0.030	0.019
	3 *Suillus grevillei*	0.028	0.020
	3 *Pholiota lubrica*	0.022	0.029
	4 *Pholiota lubrica*	0.031	0.022
UTHF (Hokkaido) June 2012	1 *Lyophyllum connatum*	0.061	0.053
	2 *Suillus grevillei*	0.047	0.035

(Minato 2011). The dose rate because of the Fukushima accident was approximately 50 nGy/h (0.05 μSv/h equivalent dose rate of radiocesium) in UTCF and <25 nGy/h in UTCBF, FTRI, and ARI (Izu).

The radiocesium concentrations in *Panellus serotinus* and *Trametes versicolor* samples collected from UTCF exceeded the present limit of 100 Bq/kg FW (Table 15.2). *Pholiota lubrica* from FTRI had a radioactivity concentration of >200 Bq/kg FW, despite low contamination in the soil. Radiocesium accumulated in the litter layer at UTCBF and FTRI, despite the normal dose rate. Both ^{134}Cs and ^{137}Cs were detected in *L. edodes* from ARI, indicating that the radionuclides from F1-NPP travelled 360 km. No ^{134}Cs was detected in the mushroom and soil samples from UTHF (Hokkaido) and ERI, confirming that these forests were not contaminated by the Fukushima accident.

15.4 Relationship Between Mushroom Contamination and Radiocesium Concentration in the Fungal Substrates

Cesium is strongly adsorbed to soil, particularly clay. Forest soil is abundant in organic compounds; therefore, less radiocesium should be adsorbed. However, considerable proportion of ^{137}Cs in forest soil is retained by the fungal hyphae, and fungi are considered to prevent the elimination of cesium 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 have been

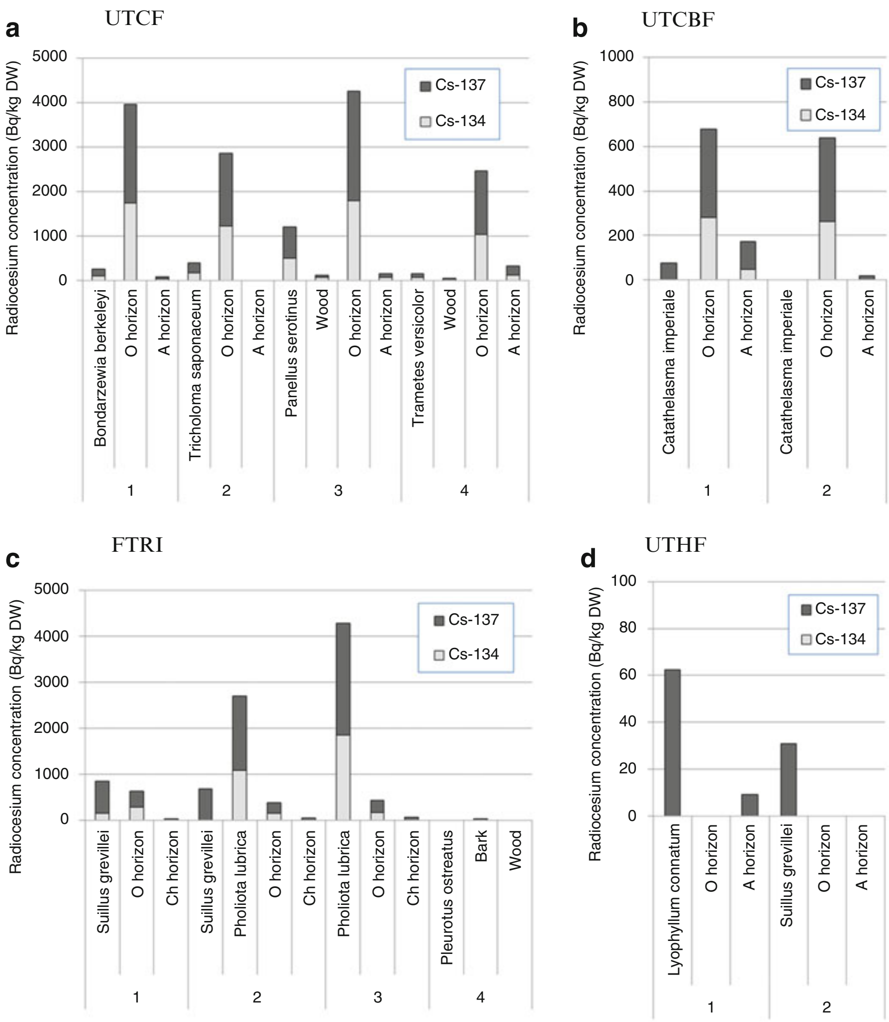

Fig. 15.4 Association of radiocesium contamination in mushrooms with that in their possible substrates. Note the differences in scale

reported where the ^{137}Cs radioactivity in mushrooms persisted for a long period in forests and was transferred to animals, whereas that in plants had short ecological half-lives (e.g., 3–3.5 years) (Fielitz et al. 2009; Kiefer et al. 1996; Zibold et al. 2001).

Because radiocesium contamination was higher in the O horizon than that in the A/Ch horizon of soil in the autumn of 2011, 6 months after the Fukushima accident (Fig. 15.4), most of the radiocesium had not migrated from the litter layer. Moreover, most mushrooms did not accumulate radiocesium compared with the O horizon, except for *P. lubrica* and *Suillus grevillei*. This suggests that the contamination had not reached the soil layer where most fungal hyphae are distributed. Mushrooms incorporate radiocesium as litter decomposition advances and cesium migrates into the soil. Thus, the radiocesium concentration in European mushrooms increased for a few years after the Chernobyl accident (Borio et al. 1991; Smith and Beresford 2005). Radioactivity was already incorporated into mushrooms in UTCF. Given the high radiocesium concentration in the O horizon, the radioactivity in mushrooms will increase greatly in UTCF and in areas with a normal dose rate. Therefore, it is necessary to monitor mushroom radioactivity in low-contamination areas as well as the immediate vicinity of F1-NPP.

^{137}Cs concentration was much higher in *P. lubrica* and *S. grevillei* compared with the O horizon and Ch horizon in FTRI. However, the causes of this may differ between the two fungal species. Radionuclides from the Fukushima accident caused the contamination of *P. lubrica*, whereas the contamination of *S. grevillei* appeared to be due to NWT or the Chernobyl accident. High concentrations of radioactivity incorporation may be induced by widespread distribution of mycelia near the litter surface. 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). ^{137}Cs is retained for a long time in the O horizon, which includes the litter layer of forest soil. A part of ^{137}Cs migrates very slowly into the A horizon (Kammerer et al. 1994; Pietrzak-Flis et al. 1996; Rühm et al. 1998). Thus, the changes are expected to be different between *P. lubrica* and other fungal species.

15.5 Radioactive Contamination due to Nuclear Weapons Tests or the Chernobyl Accident

Many atmospheric NWT were conducted up to 1980, which produced large amounts of radioactive fallout. Peak contamination in Japanese soils and crops was detected in 1963 (Komamura et al. 2006), after which the residual concentration of radioactive cesium decreased gradually. Chernobyl radionuclide plumes reached Japan in 1986; however, these depositions were temporary and less abundant.

To evaluate the contribution of NWT and the Chernobyl accident to the radioactive contamination of mushrooms and to distinguish it from that of the Fukushima accident, we calculated the ratio of ^{137}Cs derived from NWT or the Chernobyl accident to that from the Fukushima accident based on the ^{134}Cs/^{137}Cs ratio of the Fukushima derivatives. The half-lives of ^{134}Cs and ^{137}Cs are 2.07 and 30.1 years, respectively. We did not need to consider any evident decay of ^{137}Cs in our study. The residual ^{134}Cs was considered to decrease to 100/129 of its initial value. Estimates on the contribution of NWT or the Chernobyl accident and the Fukushima accident in ^{137}Cs contamination are shown in Fig. 15.5.

The contribution of NWT or the Chernobyl accident was high in several fungi such as *S. grevillei* and *P. lubrica* (Fig. 15.5). ^{137}Cs was detected in mushrooms collected from UTHF where no contamination due to the Fukushima accident was confirmed. In the soil, there was a higher contribution of NWT and the Chernobyl accident in the A or Ch horizon than the O horizon. No ^{137}Cs was detected in the

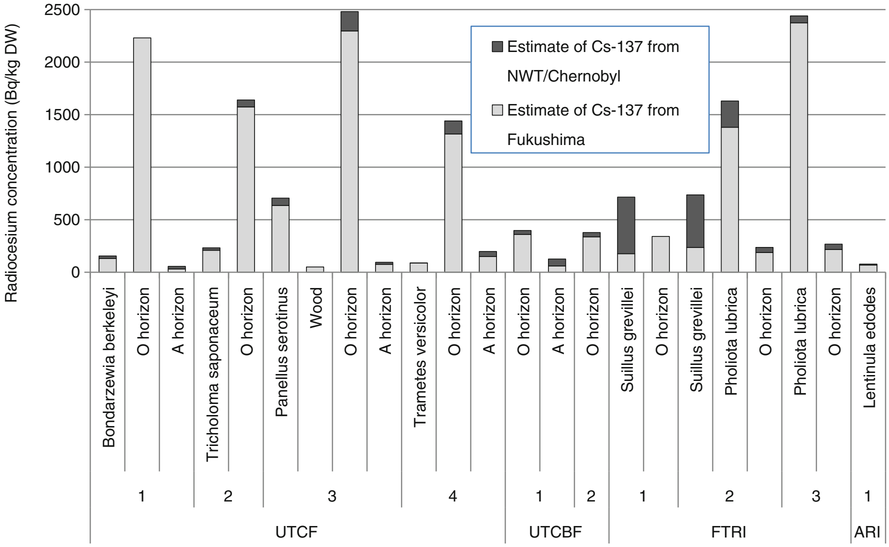

Fig. 15.5 Contribution of nuclear weapons tests or the Chernobyl accident to the radiocesium contamination of mushrooms

O horizon, despite its detection in the lower A horizon in some samples from ERI and UTHF. These results suggested that NWT- and Chernobyl-derived ^{137}Cs had already migrated from the O horizon to the A or Ch horizon and were being incorporated into the fungi.

Extremely low radiocesium activity was detected in A/Ch horizon at UTHF and FTRI, whereas the presence of ^{137}Cs in *L. connatum* and *S. grevillei* indicated ^{137}Cs retention in fungal hyphae for several decades. In particular, *S. grevillei* samples from FTRI contained >500 Bq/kg DW of ^{137}Cs derived from NWT or Chernobyl accident. Similarly, *P. lubrica* was calculated to contain 66–250 Bq/kg DW. Sugiyama et al. (2000) reported the presence of ^{137}Cs in *P. lubrica* (41 Bq/kg FW) and *S. grevillei* (91 Bq/kg FW) around Mt. Fuji in 1996. Both fungal species can be characterized by their ability to retain cesium.

The widespread distribution of radiocesium contamination before the Fukushima accident was reconfirmed by the radioactivity analysis of mushrooms, including those from low-contamination areas. This originated from the global fallout due to NWT, which peaked in 1963, and the Chernobyl accident in 1986, and is still being accumulated in the ground surface layers (Yoshida and Muramatsu 1994) almost 50 and 25 years after deposition, respectively. Takenaka et al. (1998) reported that Japanese soil was contaminated with ^{137}Cs due to NWT fallout at concentrations of 100 Bq/kg DW. Similar results were obtained in this and other studies. NWT affected the wild mushrooms in Japan, more than the Chernobyl accident. The contribution of Chernobyl accident was estimated to be in the range of 7–60% and 10–30% on an average (Igarashi and Tomiyama 1990; Muramatsu et al. 1991; Shimizu et al. 1997; Yoshida and Muramatsu 1994; Yoshida et al. 1994). Because approximately 50 years have passed since NWT, even if biological elimination from the ecosystem can be ignored, the radioactivity attributable to ^{137}Cs deposition 50 years ago was thrice as high as the present levels.

15.6 Conclusion and Future Perspectives

In addition to NWT and the Chernobyl accident, there has been serious radionuclide fallout in Japan due to the Fukushima accident. The radioactivity in forest ecosystems has been circulating. It is important to ensure the safety of forest products, such as mushrooms, game, and charcoal, derived from low and moderate-contamination areas. It is necessary to understand the behavior of radiocesium in mushrooms and their substrates as a part of forest ecosystems. Dose rate is a useful index, but it is insufficient for completely understanding mushroom contamination. It is necessary to distinguish the persistence of radiocesium due to NWT or the Chernobyl accident from the additional radiocesium attributable to the Fukushima accident, which is being absorbed into mushrooms and incorporated into the cycle of forest ecosystem. Long-term monitoring is required for the future assessment of radiocesium levels.

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Chapter 16
Diffusion and Transportation Dynamics of ^{137}Cs Deposited on the Forested Area in Fukushima After the Fukushima Daiichi Nuclear Power Plant Accident in March 2011

Nobuhito Ohte, Masashi Murakami, Takahiro Suzuki, Kohei Iseda, Keitaro Tanoi, and Nobuyoshi Ishii

Abstract A massive amount of radioactive substances, including cesium-137 (^{137}Cs), emitted from the disabled nuclear power plant has deposited on the forested areas in the northeastern region of Honshu Island, Japan after the Fukushima Daiichi nuclear power plant accident. Forests in these regions are particularly important not only for the forest products industry but also for source areas of drinking water and for residential environments. To clarify the mechanisms of diffusion and export of ^{137}Cs deposited on the forested ecosystem, we initiated intensive field observations in a small catchment, including forest and farmlands, in the Kami-Oguni River catchment in the northern part of Fukushima Prefecture. The following expected major pathways of ^{137}Cs diffusion and export were investigated: (1) transportation by water movement of dissolved and particulate or colloidal forms through hydrological processes and (2) diffusion through the food web in terrestrial and aquatic ecosystems of forests. Preliminary findings indicated the following: (1) most of the ^{137}Cs was discharged as suspended matter, and particulate organic matter appeared to be the most important carrier of ^{137}Cs. High water flow generated by storm accelerated the transportation of ^{137}Cs from the forested catchments. Estimation of ^{137}Cs export thus requires precise evaluation of the high flow acceleration during storm events. (2) Because litter and its detritus may form the biggest pool of ^{137}Cs in the forested ecosystem, ^{137}Cs diffusion occurs more rapidly through the detritus food chain than the grazing food chain. Most predators have already ingested ^{137}Cs, particularly in aquatic environments. An urgent question is when and how ^{137}Cs diffuses through grazing food chains and how rapidly

N. Ohte (✉) • K. Iseda • K. Tanoi
The Graduate School of Agricultural and Life Sciences,
University of Tokyo, Tokyo 113-8657, Japan
e-mail: nobu@fr.a.u-tokyo.ac.jp

M. Murakami • T. Suzuki
Graduate School of Science, Chiba University, Chiba 263-8522, Japan

N. Ishii
National Institute of Radiological Sciences, Chiba 263-8555, Japan

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this process occurs. To elucidate or predict these phenomena, the mechanisms of ^{137}Cs release from litter and soil organic matter need to be clarified.

Keywords ^{137}Cs • Food web • Forest • Fukushima Daiichi nuclear power plant • Hydrological processes

Abbreviations

^{137}Cs	Cesium-137
^{131}I	Iodine
MAFF	Ministry of Agriculture, Forestry and Fisheries
MEXT	Ministry of Education, Culture, Sports, Science and Technology

16.1 Introduction

The Fukushima Daiichi nuclear power plant accident in March 2011 emitted 1.5×10^{17} Bq of iodine-131 (^{131}I) and 1.2×10^{16} Bq of cesium-137 (^{137}Cs) into the surrounding environment [Ministry of Education, Culture, Sports, Science and Technology (hereafter, MEXT) and Ministry of Agriculture, Forestry and Fisheries (hereafter, MAFF) 2012]. These radioactive substances, including ^{137}Cs, were deposited on the forested areas in the northeastern region of Honshu Island, Japan. Forests in these regions are particularly important for humans not only for their association with the forestry industry but also for the source areas of drinking water and for residential environments. The first phase of government surveys and investigations showed that the major portion of deposited ^{137}Cs was trapped in the canopy and litter layer at the soil surface (Hashimoto et al. 2012; MEXT and MAFF 2012). ^{137}Cs has been shown to be easily adsorbed onto clay minerals and soil organic matter (Kruyts and Delvaux 2002), which can be transported by eroded soils and particulate and dissolved organic matter through hydrological channels, streams, and rivers (Fukuyama et al. 2005; Kato et al. 2010; Wakiyama et al. 2010). Dissolved ^{137}Cs, which is relatively free from soil adsorption, can also be taken up by microbes, algae, and plants in soil and aquatic systems. This form of ^{137}Cs will eventually be introduced into soil insects and worms, fishes, and birds through the food web (Fig. 16.1).

To precisely describe the mechanisms of diffusion and export of ^{137}Cs deposited on the forested catchment, field observations of two primary but different pathways were conducted. One was the transportation of ^{137}Cs through hydrological flow paths as particulate or dissolved substances. The other was its diffusion through the food web of plants and animals in forested and aquatic ecosystems.

In this chapter, we present the concept, method, and implementation of the field experiments and also report the results of preliminary investigations 15 months after the accident. The main focus of the field observations was the flow of materials not only of ^{137}Cs itself but also of related compounds through the continuum of forested

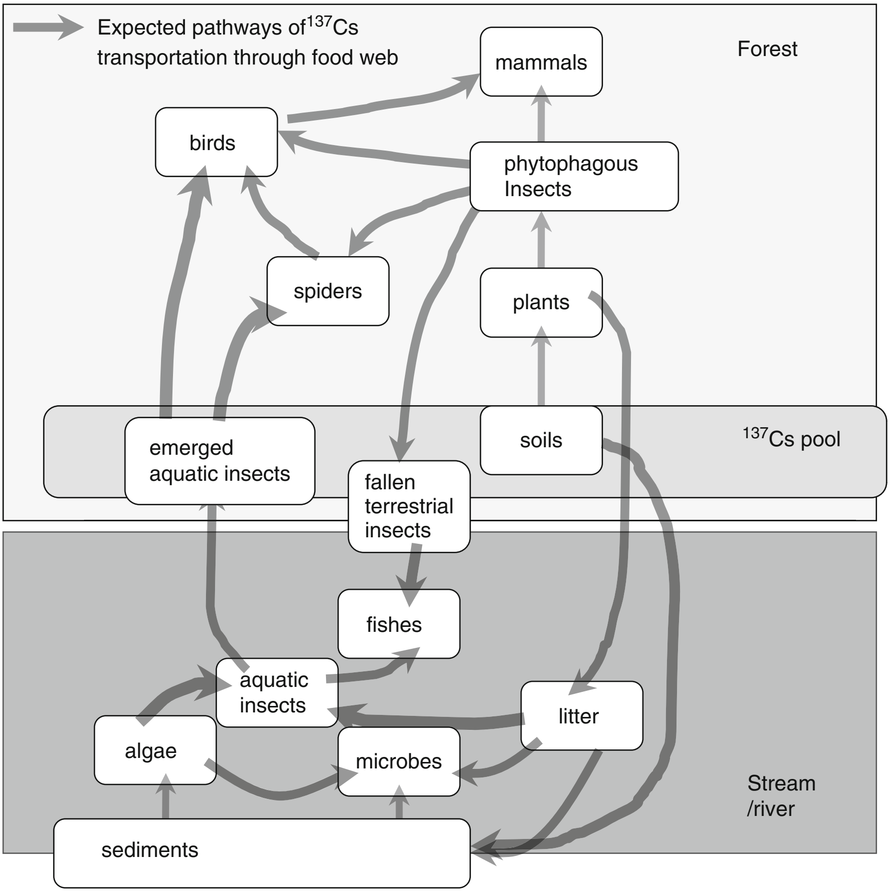

Fig. 16.1 Conceptual diagram of the pathways of cesium-137 (^{137}Cs) transfer thorough the food webs in aquatic and terrestrial ecosystems (based on Ohte et al. 2012)

and stream ecosystems. We are currently investigating how ^{137}Cs is transported by water and suspended materials and how it diffuses through the food webs in the forest–stream ecological continuum.

16.2 Materials and Methods

16.2.1 Study Site

The study was conducted at the Kami-Oguni River catchment in the northern part of Fukushima Prefecture. The area is located approximately 50 km from the Fukushima Daiichi nuclear power plant (Fig. 16.2a). According to a radioactivity survey report by MEXT that was performed using aircraft survey devices, the air

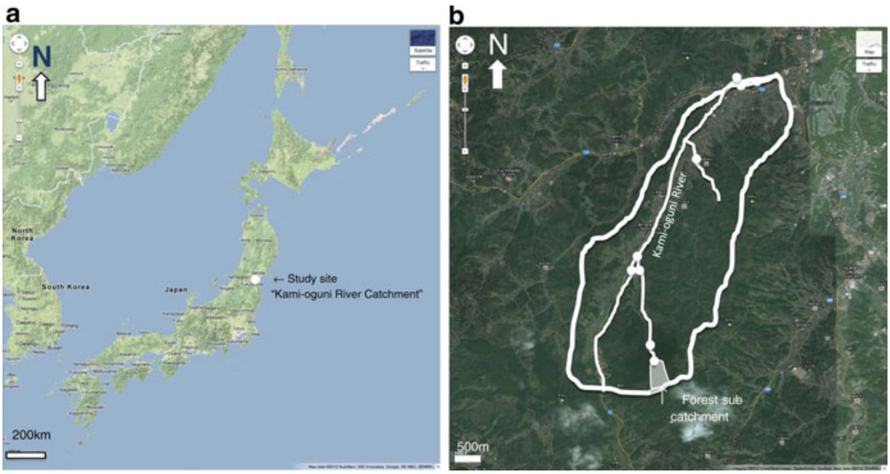

Fig. 16.2 (**a**) Location of the study site. (**b**) Observation facilities in the study catchment

dose rate in this region was 1.9–3.8 μSv/h and the total deposition rate of ^{137}Cs was 300–600 kBq/m^2 at June 28, 2012 (MEXT 2012).

The study catchment predominantly comprises forests and paddy fields, and the dominant tree species are broadleaf deciduous trees because the forested regions were mainly used as coppices until the 1960s (Watanabe, personal communication). Some parts were used as Japanese cedar and cypress plantations for timber production. The agricultural land also includes orchards producing peaches and Japanese persimmons.

In 2011, brown rice produced in several paddies in this area was found to have high radiocesium concentrations exceeding the provisional regulation level set by the Ministry of Health, Labour and Welfare of Japan. Based on this survey, MAFF decided to prohibit rice plantation during the 2012 season (MAFF 2011). The Date City government and local farmers only planted test plantations to investigate the rate of ^{137}Cs uptake by rice and the efficacy of countermeasures (applying potassium and materials that adsorb ^{137}Cs) in several selected paddies.

The upstream part of the study catchment is mostly forested, whereas the farmland areas, including the paddy fields, are mostly found in the middle to downstream parts of the catchment.

16.2.2 Survey

16.2.2.1 Forested Catchment

Within the forested parts of the study catchment, a small subcatchment was selected for intensive observation and sampling (Fig. 16.2b). Three rectangular plots

(20 m × 20 m), including two deciduous stands and one cedar plantation, were studied to determine the processes involved in ^{137}Cs pools and flows.

To describe the distribution and sizes of the ^{137}Cs pools found within the subcatchment, we collected periodical samples of litter, plants, soil, and stream water and measured the ^{137}Cs concentration of each sample.

To determine fluxes in ^{137}Cs deposition, movement within the subcatchment, and discharge from the catchment, we measured the water flux and ^{137}Cs concentration for every factor associated with ^{137}Cs movement, including rain, throughfall, stem flow, and streamwater discharge. The water discharge rate from the subcatchment was continuously measured using a partial flume with a water level gauge. Throughfall and stem flow water were collected from both the secondary deciduous stands and the cedar plantation.

Samples were collected from all terrestrial food web members, including litter and living leaves, soil worms, insects, lizards, and snakes.

16.2.2.2 River Transect

Water samples were also collected from nine points along the Kami-Oguni River to elucidate the riverine distribution of ^{137}Cs (Fig. 16.2b). The total length of the river transect was 7 km from the outlet of the forested subcatchment. Confluence locations were generally selected as sampling points. Samples were collected from members of the aquatic food web, including litter detritus, benthic algae, aquatic macrophytes, benthonic organisms, and fish.

16.2.3 Analysis

Water samples were filtered with a glass fiber filter (Whatman GF/F, $\phi = 0.7\ \mu m$) immediately after sampling. The ^{137}Cs concentration of suspended matter present on the filter was measured. Condensing treatment using a germanium semiconductor detector is required to measure the dissolved ^{137}Cs concentration. This will be performed in the next phase of the study. Samples of litter and organisms were dried in an oven, ground into a powder, and homogenized. Germanium semiconductor detectors at the University of Tokyo and the National Institute of Radiological Sciences were used for measuring ^{137}Cs concentrations of all the samples.

Carbon and nitrogen concentrations and the isotope composition ($\delta^{13}C$ and $\delta^{15}N$) of the samples were also determined. Isotope data were used to evaluate the trophic levels of sample organisms according to the methodology reported by Lajtha and Michener (1994).

16.3 Preliminary Results and Discussion

16.3.1 ^{137}Cs Export by Hydrological Processes

The observed spatial distribution of ^{137}Cs in suspended matter along the Kami-Oguni River in June 2012 is shown in Fig. 16.3c. A large storm event, a typhoon, occurred in the study area from June 19 to 20 (Fig. 16.3a, b). The increased runoff caused by this storm event exported much of the suspended matter from the source areas of the stream, such as the riparian zones of the forested areas (riparian topography is rarely found in agricultural areas because of their well-developed artificial drainage systems). The ^{137}Cs concentration in the suspended matter increased significantly during the storm event; however, it was diluted by the water discharged from the farmlands in the middle to downstream parts of the catchment. The contribution of the forested catchments located in the uppermost regions was significant.

The ^{137}Cs concentration decreased rapidly over the next day (June 20). This quick response suggests that the mechanism of ^{137}Cs discharge was highly dependent on rapid flows such as the overland flow that occurred in the riparian zones of the forested areas. While this response was rapid, it should be noted that the ^{137}Cs concentration in the suspended matter was higher than that before the storm event even after 1 week.

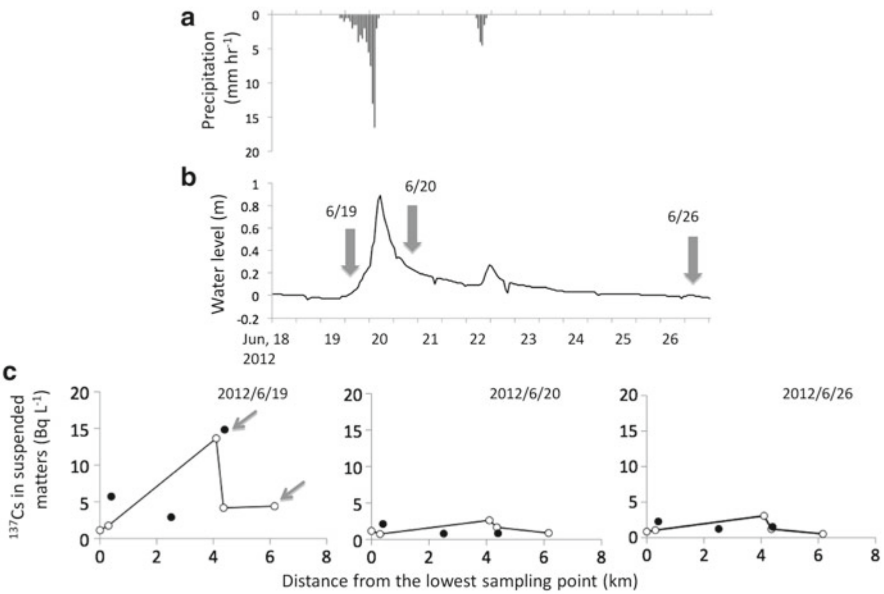

Fig. 16.3 Observed spatial distribution of cesium-137 (^{137}Cs) concentration in suspended matter along the Kami-Oguni River in June 2012. (**a**) Precipitation at Iidate, (**b**) water level of the Hirose River at Ohzeki (downstream of the Kami-Oguni River), (**c**) ^{137}Cs concentration of suspended matter. The *arrows* on the panel of 2012/6/19 indicate the influxes from the forested catchments (based on Ohte et al. 2012)

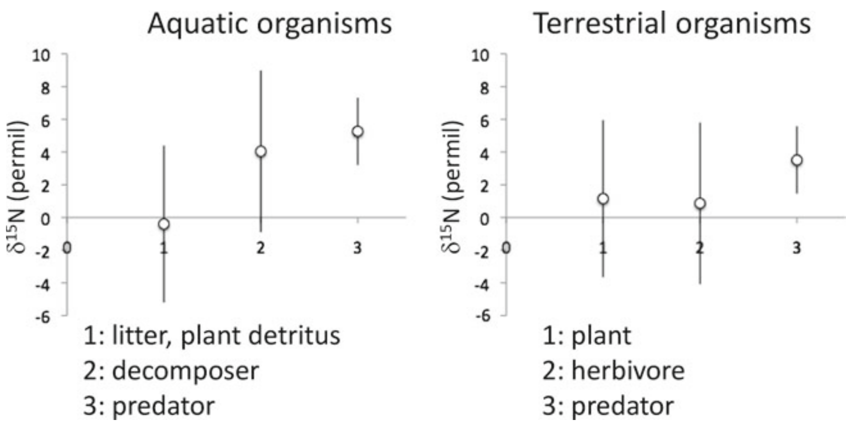

Fig. 16.4 $\delta^{15}N$ value for each expected trophic level of functional groups of aquatic and terrestrial organisms (based on Ohte et al. 2012)

It is generally accepted that the amount of suspended matter increases during conditions of high flow in headwater streams during storm events. It is also often observed that sources of suspended matter are the stream bank and the riparian zone surface. Our data suggest that suspended matter plays the most important role in the export of ^{137}Cs from the forested catchment. This interpretation is also supported by a study on other forests in Fukushima, which found that most of the ^{137}Cs pool remained in litter and decomposing organic materials found on the forest floor and soil surface (Hashimoto et al. 2012).

These findings indicate that high flows generated by storm events accelerate the transportation of ^{137}Cs by water from forested headwater catchments (Fig. 16.3). Precise estimation of the extent of ^{137}Cs export from forested catchments requires a realistic evaluation of high flow acceleration during storm events.

16.3.2 ^{137}Cs Diffusion Through the Food Web

The $\delta^{15}N$ values increased with the expected trophic levels of functional groups of aquatic and terrestrial organisms (Fig. 16.4). The ^{137}Cs concentrations in these trophic groups are shown in Fig. 16.5. The ^{137}Cs concentration in litter was significantly higher than that in the other groups. The concentration in mushrooms was also high, showing a similar tendency to that seen in a previous report (MEXT and MAFF 2012).

The ^{137}Cs concentration in aquatic organisms was generally higher than that in terrestrial herbivores, which clearly indicates that the majority of the aquatic primary consumers feed on litter detritus, whereas the primary consumers of the terrestrial food web mainly feed on living plant tissues. In other words, the fastest diffusion of ^{137}Cs occurs through the detritus food chain, and it is dominant in the food web of aquatic organisms. In contrast, in terrestrial organisms, herbivorous insects did not ingest ^{137}Cs, although the ^{137}Cs concentration of predators was

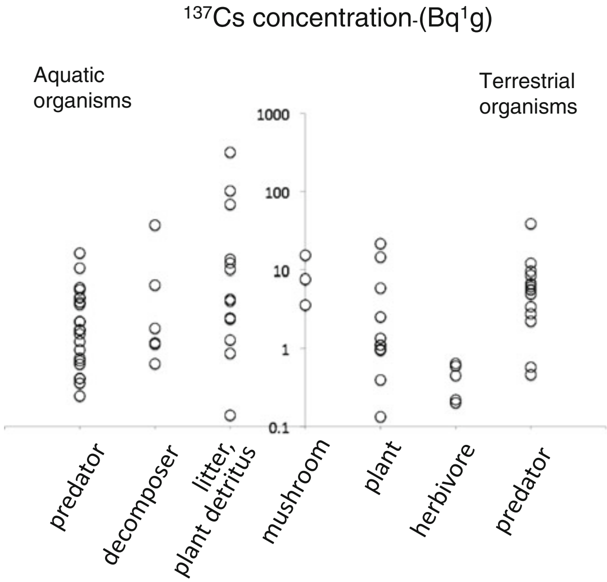

Fig. 16.5 Cesium-137 (^{137}Cs) concentrations of three trophic groups of aquatic and terrestrial organisms (based on Ohte et al. 2012)

significantly high. This suggests that many plants have not yet absorbed ^{137}Cs and that terrestrial predators such as lizards and snakes may consume some decomposers found in the shallow soil layers, e.g., angleworms and galleyworms. Figure 16.5 also indicates that ^{137}Cs is substantially diffused to primary consumers but has not completely reached yet high-level predators, particularly in the aquatic ecosystem.

An urgent question is when and how ^{137}Cs diffuses through the grazing food chain and how rapidly this process occurs. To predict this phenomenon, mechanisms of ^{137}Cs release from litter and soil organic matter as well as the ^{137}Cs absorbing behavior of plants need to be clarified.

16.4 Future Studies

Since the Chernobyl nuclear power plant accident, the dynamics of radioactive substances has been monitored in many forested ecosystems in Russia, the Baltic countries, and Scandinavia. Some of these have continued long-term ecosystem monitoring (Fesenko et al. 1995; Realo et al. 1995; Lehto et al. 2008). In southern Finland, for example, it was reported that the Chernobyl fallout remained on site, with more than 3,000 Bq/kg being found in the uppermost (0–3 cm) humus layer of forest soil (Ylipieti et al. 2008).

At present, in northeastern Japan, including Fukushima, the largest pool of fallout ^{137}Cs has been found in the litter layer of deciduous forest floors as well as in

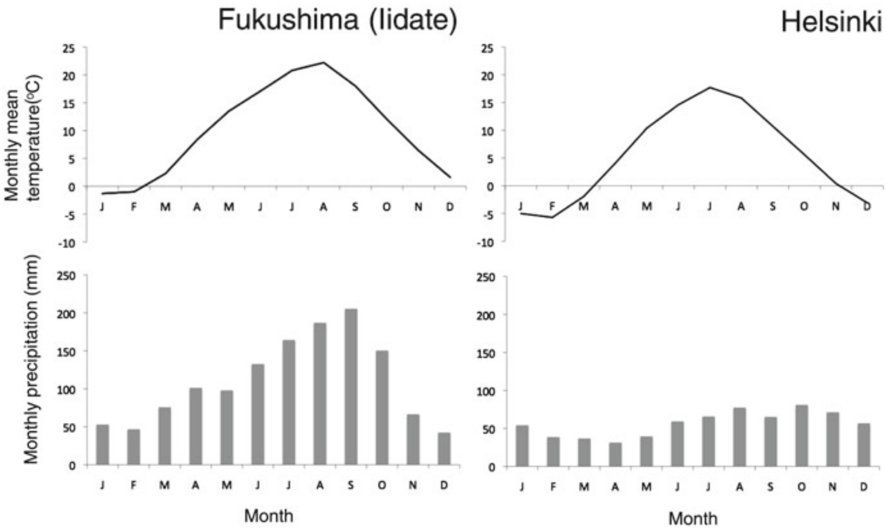

Fig. 16.6 Monthly mean air temperature and precipitation in Helsinki and Fukushima (Iidate town)

the canopies of evergreen coniferous forests. Therefore, the urgent and most important objective of future studies is to predict how fast ^{137}Cs moves from relatively fresh litter to the humus or mineral soils found beneath the litter layer and also how it is adsorbed to these soils. Although the Scandinavian case studies suggested high retention of fallout in humus, there are geographical differences between Scandianvia and Japan, including differences in climate and geology.

Figure 16.6 shows the mean monthly air temperature and precipitation for Helsinki and Fukushima (Iidate town). The most important point to note is that the total precipitation in Fukushima is twice that in Helsinki and that ~60% of the total precipitation falls in June, July, August, and September. This is a typical seasonal variation caused by the Asian monsoon system. High-temperature summers with high precipitation also accelerate not only the decomposition of litter on the forest floor but also the transportation of detritus and dissolved organic matter by hydrological pathways. Thus, mobilized ^{137}Cs may be transported more easily into the deeper soils and exported by streams in Fukushima than in Scandinavia.

In addition, the geological and geomorphological characteristics of Fukushima forests are substantially different from those of Finland. Japanese geological settings are generally affected by active orogenic movement. Uplift and erosion activities are much more active than those in Scandinavia and the northwestern part of the Eurasian continent. This may accelerate the erosion and export of surface materials from the forest floor because of the high precipitation rate in summer (Ohte and Tokuchi 1999).

Thus, we have to be careful when referencing information obtained from Russia, Ukraine, and Scandinavia because of environmental differences. For comprehensive comparison between Japan and these countries, intensive and long-term monitoring of various forested ecosystems in the northeastern parts of Japan is required.

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Chapter 17
Development of an Information Package of Radiation Risk in Beef After the Fukushima Daiichi Nuclear Power Plant Accident

Hiromi Hosono, Yuko Kumagai, and Tsutomu Sekizaki

Abstract This study aimed to improve public knowledge on radiation risk and its management system by providing information for the consumers in order to assist in food purchase decision making. To develop the necessary information, we administered web-based questionnaires twice and conducted two rounds of focus group interviews (FGIs) involving five groups between each questionnaire. Attitude toward food from suffered areas, trust in the risk management sector, and change in the knowledge level with and without providing the information were examined in each questionnaire. FGIs were conducted to identify any insufficient or confusing point in the provided information on radiation risk. As a result, although the risk of radiocesium was not regarded as high compared with other risks in beef, the willingness to pay (WTP) for food from affected areas was lower even if the contamination was checked to be below the regulation level. Public knowledge on current radiocesium contamination in food, risk control measures such as the regulation level and inspection, and health effects of low-dose radiation exposure was limited. The developed information package is available on the website of the Research Center for Food Safety belonging to the University of Tokyo.

Keywords Attitude • Information • Knowledge • Risk perception

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H. Hosono (✉)
Institute for Sustainable Agro-Ecosystem Services, Graduate School of Agricultural and Life Sciences, the University of Tokyo, 1-1-1 Midori-cho, Nishitokyo-shi, Tokyo, Japan
e-mail: hiromix@isas.a.u-tokyo.ac.jp

Y. Kumagai • T. Sekizaki
Graduate School of Agricultural and Life Sciences, The University of Tokyo, 1-1-1 Yayoi Bunkyo-ku, Tokyo, Japan

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17.1 Introduction

On March 11, 2011, the greatest earthquake ever recorded hit Japan. Even more devastating than the earthquake itself was the resulting tsunami and the subsequent accident at Tokyo Electric Power Company's Fukushima Daiichi nuclear power plant. Soon after the explosion at the power plant, the health risks of radioactive substances were of central concern. In terms of food safety, levels of radioiodine and radiocesium that exceeded the provisional regulation levels were detected on March 19, and abnormal levels of these radioactive substances have continuously been detected thereafter, particularly from the area surrounding the power plant. Radioactive contamination caused by the accident was detected at the 1st stage in water, vegetables, fruits, and milk and then in tea leaves in May, beef in July, and rice in September. Thus, public anxiety regarding radiation risk is widespread and has affected the food market, particularly in the Kanto and Tohoku regions, despite the implementation of measures to ensure that foods contaminated above the regulation level were not supplied for human consumption.

On March 17, 2011, the Japanese Ministry of Health, Labor, and Welfare (MHLW) adopted a provisional regulation level of radioactive substances in food to control radiation exposure from foods; this was set at <5 mSv/year. Under this provisional regulation, the upper limit of radiocesium contamination in drinking water and dairy products was set at 200 Bq/kg, and the upper limit for other foods such as meat, vegetables, and grains was set at 500 Bq/kg (Table 17.1). After risk assessments were made by examining the related literature and consideration was given to public concern, the regulation level was revised upward in April 2012. Under the new standard that considers the maximum permissible dose to be 1 mSv/year for foods, the upper limit of radiocesium in general food was set at 100 Bq/kg (50 Bq/kg for milk).

Along with setting the maximum permissible dose of radioactive nuclide contamination in food, public inspections have been intensively conducted around the affected area. Up to the end of March, a total of 136,975 food samples were inspected, and 1,204 of these were found to have radioactive nuclide levels higher than the regulation levels (inspection results are available at the Ministry of

Table 17.1 Provisional regulation levels and the new standard for radioactive nuclides in foods

Provisional regulation level				New standard	
Radioiodine in food		Radiocesium in food		Radiocesium in food	
Category	Limit (Bq/kg)	Category	Limit (Bq/kg)	Category	Limit (Bq/kg)
Drinking water	300	Drinking water	200	Drinking water	10
Milk, dairy products	300	Milk, dairy products	200	Milk	50
Vegetables	2,000	Vegetables	500	General foods	100
				Infant foods	50

Note: Ministry of Health, Labour and Welfare

Agriculture, Forestry and Fisheries website). Once foods with radiocesium/iodine concentrations higher than the regulation level are detected, shipments of such foods from the same area are restricted, and it takes at least 1 month before normal shipping can be resumed.

Information on the inspection results and shipment restriction areas is available on the MHLW website, which has been updated daily since March 2011. Other information related to radiation risk such as the type of radiation, annual exposure dose under usual conditions, internal and external exposures, and epidemiological findings related to radiation exposure as well as risk control measures taken in Japan after the accident is also provided on the websites of government ministries and agencies. Public forums to discuss radiation risks have been held in each locality.

Many TV programs and newspapers provided commentaries on the radiation risk, measures taken, and current situation regarding radiation contamination of foods as well as efforts to recover from the damage. Other internet-based tools such as social networks and blogs were used by consumers to gather information on radiation risks. Nevertheless, the information gathered from all these sources was sometimes inconsistent, and there have been numerous conflicts of opinions, even among specialists. Given the flood of information, it was difficult for people to judge the radiation risk by consuming food from the affected areas. In this context, many people avoided purchasing food from the affected areas even if these areas had been inspected. Consumer surveys administered after the explosion at the Fukushima Daiichi nuclear power plant revealed that radioactive contamination of food was of major concern and was regarded as the highest risk associated with food consumption (Food Safety Commission 2011; Kito 2012; Hosono and Nakashima 2012; Hosono et al. 2012a).

This study aimed to improve public knowledge on radiation risk caused by the accident and its management in order to assist with food purchasing decisions by providing information.

17.2 Radioactive Contamination of Beef

As noted above, no radioactive contamination of beef above the regulation levels was detected until the beginning of July. On July 8 and 9, 11 heads of beef cattle shipped to Tokyo from Minamisoma City in Fukushima Prefecture were found to contain 1,530–3,200 Bq/kg of radiocesium. Thereafter, their feed, water, and environment were intensively investigated. On July 14, rice straw on the beef cattle farm was found to be contaminated with high doses of radiocesium. On July 19, shipments of beef cattle from Fukushima Prefecture were restricted. Thereafter, shipments of cattle from Miyagi, Iwate, and Tochigi prefectures were also restricted. The distribution of the contaminated rice straw was traced, and a blanket inspection of cattle farms was implemented. The shipment ban was gradually reduced and was lifted at the end of August because of the application of intensive inspections at slaughter houses. If the source of cattle was a farm where feed control was not confirmed, where there was a possibility of feeding contaminated rice straw, or where radiocesium contamination over regulation levels had been previously identified, the cattle were subjected to intensive inspections at the slaughterhouse (the Government of Japan website 2011). Comprehensive examination of farms was also performed even if it was confirmed that contaminated feed was not provided to the cattle. After the accident, a total of 90,661 beef samples produced in eastern Japan were submitted for public inspection of radiation levels before the end of March 2012. These inspections identified 157 samples that exceeded the provisional regulation level for radiocesium (500 Bq/kg) and 1,092 samples that exceeded the new standard (100 Bq/kg). Although a blanket inspection of beef was not required publicly, this was voluntarily implemented and covered all slaughtered beef from the affected area to ensure food safety.

During our study on the food safety risk posed by beef, we experienced an outbreak of enterohemorrhagic *Escherichia coli* O111 and O157 from raw beef provided by the BBQ franchise in April 2011; this claimed five lives and more than 100 patients were infected. On this occasion, the standards used to assess raw meat at restaurants and/or shops were re-examined and enhanced to prevent further food poisoning outbreaks. The new code took effect on October 1, 2011. Since the BSE crises in September 2001 and December 2003 (during which the 1st cases were detected in Japan and the US, respectively) and the economic downturn and foot-and-mouth disease outbreak in 2010, the beef market in Japan has been experiencing a tough time.

17.3 Research Outline

We conducted a series of studies to develop an information package on radiation, associated health risks, and measures taken to control food radiation risk after accidents for improving the knowledge of consumers. To collect the necessary information, we administered two web-based questionnaires and conducted two rounds of focus group interviews (FGIs) involving five groups between the administration of each questionnaire (Fig. 17.1). Prior to the 1st web questionnaire, we prepared ten slides of information to check its effectiveness. FGI participants were then recruited from the respondents to the 1st questionnaire. Those who were able to attend two FGIs and who had an average level of knowledge were selected as FGI participants.

In the 1st FGIs, we asked the participants to identify any points of confusion and to note any additional information that would help increase the understanding of radiation risk and the measures taken to ensure food safety. During the 1-month interval between FGIs, we prepared 45 slides of information considering the opinions collected during the 1st FGIs. The 45 slides of information were prioritized by each group, and their responses were collated during the 2nd round of FGIs. The information package provided was developed according to the FGI outcomes.

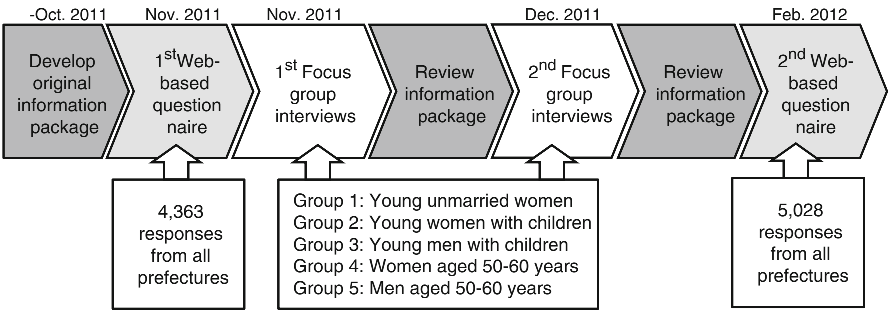

Fig. 17.1 Outline of the study

Table 17.2 Internet-based questionnaire respondents

		October 2011		March 2012	
		No. of respondents	%	No. of respondents	%
Total		4,363	100.0	5,028	100.0
Sex	Male	2,165	49.6	2,641	52.5
	Female	2,198	50.4	2,387	47.5
Age group	20–29	882	20.2	873	17.4
	30–39	839	19.2	1,014	20.2
	40–49	864	19.8	1,078	21.4
	50–59	861	19.7	1,047	20.8
	60–69	917	21.0	1,016	20.2
Residential area	Hokkaido	70	1.6	95	1.9
	Tohoku	433	9.9	582	11.6
	Kanto	1,462	33.5	1,255	25.0
	Hokuriku	281	6.4	382	7.6
	Chubu	347	8.0	516	10.3
	Kinki	500	11.5	624	12.4
	Chugoku	367	8.4	479	9.5
	Shikoku	292	6.7	383	7.6
	Kyushu	526	12.1	629	12.5
	Okinawa	85	1.9	83	1.7

Two internet-based questionnaires monitored by Nikkei Research Inc. were offered in October 2011 and March 2012 to respondents in all the prefectures in Japan. As shown in Table 17.2, the 1st survey had 4,363 respondents and the 2nd had 5,028 respondents. The survey covered the following subjects: (1) risk perception of seven hazards that may be derived from beef; (2) knowledge about radioactive substances, health risks, and control measures taken in Japan after the accident; (3) trust in radiation risk management implemented by the government and food industry; (4) attitude toward food safety and radiation risk management; and (5) intention to purchase food from the affected area. We provided the prepared information pack during the final phase of both the surveys to check whether it led to any improvement in public knowledge.

17.4 Results

17.4.1 Risk Perception, Knowledge, and Attitude

Risk perceptions of the seven beef-derived hazards were rated on a scale from 0 (no risk) to 5 (very high risk). The average risk perception score for each hazard is shown in Fig. 17.2. In the 1st survey, the risk of radioactive contamination was ranked 6th among men and 5th among women. The risk of enterohemorrhagic *E. coli* was ranked the highest by both men and women, followed by the risk of *Salmonella* spp. infection. Women perceived the risk of bovine spongiform encephalopathy as the 3rd highest, whereas men perceived the risk of *Campylobacter* spp. infection as the 3rd highest. In the 2nd survey, which was administered 1 year after the Fukushima Daiichi nuclear power plant accident, enterohemorrhagic *E. coli* infection was also regarded as the highest risk. The perceived risk levels of the three bacterial hazards were lower than those in the 1st survey. However, the perceived risk of bovine spongiform encephalopathy, radioactive contamination, and antibiotic residues was similar. Consequently, the risk of radioactive contamination was ranked 2nd among women and 5th among men in March 2011. Risk perception by women was higher than that by men for all the presented hazards ($p < 0.05$).

Knowledge related to food poisoning, bovine spongiform encephalopathy, and radioactive contamination was classed as subjective or objective in the 1st survey. Subjective knowledge was tested in the following five categories: "I am well informed about health risks and measures to control such risks," "I have some knowledge about health risks and measures to control these," "I know about health

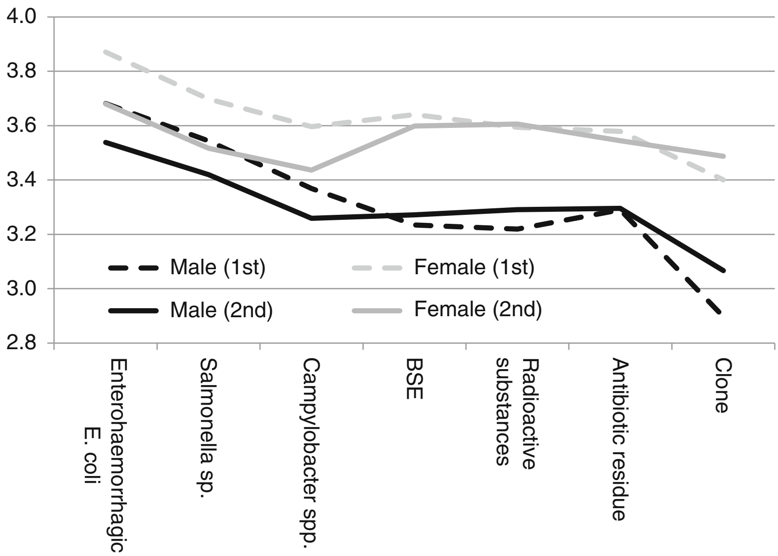

Fig. 17.2 Risk perception of seven hazards in beef. *Number* indicates the average score on a scale from 0 (no risk) to 5 (very high risk)

risks," "I do not have detailed knowledge of health risks, but I have heard of them," and "I have never heard of health risks." For each hazard, the 1st row indicates the number of respondents who indicated that they had respective subjective knowledge of the hazard. To evaluate their objective knowledge, five correct or incorrect descriptions of three hazards were included, and the respondent had to indicate whether he/she thought that these descriptions were correct, incorrect, or whether he/she did not know. The proportion of correct responses is shown in relation to subjective knowledge responses in Table 17.3.

Knowledge related to food poisoning, such as the implementation of stricter raw beef hygiene restrictions and the fact that food poisoning cannot always be prevented even in frozen or heat-treated foods, was comparatively well known. The 1st case of bovine spongiform encephalopathy in Japan was detected in September 2001, and this disrupted the beef market. One month later, blanket inspections were introduced to stabilize the market. Even among respondents who thought that they knew about the disease, 30% did not realize that bovine spongiform encephalopathy had occurred in Japan. Less than half people realized that the causative agent of bovine spongiform encephalopathy is concentrated in the brain or neurological system and is rarely found in beef. This indicates the possibility that the information provided during and after the bovine spongiform encephalopathy outbreak focused more on the introduction of blanket testing and less on scientific knowledge and effective control measures for preventing the disease[1] (Hosono et al. 2012b).

Objective knowledge on radioactive contamination was correlated with subjective knowledge. However, more than 70% of the respondents did not realize the radiation exposure from food under control, the fact that careful inspection is required before lifting the shipment restriction, and the fact that gene has repair function.

The knowledge section of the 2nd survey was focused more on radiation risk. Similar to the 1st survey, the questions used to evaluate the knowledge level consisted of correct and incorrect descriptions that the respondents had to rate as "true," "false," or "do not know." The proportion of correct answers to each question is shown in Fig. 17.3. Radioactive cesium control measures, the level of exposure from food, as well as the fact that radiation does not always cause cancer were well known. Although the terms Becquerel and Sievert appeared frequently in the media after the accident, only 38% of the respondents realized the meaning of these units. Moreover, <20% of the respondents realized the probability of health effects caused by radiation, knew about the procedures used to lift shipment restrictions, or knew that the interim regulation level for food was set to <5 m Sv/year. Public understanding of the health effects of exposure to low-dose radiation and knowledge of the current contamination level and food control measures were shown to be limited.

Approximately 70% of the respondents thought that food should not be sold if even the slightest radiocesium was detected, and only 20% thought that blanket farm inspections were costly in terms of both money and time and were therefore unnecessary (Table 17.4). In contrast, >40% of the respondents did not care about

[1] Approximately 70% Japanese do not feel safe to consume beef that had not been subjected to inspection for bovine spongiform encephalopathy and agree to continuous blanket inspections.

Table 17.3 Subjective and objective knowledge on food poisoning, bovine spongiform encephalopathy, and radioactive contamination

		Never heard	Heard but not know well	Know about health risk	Know health risk and control	Well know health risk and control	Total
Food poisoning	EHEC health risk/control total	122	817	1,898	1,181	345	4,363
	Food poisoning occurs two times more frequent in summer than in winter	31	354	822	493	141	1,841
		25.4%	43.3%	43.3%	41.7%	40.9%	42.2%
	The most popular agent of foodborn disease in Japan is EHEC O157	45	517	1,266	793	229	2,850
		36.9%	63.3%	66.7%	67.1%	66.4%	65.3%
	Hygiene restriction of raw meat has been enhanced because of the outbreak of *Escherichia coli* O111 and O157 in spring 2011	40	482	1,360	923	274	3,079
		32.8%	59.0%	71.7%	78.2%	79.4%	70.6%
	Adequate heating will perfectly prevent food poisoning	43	415	1,097	696	208	2,459
		35.2%	50.8%	57.8%	58.9%	60.3%	56.4%
	Once frozen, food poisoning caused by microorganisms can be prevented	64	661	1,598	1,058	308	3,689
		52.5%	80.9%	84.2%	89.6%	89.3%	84.6%
BSE	BSE health risk/control total	222	1,143	1,314	1,280	404	4,363
	BSE has not been detected in Japan	47	531	734	895	279	2,486
		21.2%	46.5%	55.9%	69.9%	69.1%	57.0%
	BSE spread by aerial infection	54	601	865	987	318	2,825
		24.3%	52.6%	65.8%	77.1%	78.7%	64.7%
	BSE became easy to control because effective vaccine has been developed	58	618	869	991	326	2,862
		26.1%	54.1%	66.1%	77.4%	80.7%	65.6%
	The causative agent of BSE is concentrated in the brain and neurological system	35	334	536	660	214	1,779
		15.8%	29.2%	40.8%	51.6%	53.0%	40.8%
	BSE risk in Japan is internationally regarded as controlled because of implementing blanket testing	39	308	423	442	113	1,325
		17.6%	26.9%	32.2%	34.5%	28.0%	30.4%

Radioactive substance	Radioactive substance health risk/control total	185	941	1,501	1,339	397	4,363
	Additional radiation exposure by consuming 200 g of beef contaminated with 500 Bq/kg of ^{137}Cs daily for one year will not exceed 1 mSv	22	108	215	270	109	724
		11.9%	11.5%	14.3%	20.2%	27.5%	16.6%
	Epidemiological result from Hiroshima and Nagasaki victims indicates the exposure to radiation would increase the health risk in offspring	51	326	642	668	210	1,897
		27.6%	34.6%	42.8%	49.9%	52.9%	43.5%
	The health risk of 1 mSv radiocesium exposure is higher than that of 1 mSv radioiodine because the half life is longer	30	190	499	574	202	1,495
		16.2%	20.2%	33.2%	42.9%	50.9%	34.3%
	Once shipment restrictions are applied by detecting radiation from a food, it takes 1 month to resume reshipping	9	88	198	220	69	584
		4.9%	9.4%	13.2%	16.4%	17.4%	13.4%
	Even cells are damaged by exposed to radiation, DNA has repair function	16	64	175	211	95	561
		8.6%	6.8%	11.7%	15.8%	23.9%	12.9%

Note: The 1st row of each hazard indicates the number of respondents that indicated that they had subjective knowledge. The other numbers indicate the proportion of correct responses and the accuracy rate for each question

BSE bovine spongiform encephalopathy

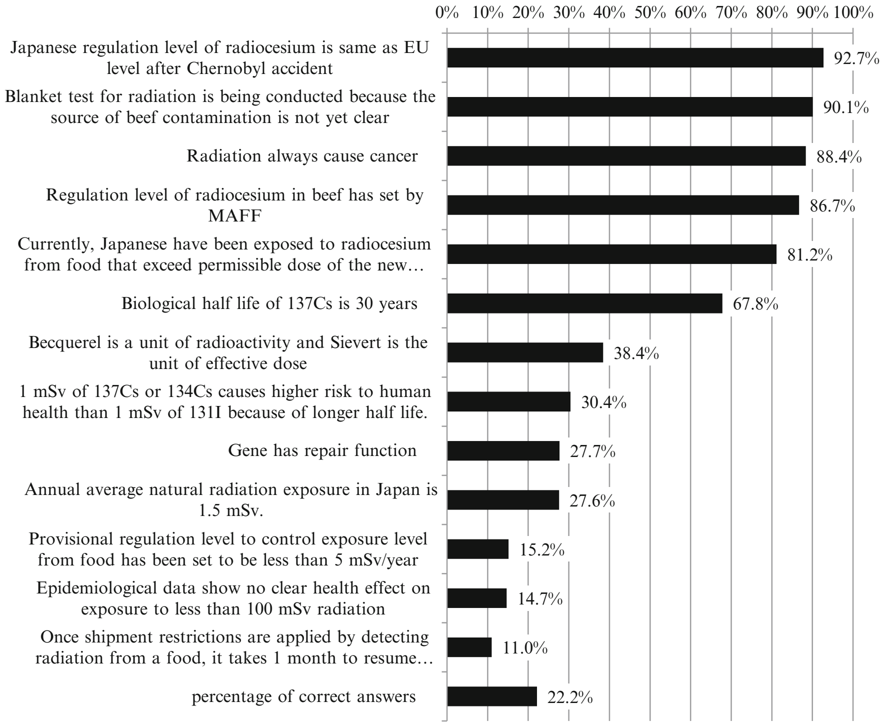

Fig. 17.3 Objective knowledge of the 2nd survey respondents. The *numbers* indicate the proportion of correct answers for each description

Table 17.4 Attitude on radiation control in food

	1st survey		2nd survey	
	Agree (%)	Relatively agree (%)	Agree (%)	Relatively agree (%)
Food should not be sold if a slightest radiocesium is detected	30.0	27.0	27.7	30.7
Not necessary to inspect all farms because it is costly in terms of time and money	6.9	15.8	4.6	16.5
I do not care because food containing radiation above regulation level would not be sold	16.4	30.3	14.4	32.7
I try not to care because the risk of radioactivity from food is not so high	13.3	31.3	11.5	31.6

the radiation risk because food containing levels above the regulation level would not be sold or because the risk of radioactive contamination from food is not high. However, the trustworthiness of central and local government agencies in terms of their ability to control radioactive contamination of food was not high (Fig. 17.4). The trustworthiness of food manufacturers and retailers was comparatively higher; however, only 30% of the respondents trusted their ability to implement radioactive

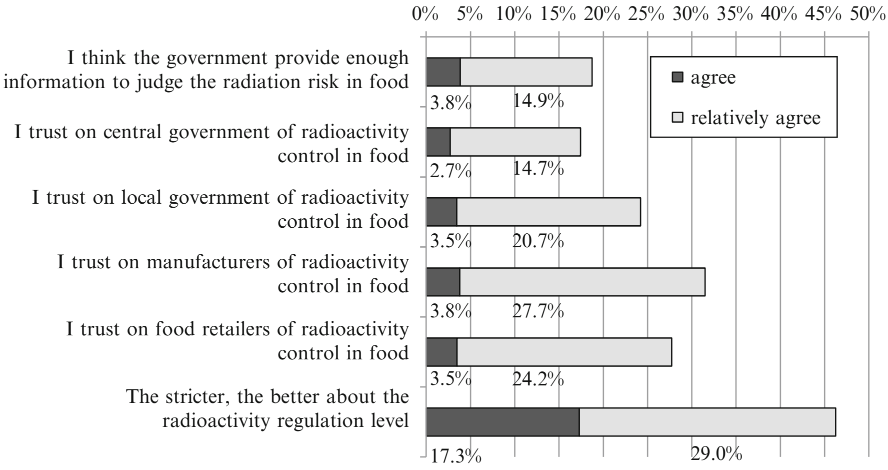

Fig. 17.4 Trustworthiness of risk management agents and risk control (2nd survey)

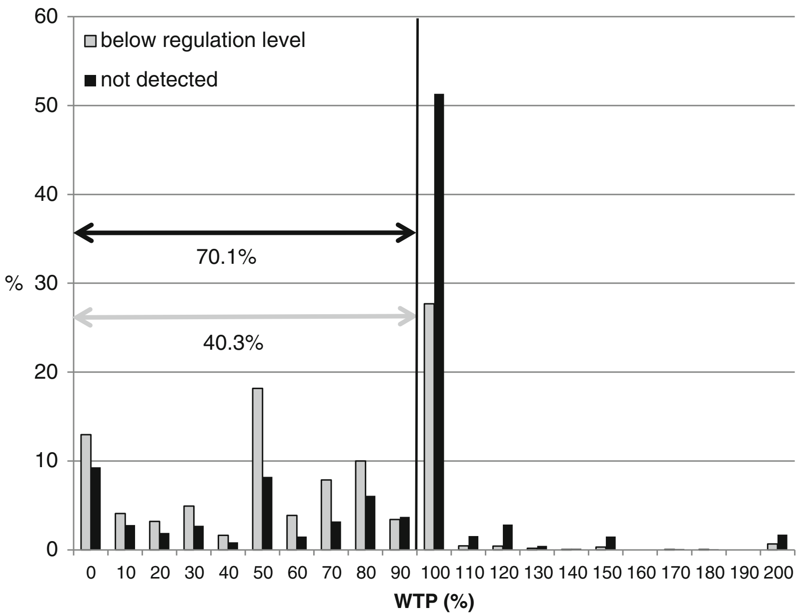

Fig. 17.5 Willingness to pay (WTP) for the food from affected area (1st survey)

contamination control. To ensure food safety, 17.3% of the respondents (46.3% of those who indicated relative agreement) considered "the stricter, the better" about the radioactivity regulation level. Approximately 20% of the respondents considered the provided information to be insufficient to judge the radiation risk in food.

The willingness to pay (WTP) for the food from affected areas if the radioactive contamination level was inspected and proven to be lower than the regulation level or not detected was also investigated (Figs. 17.5 and 17.6). Answers ranged from 0% to

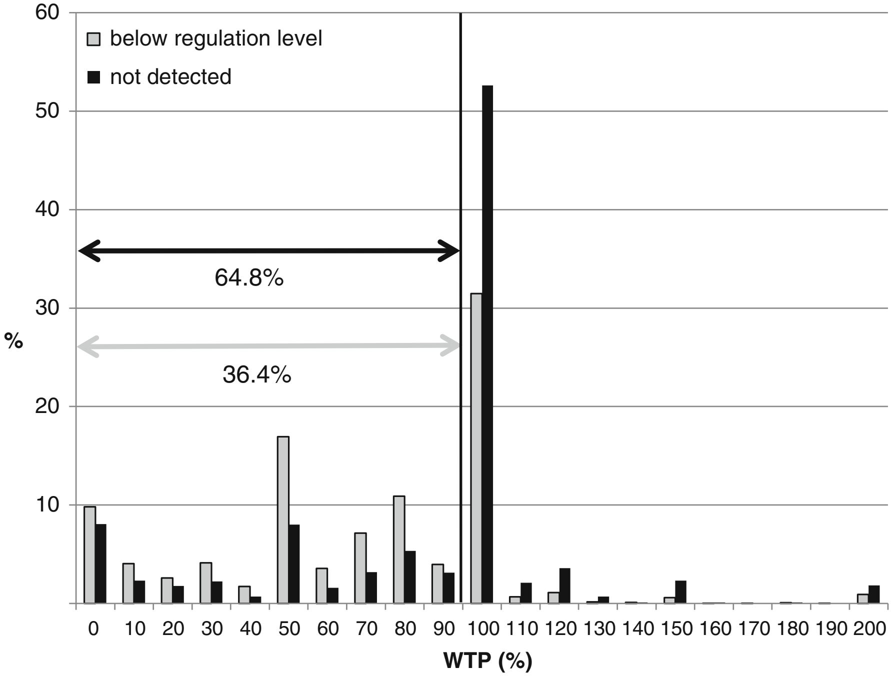

Fig. 17.6 Willingness to pay (WTP) for the food from affected areas (2nd survey)

200% assuming that the price of food derived from an unaffected area was 100%. The average relative WTP if radioactive contamination was lower than the regulation level and was not detected was 62.1% and 80.5%, respectively, in the 1st survey and 67.6% and 83.8%, respectively, in the 2nd survey. Approximately 70% (1st survey) and 65% (2nd survey) of the respondents provided WTP ratings lower than 100% for food from affected areas even if radioactive contamination was below regulation levels. Even if radioactive substances were not detected, 40.3% (1st survey) and 36.3% (2nd survey) of the respondents provided WTP ratings less than 100%. Approximately 10% of the respondents did not want to consume food from affected areas (0% WTP). In contrast, 8.4% (1st survey) and 11.0% (2nd survey) of the respondents provided WTP ratings over 100% if radioactive substances were not detected.

17.4.2 Effectiveness of the Information Package

To determine whether the prepared information improved the understanding of radiation risk and control measures, we provided the respondents with four types of information: narration and captions with and without information on beef cattle raising processes (slide 10) in the 1st survey. It took 2–3 min to view the slide. The contents of the prepared slide included the following information: (1) internal and external radiation exposure, (2) natural radiation exposure levels, (3) regional

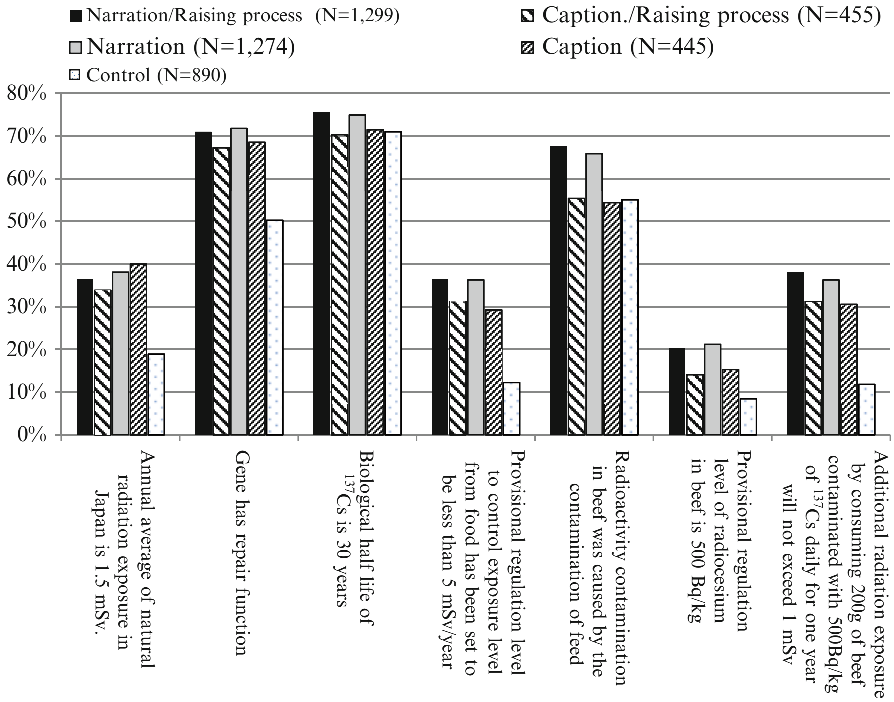

Fig. 17.7 Information pattern and proportion of correct answers (1st survey)

differences in natural radiation exposure levels, (4) effects of exposure on cells/genes and the repair function of genes, (5) physical half life and biological half life, (6) the provisional regulation level in food, (7) the concept behind the setting of provisional regulation levels, (8) radioactivity inspections of food, (9) reasons for radiocesium contamination in beef and exposure levels after consuming 200 g of 500 Bq/kg radiocesium-contaminated beef daily for 1 year, and (10) the process used to raise beef cattle.

One-fifth of the respondents formed a control group and were not provided with any of the information outlined above. We included seven correct or incorrect descriptions in the questionnaire, and the respondents were asked to indicate if these were "true," "false," or "did not know" after the information was provided. Figure 17.7 shows the proportion of correct answers, which indicates the effectiveness of the information provided to improve understanding. The proportion of correct answers among the control group was 32.5%, whereas that among respondents provided information with and without narration was 49.2% and 43.8%, respectively. Information with narration was better at improving understanding than that with figure captions.

Because narration was shown to improve understanding about radiation risk in the 1st survey, we prepared two information packages for the 2nd survey. These packages took 5 or 10 min to view, and the respondents chose one of them by themselves. If they began viewing the information and thought it was too long or not

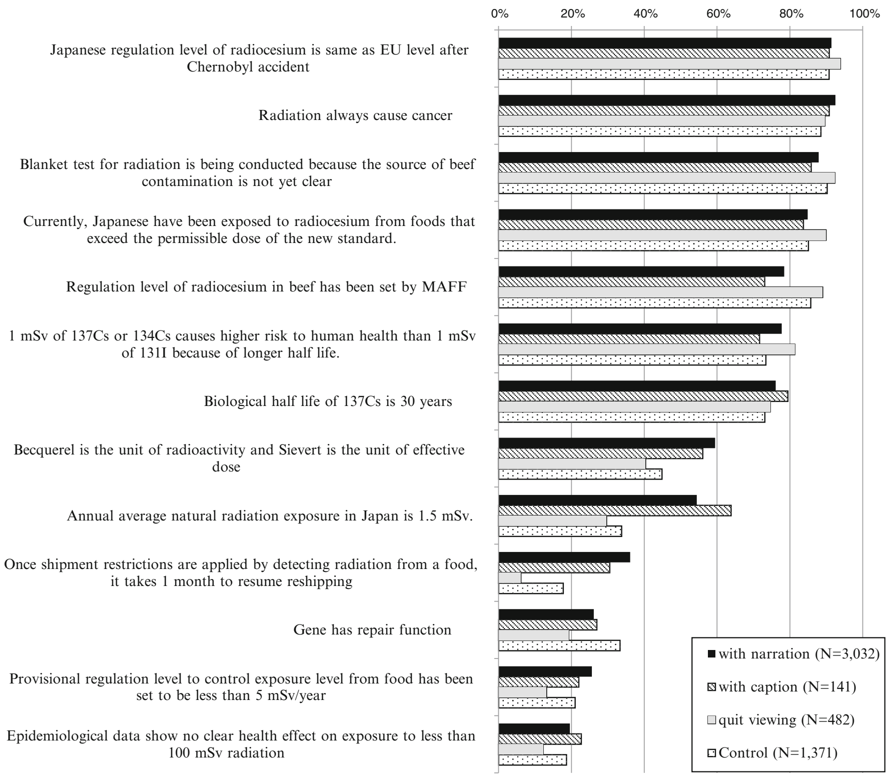

Fig. 17.8 Information pattern and proportion of correct answers (2nd survey)

interesting, they could stop watching it. For those who could not hear the narration when they viewed the slides on the website, we prepared 5- and 10-min versions with captions. Only 353 respondents selected the 10-min version, and 327 of these stopped viewing the slides in the middle of the show. As shown in Fig. 17.8, the information provided in the 2nd survey improved understanding of the meaning of Becquerel and Sievert, annual natural radiation exposure levels, and the process of lifting shipment restrictions. The responders who stopped viewing the information already knew about the topics that were well known among Japanese at the time of the survey. However, the knowledge on control measures, e.g., the regulation level and shipment restriction, as well as the health effects of low-dose exposure was limited among those who quit viewing.

17.4.3 Results of FGIs

Two rounds of FGIs were held for 2 h in November and December 2011; these involved 30 participants in five groups: unmarried women, women with children

<12 years old, men with children <12 years old, women aged 50–60 years, and men aged 50–60 years. The topics discussed were the concern for general food safety issues, including radiation exposure from food, as well as any additional information they enquired about related to radiation risk and its management. The results of FGIs are summarized in Table 17.5. Regarding food safety in general, the participants addressed pesticide residues, food additives, genetic engineering, food poisoning (particularly from raw meat), bovine spongiform encephalopathy, dioxin, allergens, radionuclide contamination, and anxiety around animal cloning. Because of a recent repeated mislabeling incident, distrust of processed food was mentioned by both women and men who had young children.

The information required to improve understanding about the radiation risk was similar among the groups. All the groups indicated that they lacked information on the relationship between the exposure level and adverse health effects. Among the younger groups, radiation risk according to age, particularly the adverse health

Table 17.5 Summary of the focus group interviews

Group	Consciousness for food safety	Comments of necessary information on the problems caused by radioactive substances in foods
Mothers with children <6 years	• Country of origin • Adverse effect on children – Agricultural chemicals, dioxin • Price	• Easy-to-understand explanation on – Examination methods, examination plans of local governments and the central government – Process of establishing the standards of radioactive substances in foods – Relationship between Sv and Bq – Differences of adverse effects between children and adults • Interpritation of "half life", "probabilistic effect", and "determistic effect" • Cuntermeasures for radioactive substances at foreign areas indicating higher level of radioactivity • Comparison with other risks • Management of risk for radioactive substances in foods
Married women (from 50 to 60 years old)	• Country of origin • Adverse effect on children – Agricultural chemicals, dioxin • Price • Degree of confidence for retails, super-market, restaurants, and others	• Easy-to-understand explanation of examination methods • Easy-to-understands explanation of the process of establishing the standards of radioactive substances in foods • Interpritation of "half life" • Information on what we have to do now • Countermeasures for radioactive substances at foreign areas indicating higher level of radioactivity

(continued)

Table 17.5 (continued)

Group	Consciousness for food safety	Comments of necessary information on the problems caused by radioactive substances in foods
Un married women (from 20 to 30 years old)	• Country of origin • Foodborne diseases • Agricultural chemicals • Food additives • Radioactive substances	• Easy-to-understand explanation of examination methods, inspection plans of local government and the central government • Easy-to-understand explanation of the process of establishing the standards of radioactive substances in foods • Relationship between Sv and Bq • Interpritation of "half life" • Countermeasures for radioactive substances at foreign areas indicating higher level of radioactivity
Fathers with children <6 years	• Country of origin • Risks for hazards in foods, for example GMO, BSE, food additives, environmental endocrine disruptors, agricultural chemicals, radioactive substances • Food labeling	• Adverse effect on children, – What type of foods cause adverse health effect – How much children can eat • Comparison with other risks • Comparison with similar accidents such as Chernobyl and atomic bomb
Married men (from 50 to 60 years old)	• Management of food safety	• Easy-to-understand explanation of the process of establishing the standards of radioactive substances in foods • Comparison with similar accidents • Epidemiological study for radiation technologists, pilots, and other occupations • Relationship between examination data and adverse effects on human health • Comparison with other risks • Countermeasures for radioactive substances at foreign areas indicating higher level of radioactivity

effects on young children, was of primary concern. All the groups wanted to know their radiation exposure level caused by the Fukushima Daiichi nuclear power plant accident compared with that caused by past accidents, high-risk occupations, and residence in high-dose regions.

In addition to the exposure level and health effects, a lack of information on control measures was also indicated. The participants considered that the public was not well informed of the concept of a provisional regulation level, the frequency of inspections of targeted areas, the location of the targeted areas, or the inspection regime used. Although detailed information is available on websites, it may be difficult for consumers to find the information that they would like to know or it may be difficult to understand. Some young participants required simply a declaration that the "food is safe" because it is too difficult to understand the radiation risk.

Another opinion was expressed about the objective of the information provided. We assumed consumers would be able to judge the risk of radiation from food by gaining an understanding of the radiation risk. Therefore, the information provided consists of scientific facts related to radiation and health risks as well as control measures taken after the Fukushima accident. The objective of the provided information was to improve understanding. However, the expectation of the participants was different. They wanted clearer message such as "food is safe" or "be careful when choosing food."

The information package developed through this study is available on the website of the Research center for Food Safety, the University of Tokyo (http://www.frc.a.u-tokyo.ac.jp/event/radioactive/radioactive.html).

17.5 Conclusion

After the explosion at the Fukushima Daiichi nuclear power plant, radioactive substances in food became a major concern of Japanese consumers. Some people avoided purchasing food from affected areas, particularly from Fukushima Prefecture, even if it was intensively inspected. According to opinions obtained in this study, this was because the inspections were not comprehensive. Therefore, consumers are anxious whether the food in front of them is properly controlled or has been inspected and found to be safe. Moreover, knowledge on radionuclides, the meaning and magnitude of Becquerel and Sievert measures, exposure levels, health effects, the concept of a regulation level, and risk control measures was limited. If the relationship between the exposure level, health effects, and regulation levels is not realized, people would not feel safe even if food is controlled and found to be below the regulation level. In this context, it is natural for people to require stricter radioactivity regulations. Choosing food inspected and shown to be free of radiation or food from an unaffected area is one means of feeling secure. Information connecting the current contamination level and its health effects should be provided to the public in an easy-to-understand format. Otherwise, it will be difficult for the public to feel secure, even with a lower regulation level.

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