Chapter 1
An Overview of Our Research

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Abstract Immediately after the Fukushima nuclear plant accident (FNPA), 40–50 researchers at the Graduate School of Agricultural and Life Sciences, the University of Tokyo, analyzed the behavior of the radioactive materials in the environment, including agricultural farmland, forests, rivers, etc., because more than 80% of the contaminated land was related to agriculture. Since then, a large number of samples collected from the field were measured for radiation levels at our faculty. A feature of the fallout was that it has hardly moved from the original point contaminated. The fallout was found as scattered spots on all surfaces exposed to the air at the time of the accident. The adsorption onto clay particles, for example, has become firm with time so that it is now difficult to be removed or absorbed by plants. ^{137}Cs was found to bind strongly to fine clay particles, weathered biotite, and to organic matter in the soil, therefore, ^{137}Cs has not mobilized from mountainous regions, even after heavy rainfall. In the case of farmland, the quantity of ^{137}Cs in the soil absorbed by crop plants was small, and this has been confirmed by the real-time imaging experiments in the laboratory. The downward migration of ^{137}Cs in soil is now estimated at 1–2 mm/year. The intake of ^{137}Cs by trees occurred via the bark, not from the roots since the active part of the roots is generally deep within the soil where no radioactive materials exist. The distribution profile of ^{137}Cs within trees was different among species. The overall findings of our research is briefly summarized here.

Keywords ^{137}Cs · Fukushima nuclear plant accident · Agriculture · Soil · Plant · Forest

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1.1 General Features of the Fallout

When the fallout from the nuclear power plant between Fukushima and Chernobyl was compared, the total radioactivity released into the environment by FNPA was estimated at 770,000 TBq, which is approximately 15% of that released by the Chernobyl accident. The radioactive nuclides released by FNPA contained 21% ^{131}I (half-life: 8 days), 2.3% ^{134}Cs (half-life: 2 years) and 1.9% ^{137}Cs (half-life: 30 years). The remaining nuclides in the environment are ^{134}Cs and ^{137}Cs, and the ratio of which has changed from roughly 1:1 in 2011 to 0.12:1 in 2017.

Since the accident occurred in late winter, the only crop in the fields was wheat. The relevant feature, with regards to the fallout, is that the radioactive Cs remained at the initial contact site and had not moved since, therefore, this would imply that Cs will be difficult to remove from fields. When the radiograph of any materials exposed to the air at the time of the accident was taken, the contamination was found as scattered spots on all the surfaces investigated, including soil particles and plant material.

Today, there are no contaminated agricultural products on the market, and researchers are starting to turn their attention to the situation in the forests. At the time of the accident, most of the radioactive material was trapped in leaves located high in the evergreen trees and in the bark of these trees; therefore, the radioactivity was relatively low on the forest floor. In the past few years the contaminated leaves of evergreen trees have fallen to the ground and along with the decomposition process of the litter, ^{137}Cs has gradually moved to the soil and become firmly adsorbed by soil particles.

Since most of the radioactive Cs is adsorbed to fine clay or organic matter in the soil, radioactivity was not detected in the water itself flowing out from the mountain. A simple filtration of the water was effective to remove the radioactive fine particles suspended in water. In the forests, no biological concentration of ^{137}Cs was found in any specific animal along the food chain.

1.2 Radioactivity Measurement

Most of the radioactivity measurement and imaging was performed by the Isotope Facility for Agricultural Education and Research in our faculty. Two academic faculty members and two technicians continue to measure all samples collected from the field, as well as samplers generated from laboratory experiments. Approximately 300 samples are measured per month using two Ge counters and several hundred samples are measured using a Na(Tl)I counter with an automatic sample changer (Figs. 1.1 and 1.2). The number of the samples measured in each month does not mean the number of the samples actually collected. The number of the sample is dependent on the activity level of the sample collected, since when the radioactivity level of the sample is low, it required a longer time of the measurement, therefore, only small number of the sample was able to measure in the month.

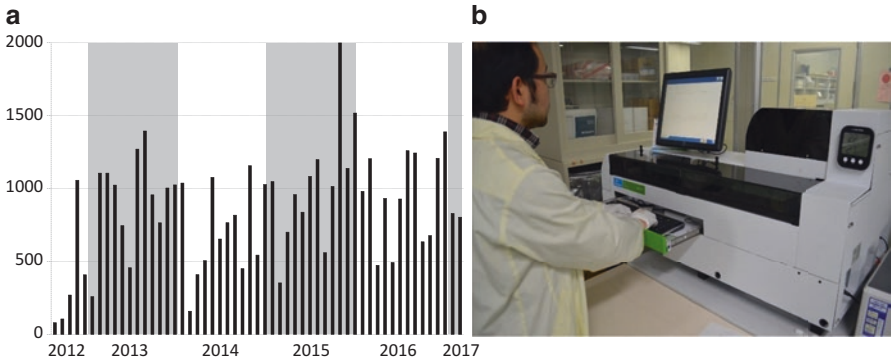

Fig. 1.1 Radioactivity measurement of samples. (**a**) The number of samples measured each month using a Na(Tl)I counter. (**b**) Picture of the Na(Tl)I counter

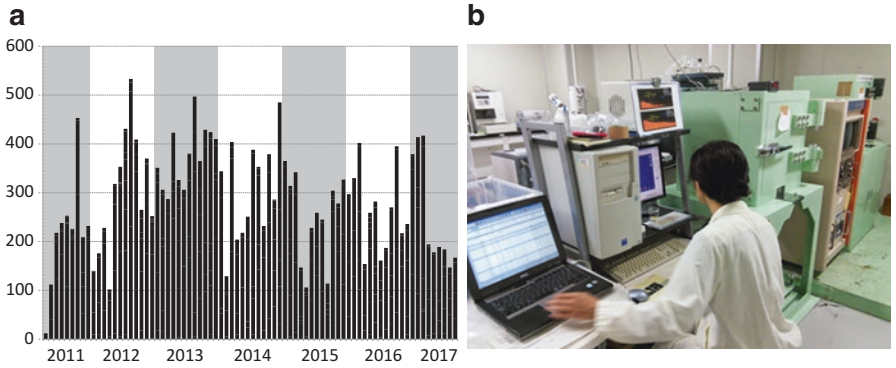

Fig. 1.2 Radioactivity measurement of samples. (**a**) The number of samples measured each month using a Ge counter. (**b**) Picture of the Ge counter

1.3 A Brief Summary of Our Findings

1.3.1 Soil

1.3.1.1 Vertical Migration of Radiocesium

To measure the vertical migration of radiocesium in the soil, a vinyl chloride cylinder was placed in a borehole in the soil. A scintillation counter covered with a lead collimator with a slit window was inserted into the pipe to measure the radioactivity vertically along the borehole. About 2 months after the accident, the vertical radiocesium (^{134}Cs and ^{137}Cs) concentration in the top 0–15 cm layer of soil was measured in an undisturbed paddy field. Approximately 96% of the radiocesium was found within the top 0–5 cm layer. The measurement was repeated every few

months to record the downward movement of the radiocesium. The radiocesium movement in soil was very fast during the first 2–3 months and then the speed was drastically reduced, indicating that the adsorption of the radiocesium to soil particles had become stronger with time, indifferent to the amount of rainfall. The speed of the downward movement of the radiocesium is now much slower than immediately after the accident, at about 1–2 mm/year.

The radioactivity of the surface soil at the bottom of a pond was measured periodically. Radioactivity had gradually decreased with time, except for one pond, where run-off water from the city flowed into the pond. Water had been used to decontaminate concrete and other surfaces in the city after the accident, and radiocesium in this contaminated water had moved to the bottom of the pond.

1.3.1.2 ^{137}Cs Adsorption Site

One study determined where radiocesium is adsorbed on soil. The soil was separated according to particle size and an autoradiograph of each fraction was taken. It was found that radiocesium was adsorbed by the fine clay and organic matter but not by the larger components of the soil, such as gravel and sand. It was an important finding which led to the development of an efficient and practical decontamination protocol for farmland.

To determine the kind of clay mineral adsorbing the radiocesium, eight mineral species were prepared and an adsorption/desorption experiment was carried out using a small quantity of ^{137}Cs tracer. It was found that the weathered biotite (WB) sorbed ^{137}Cs far more readily and firmly than the other clay minerals. The WB sample was then cut into pieces by a focused ion beam and the radioactivity of each piece was measured by an imaging plate to know the distribution of ^{137}Cs. To our surprise, each separated fraction of WB showed similar radioactivity per area, suggesting the uniform distribution of radiocesium within the clay piece. This finding has completely changed our understanding of the adsorbed site of clay minerals. The layered shape of the clay was reported to have a loosed edge due to weathering, known as a frayed edge site, where ^{137}Cs was selectively fixed, therefore, ^{137}Cs was expected to be fixed at the margin of the clay. It was also suggested that the adsorption behavior of ^{137}Cs was different when the quantity was very small, i.e. radiotracer level.

1.3.1.3 ^{133}Cs and ^{137}Cs

To compare ^{137}Cs distribution with that of ^{133}Cs, which is a stable nuclide, an agricultural field (3.6 × 30 m) in Iitate-village was selected. In this field, the total radiocesium activity was 5000 Bq/kg, which corresponded to about 10^{-3} μg/kg of ^{137}Cs, whereas the concentration of stable ^{133}Cs was about 7 mg/kg. The stable ^{133}Cs

was derived from the minerals in the field and the ^{137}Cs was derived from the fallout. Though there was a high correlation between total ^{137}Cs distribution and that of exchangeable ^{137}Cs, it was found that the extraction ratio, exchangeable ^{137}Cs/total ^{137}Cs, was higher than that of stable Cs. Since this extraction ratio is expected to be the same between stable and radioactive Cs in the future, when equilibrium is attained, the higher extraction ratio of radioactive Cs suggested that the exchangeable radioactive Cs is still moving toward the stable state, which could be interpreted that the fixing process of fallout nuclide is still proceeding.

1.3.2 Plants

1.3.2.1 Rice and Soybean

To study the transfer factor of ^{137}Cs from soil to rice, the relationship between the radioactivity in the soil and that in plants was measured. But there was a reciprocal correlation between the K concentration in soil and ^{137}Cs concentration in plants, which suggested that applying K to soil prevents rice from becoming contaminated. Actually, when an optimum amount of K was not supplied in K deficient fields, radiocesium content in rice plants was high. Although the natural abundance of Cs compared to K is only 1/1000, it was interesting that the competition between the two ions in plant absorption was observed.

When a single grain of rice was sliced and placed on an imaging plate to obtain the radiograph, it was shown that ^{137}Cs accumulated in both the hull and in the cereal germ of the grain. To study the distribution of the radioactive Cs in more detail, the micro-autography method was employed which was developed by our faculty. After slicing the grain, the film emulsion was painted on the surface of the glass to produce a thin film. The film was exposed to radiation from the sample and then developed to obtain a micro-radiograph. Examining the micro-radiograph under a microscope showed that ^{137}Cs accumulated around the plumule and radicle, suggesting that Cs was not incorporated into the newly developing tissue itself but accumulated in the surrounding tissue of the meristem, similar to the phenomenon that the meristem is generally protected and free from heavy metals or viruses.

Radiocesium accumulation in soybean seed tends to be higher than that of rice grain. One of the reasons is that a soybean seed does not have an albumen. In the case of a rice grain, radiocesium accumulates in the embryo and not in the albumen. The soybean seed itself develops into a cotyledon, a kind of embryo, therefore it contains a high amount of minerals. Another difference between rice and soybean plants is that, in the case of a rice, radiocesium absorption occurs before ear emergence and out of the total radiocesium amount absorbed, 10–20% was accumulated in the seeds. However, in the case of a soybean plant, half of the radiocesium accumulated in the seed is taken up during pod formation, and out of the total radiocesium absorbed about 42% accumulated in the seeds.

1.3.2.2 Fruit Trees

Generally, in the case of the trees, radioactive Cs moved directly from the surface of the bark to the inside of the trunk. To understand how radioactive Cs is transferred to the inner part of a fruit tree in the following season, a contaminated peach tree was transplanted to a non-contaminated site after removing twigs, leaves and fine roots. Then, 1 year later, all of the newly developed tissue, including the fruits, was harvested and the radioactivity was measured. Only 3% of the radioactive Cs had moved the following year to the newly developing tissue, including roots. That means that 97% of the radioactive Cs that had accumulated inside the tree did not move. In the case of the fruit tissue, about 0.6% of the radioactive Cs that accumulated inside the tree had moved and accumulated in the fruit.

1.3.3 Forests and Animals

1.3.3.1 Forests

In the mountain forests, leaves were only present on evergreen trees and these needle-like leaves were highly contaminated due to the fallout because the accident occurred in late winter. However, even these needle-like leaves received high amounts of radioactive material and prevented the fallout from moving to the forest floor. Therefore, the radioactivity of the soil under the deciduous trees without leaves was higher than the soil under the evergreen trees. In the case of the evergreen trees, leaves located higher on the trunk of the trees were more contaminated than those located lower on the trunk and the trunk itself was highly contaminated. Though the amount of radioactivity moved into the heartwood was different along the height of the tree, the contamination inside the tree was not due to the radioactive Cs transport from the roots. Since the radioactive Cs was only at the surface of the soil, it was not possible for the active roots to absorb Cs. The active part of the roots for most trees is at least 20–30 cm below the surface of the soil and at this depth, there was no radioactive cesium. In the past few years the contaminated leaves of evergreen trees have fallen to the ground and along with the decomposition process of the litter, ^{137}Cs has gradually moved into the soil and then firmly adsorbed by soil particles.

Mushrooms can be found growing in forests in mountainous regions all over Japan, however, the radioactivity of the mushrooms growing in the forest has not drastically decreased with time. Some of the mushrooms harvested more than 300 km from the site of the accident were found to accumulate ^{137}Cs only, indicating that they are still accumulating the global fallout from nuclear weapons testing that occurred during the 1960s. Since the half-life of ^{137}Cs is 30 years, it is much longer than that of ^{134}Cs (half-life of 2 years), all of the ^{134}Cs in the global fallout in the 1960s has decayed after 50 years. This means when only ^{137}Cs was detected in mushrooms, the ^{137}Cs found was not from the Fukushima nuclear accident. In the

case of the fallout from the Fukushima nuclear accident, the initial radioactivity ratio of ^{137}Cs to ^{134}Cs was the same in 2011.

The river water flowing from the mountains show very low radioactivity (less than 10 Bq/l). It was also found that the water itself flowing out from the mountain had low radioactivity and the radioactivity was removed after filtering out the suspended radioactive clay in the water. The amount of the radioactive Cs flowing out from the mountain was in the order of 0.1% of the total fallout amount per year.

1.3.3.2 Animals

Contaminated haylage was supplied to dairy cattle and the radioactivity of the milk was measured. It was found that radioactive Cs was detected in the milk soon after the contaminated feed was supplied. After radioactivity levels in the milk reached a plateau after 2 weeks, the non-contaminated feed was fed to the cattle and the radioactivity in the milk decreased and became close to the background level after 2 weeks. Similar results were found for animal meat, indicating that when contaminated animals are identified, it is possible to decontaminate them by feeding non-contaminated feeds. The biological half-life of ^{137}Cs was estimated to be less than 100 days because of the animal's metabolism, whereas the physical half-life of ^{137}Cs is 30 years.

At the time of the accident, radioactive Cs contaminated every surface exposed to the air, and this also included the feathers of birds. Male bush warblers were captured in a highly contaminated area of the Abukuma highlands in 2011, and it was found that the feathers were contaminated with ^{137}Cs. The accident occurred just as these birds had started molting, therefore, they had a limited home range in the highlands, which was close to the site of the accident. This contamination of feathers was not removed by washing. However, in the following year, no radioactivity was found on the feathers of the bush warbler caught in the same area.

1.4 Decontamination Trial

The most effective and efficient way to prevent radioactive Cs uptake in crops is to apply K fertilizer on farmland. Since the soil in agricultural land is a very important natural resource, the removal of the soil surface cannot be compensated by simply replacing it with non-contaminated soil. The best way to decontaminate farmland is to eliminate only the contaminated particles in the soil. Radioactive Cs was only found to be adsorbed firmly on the fine clay component of soil. Therefore, introducing water into a contaminated field and mixing it well with the surface soil (about 5 cm in depth), the soil components precipitate and the suspended fine clay particles in the water can be drained off into an adjacent ditch in the field. Thus, more than 80% of the radioactivity in the field was removed.

1.5 Conclusion

The behavior of the radioactive Cs emitted from the nuclear accident was different from that of so-called macroscopic Cs chemistry we know. Because the amount of Cs deposited on leaves was so small and carrier-free, the nuclides seem to behave like radio-colloids, or as if they were electronically adsorbed onto the tissue.

Through our activities, many scientific findings have been accumulated. The results of our research introduced above are only a small portion of our total findings since the Fukushima nuclear accident occurred.

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