

Chapter 8

Radioactivity in Agricultural Products in Fukushima

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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

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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 radio-caesium 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 (*Spinacia 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			Total
	March	April	May	June	July	August	September	October	November	December	January	February	March			
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

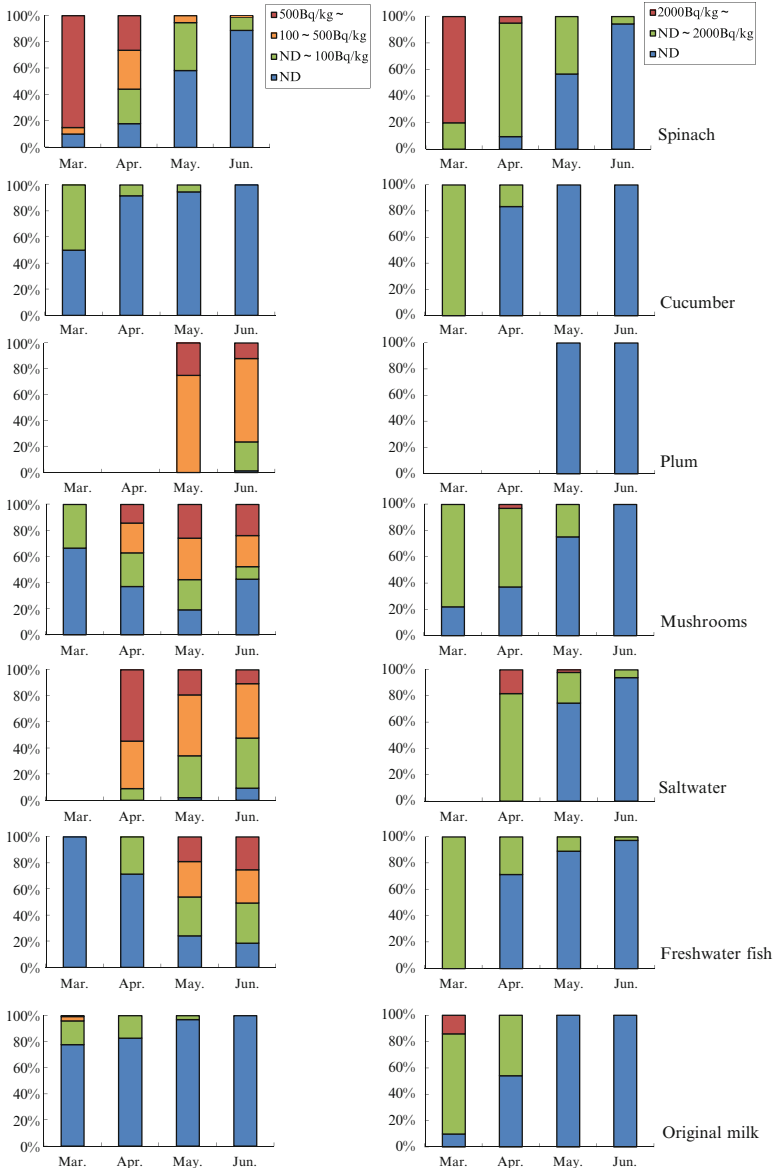


Fig. 8.2 Radioactive material concentrations in different products (from March 2011 to June 2011). *Left figures:* radiocesium; *right figures:* radioiodine

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 (*Plecoglossus 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,

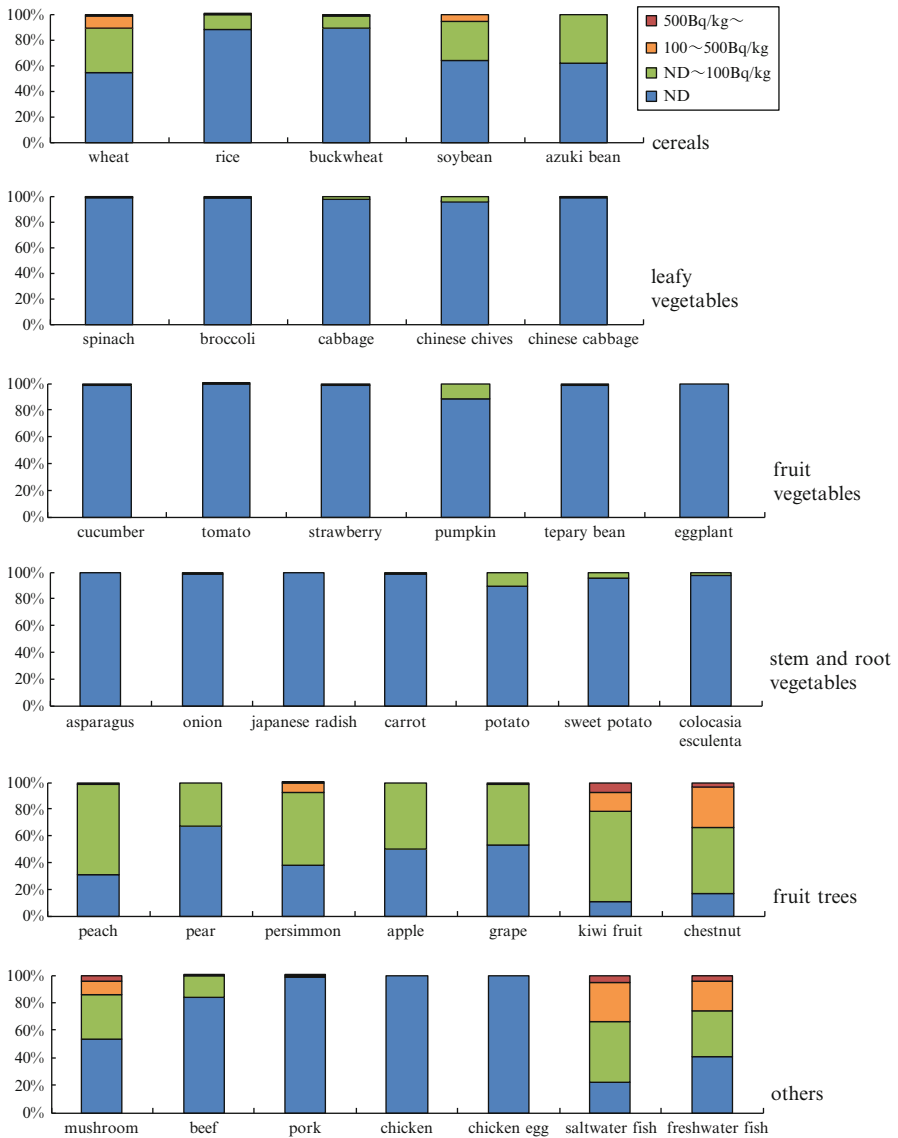


Fig. 8.3 Radiocesium concentrations in different products (from July 2011 to March 2012)

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 kenoei*), 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 leucomaenis*), seama (*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.

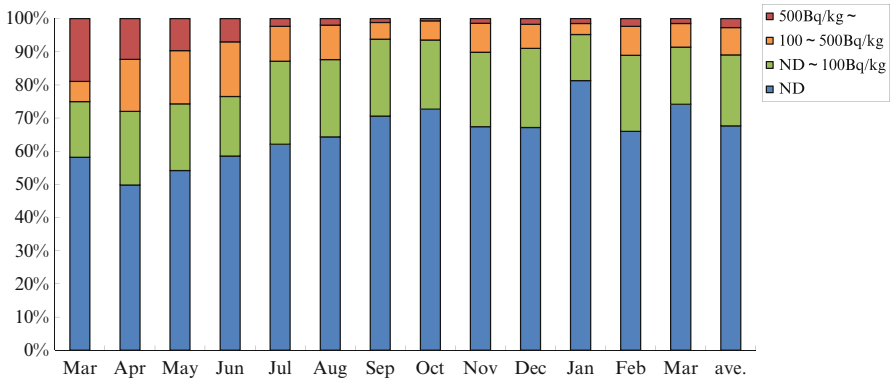


Fig. 8.4 Radiocesium concentrations in all monitoring samples

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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