Large-scale sampling and radioactivity analysis of agricultural soil and food during nuclear emergencies in Japan: Variations over time in foodstuffs inspection and sampling

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Abstract

The measurement of radioactivity in food and agricultural ecosystems is an essential task for keeping the population safe after a nuclear emergency. Prior to the Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident in 2011, the probability of such an accident, combining complex effects of natural and technical factors, was not reflected in detail in national emergency preparedness guidelines. The lack of such guidelines resulted in a limited efficiency response to the Fukushima accident in agriculture. This outcome showed a need for the preparation of such guidelines as a part of emergency preparedness for nuclear and radiological emergences. This paper provides information and generic, non-country-specific guidance on approaches to sampling food. The paper is intended for scientists, policy makers and decision makers involved in nuclear emergency preparedness and responses, particularly on large scales and at different stages of nuclear emergency based on lessons learned from the FDNPP accident.

Key words: Agriculture, Foodstuffs, Monitoring, Policy making for food import/export

1. Introduction

The continuation of agricultural activity in areas affected by man-made radionuclides requires the implementation of radiation safety standards and regular radiation inspection and monitoring.

These standards and requirements are based on relevant national legislation: they also provide for dose reference levels and derived reference levels for radionuclide concentrations in foodstuffs to be applied for managing the safety of foodstuffs following large-scale contamination of agricultural land.

After the Chernobyl accident, such standards, called temporary permissible levels (TPLs) of radionuclides in foodstuffs, were implemented at both the national and international levels (Alexakhin et al., 2007; Balonov et al., 2018). The national standards, implemented in the most affected countries, such as Belarus, Russia and Ukraine, were stricter than the international standards because they assumed that all food produced in the affected regions was consumed locally. These TPLs, adjusted considering changes in radiological situations and economic constraints, played the same role as the reference levels suggested by the International Commission on Radiological Protection (ICRP) in 2007. These temporary permissible levels for radionuclides in food were implemented in the Chernobyl-affected countries not only for the inspection of radionuclide concentrations in the foodstuffs but also to encourage the producers to apply maximum efforts in the reduction of radionuclide concentrations in foodstuffs (Balonov et al., 2018). The corresponding guideline levels for radionuclides in foods were also implemented for international trade by the FAO/WHO Codex Alimentarius Commission (Codex Alimentarius Commission, 2006).

After the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, large amount of radionuclides were dispersed and contaminated a large area of eastern Japan including agricultural field. The main radionuclides were ^{131}I, ^{134}Cs and ^{137}Cs, and during the very early initial stage of contamination ^{131}I was most worried, then the radiocaesium become in concern because of its rather longer half-life. Plants were suffered by radionuclide by direct fallout at first then transfer from soil become dominant. Though the transfer of radionuclide from soil is regulated by the properties of soil and plant, environmental contamination level had a decisive influence (Shinano et al. 2020). To avoid internal contamination by these radionuclides, Provisional Regulation Values for both radioiodine and radiocaesium were set by the Ministry of Health, Labour and Welfare (MHLW) on 17 March 2011 under the Food Sanitation Act by adopting criteria from the National Security Council (NSC) Nuclear Emergency Preparedness Guide (MHLW, 2011 a, b). For radiocaesium, these values were 200 Bq kg^{-1} for drinking water and milk dairy products and 500 Bq kg^{-1} for the all other foodstuffs, such as grain, meat, vegetables and fish. For radioiodine, values of 300 Bq kg^{-1} were adopted for drinking water and milk, while 2,000 Bq kg^{-1} was used for vegetables and some fishery products (MHLW, 2011b). The Provisional Regulation Value was used immediately after the FDNPP accident until 31 March 2012.

On 1 April 2012, Standard Limits restricting food contamination were introduced to replace Provisional Regulation Values. These Standard Limits were set at 100 Bq kg^{-1} for general food, 10 Bq kg^{-1} for drinking water, and 50 Bq kg^{-1} for milk and infant food (MHLW 2019a). Thus, it should be noted that these values are subject to reconsideration based on the dose reference levels, food self-sufficiency and economic constraints.

Regulation values for food safety are important; however, the emergency response systems themselves, which include sampling and measurements of the agricultural soil and foodstuffs, are also very worthwhile to consider. In Japan, sampling of agricultural products is performed based on the Environmental Radiation Monitoring procedure. This guideline was originally published by NSC of Japan in 1984 (NSC, 1984) as the Guideline for Environmental Radiation Monitoring in Emergency; it was updated in March 2008, with the integration of normal and emergency monitoring considerations as the Guideline for Environmental Radiation Monitoring (NSC, 2008).

Manuals for the measurement of radioactivity under normal and emergency situations were published by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) and the Nuclear Regulation Authority (NRA) and were considered a subject for regular updates provided by the working group of Environmental Radiation Monitoring of NRA (MHLW, 2002; NRA, 2016,).

Based on the sampling procedures provided by Operational Intervention Levels (OIL) for Reactor Emergencies (IAEA 2017), the process of assessing a nuclear or radiological emergency for contamination of a large area was focused. For the monitoring of food, milk and drinking water samples, the default OIL values of 1,000 Bq kg^{-1} of ^{131}I and 200 Bq kg^{-1} of ^{137}Cs as marker radionuclides are applied after the end of a major release of radioactive material. Furthermore, 100 mSv year^{-1} is set when early protective actions are taken. It is also mentioned that, once sufficient resources and time become available, the generic criterion of 10 mSv year^{-1} is used with all required radionuclide analyses. In Japan, since it is believed that the required analysis is able to be carried out, more protective actions were undertaken to decide the Provisional Regulation Values.

In January 2014, the NRA published a supplement reference document of the Nuclear Emergency Response Guidelines. This document describes the radionuclide measurements in foods and drinks: the document is used as a standard to decide on intake regulation as quickly as possible under OIL to decide the protective measure indices for intake of foods and drinks under OIL referring to the FDNPP accident (NRA, 2017 in revised version). Since the purpose of monitoring in emergencies is to evaluate the radiological significance of the radionuclide intake with foodstuffs to limit internal exposure, the guidance from MHLW published in 2012 (MHLW, 2012a) was considered sufficient. The guidance recommended that measurement and analyses of the sample be performed within 1 week after the nuclear accident in the framework of emergency monitoring. It was also recommended that areas with an air dose rate (measured at 1 m in height) greater than 0.5 μSv h^{-1} had to be selected for monitoring, including sampling and measurement of radionuclide levels in agricultural products.

The 2011 FDNPP nuclear accident underscored the importance of emergency preparedness for potential large-scale nuclear disasters. This paper describes lessons learned from the response process and decisions made by the Japan national and local governments for large-scale sampling and measurements of radioactivity in agricultural soil and foodstuffs.

2. Inspection and sampling guidelines used in response to the FDNPP accident

2.1 Inspection and sampling guidelines used in response to the FDNPP accident

Before the accident at FDNPP, the regulation values for radioactive materials in food were based on the Food Sanitation Act (MHLW, 1947). Immediately after the accident, MHLW released a Provisional Regulation Value based on index values indicated by NSC in Japan from 17 March 2011 (MHLW, 2011a). The Provisional Regulation Values included radioiodine, radiocaesium, uranium, plutonium and other transuranic elements, which were dedicated to being applied in response to the emergency situation after the nuclear accident and were positioned as "one of the protective measures as a standard to introduce restriction of consumption of food"

The Provisional Regulation Values, which were defined by MHLW (MHLW, 2011a), were based on the upper limit of radiation exposure based on an effective dose of 5 mSv $year^{-1}$ for radiocaesium, uranium, plutonium and transuranic elements and 50 mSv $year^{-1}$ for radioiodine for the thyroid equivalent dose. These limitations of effective doses were discussed by the Food Safety Commission of Japan in March 2011 (Food Safety Commission of Japan, 2011) and were decided to be based on the guidelines from ICRP (1992) and WHO (1988). In the case of radiocaesium, ICRP advised that 10 mSv $year^{-1}$ was an effective dose for each food and drink (ICRP 1992), and the commission suggested 5 mSv $year^{-1}$ as more conservative position. In the case of radioiodine, the thyroid equivalent dose of 50 mSv $year^{-1}$ (2 mSv $year^{-1}$ as an effective dose) was adopted based on the WHO (1988). After 2012, the regulation in Japan moved from the emergency exposure situation to the existing exposure situation. Therefore, temporary levels for radionuclide activity concentrations in the foodstuffs specific to the emergency phase were shifted to the reference levels in foodstuffs accounting for long-term exposure. These levels were provided as new Standard Limits based on the discussion in the Subcommittee on Animal Origin Foods Sanitation (MHLW, 2012a). The new Standard Limits were determined using the list of radionuclides released into the environment after the accident (MEXT, 2011a), which included radionuclides with half-lives of more than one year, i.e., ^{134}Cs, ^{137}Cs, ^{90}Sr, $^{238,239,240,241}Pu$, and ^{106}Ru. Not all of the radionuclides were determined separately, and ^{106}Ru was estimated based on the relative ratio of ^{106}Ru to ^{137}Cs based on estimations made by NISA (NISA, 2011). The sum of Pu isotopes was based on the actual measured values provided by MEXT and was calculated based on the relative ratios of $^{239,240,241}Pu$ to ^{238}Pu as reported by the "Working Group to Consider Countermeasures against Radioactive Materials" of the "Subcommittee on Animal Origin Foods Food Sanitation Committee, MHLW" (MHLW, 2012a). In the new regulation, it was assumed that radiocaesium is a reference radionuclide, and its concentrations reflect other radionuclides listed above based on the relative concentration of each radionuclide in the environment (MHLW, 2012 b,c). Since the short half-life of radioactive ^{131}I could not be detected after one year, and the concentrations of uranium isotopes were similar to the background levels, these radionuclides were removed from the regulation. Based on these new standards, the document "Concepts of Inspection Planning and the Establishment and Cancellation of Items and Areas to Which Restriction of Distribution and/or Consumption of Food Concerned Applies" was revised (MHLW, 2012e). For the actual inspections, the target municipality and agricultural products required to be checked based on the inspection program (Concepts of Inspection Planning and the Establishment and Cancellation of Items and Areas to which Restriction of Distribution and/or Consumption of Foods concerned Applies) (MHLW, 2011b). Since then, the concept has been updated 9 times through the end of 2017 (MHLW, 2011d, h, 2012c, d, 2013, 2014, 2015, 2016, 2017b). MHLW paid major attention to ^{131}I at the initial stages since a large number of products were reported to be contaminated by ^{131}I (MHLW, 2011b). The policy was revised to change the focus of foodstuffs inspection from radioiodine to radiocaesium on 27 June 2011, and tea, marine products, wheat and barley were added at the same time (MHLW, 2011d). Beef and rice were added on 4 August 2011 (MHLW, 2011e), while soybeans and buckwheat were included in the regulation on 12 July 2012 (MHLW, 2012f). On 19 March 2013, regulatory limits were introduced for cultivated mushrooms with cut logs (MHLW, 2013). In the documents above, not only the list of monitored radionuclides but also the conditions for the termination of foodstuffs inspection were prescribed.

The frequency of sampling was approximately once per week, and the inspection area was determined by the municipality by dividing each prefectural area into several areas and each municipality being responsible for sample collection. The obligation to label origins in prefectural areas is required for the traceability of agricultural products in Japan (Japan Agricultural Standards, JAS). The traceability was based on the units of prefectures, but it also allowed for separation of the prefectural area into several subunits if monitored by prefectures and/or municipalities.

The Prime Minister, who was the head of the Nuclear Emergency Response Headquarters (NERH) at the time, issued a directive for the formulation of a monitoring protocol for agricultural produce and lands in the Fukushima, Ibaraki, Tochigi and Gunma prefectures, the surrounding municipalities (Miyagi, Yamagata, Niigata, Nagano, Saitama and Chiba prefectures), and the municipalities where concentrations of radionuclides in agricultural products exceeded the regulation value (i.e., Tokyo on 4 April 2011 (MHLW, 2011b), Kanagawa, Yamanashi, and Shizuoka prefectures on 27 June 2011 (MHLW, 2011d), and Iwate, Akita, and Aomori prefectures on 4 August, 2011(MHLW, 2011e)). Spinach, crown daisy, Kakina (a variety of *Brassica napus*), Potherb mustard, *Brassica rapa* var., milk and other major products were selected to be reference food items and had to be monitored regularly.

The food and drink intakes regulation levels were defined after specific radionuclides were selected as contamination representatives, based on the actual contamination level of food items. As a first step, conventional screening measurements using survey meters were requested. In the second step, accuracy became more important than urgency in the data provision, and a broad range of food items were analysed in laboratories and used to assess the internal exposure of the public (MHLW, 2002).

Monitoring was initially performed by each prefecture and supported by the government at the time of the accident. With revision of the nuclear emergency response guidelines (NRA, 2013), it was changed to a system in which the government supervised each prefectural monitoring system and categorized the area into a Precautionary Action Zone (PAZ), an Urgent Protective Action Planning Zone (UPZ), and a Plume Protection Planning Area (PPA). It is generally considered that the PAZ is within a 5-km radius, the UPZ is within a 30-km radius, and the PPA is where protection is required outside a 30-km radius from the nuclear facilities, enabling the monitoring of larger areas beyond the prefectural boundaries.

2.2 Collection of samples for foodstuffs inspection

Food sample analyses were performed by the national food safety agencies, the Fukushima prefecture, and other municipalities. The data collected were published by MHLW (MHLW, 2019b)[1].

Sampling of agricultural products during the emergency exposure situation was conducted based on the Manual for the Measurement of Radioactive Materials in Food in Emergency (MHLW, 2002). Samples are pre-treated before measurement based on the Manual for Radioactivity Measurement Series 24 (NRA, 2019-originaly published by MEXT in 1992). In this manual, vegetables are

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[1] Regarding the survey results of radionuclide activity concentrations in agricultural produce, more than 1,000 reports have been published, and this activity was ongoing in 2019.

recommended to be cut into small pieces before measurement. To protect against cross-contamination among samples, the user is instructed not to wash leafy vegetables with water.

The pretreatment of food before measurement and pre-washing methods for several vegetables were added to the instruction manual (MHLW, 2012d). After a substantial amount of soil was removed manually after collection of the sample, the samples were washed based on the category of the foodstuff properties, such as rinsing the sample for approximately 20 seconds under running tap water and gently wiping the surfaces of the samples with wet paper towels. The details of the washing procedures according to the category of food are described in the attachment from MHLW (2012d).

Grid basis sampling is ideal for environmental sampling, but it is considered unsuitable for foodstuffs sampling because most food is produced under the control of producers. Therefore, sample collection was performed by the agricultural authority of the local government and/or agricultural cooperatives in the area. This process allowed for sampling with traceability of the samples obtained from the field to the producer or to the food distributor levels.

The analytical flow of the sampled agriculture, forestry and fishery products is shown in Fig. 1.

Fig. 1 is about here

2.3 Sampling of arable soil

Fukushima prefecture used the guiding document of Radioactivity Measurement Series No. 16 (MEXT, 1983) for the sampling of farmland soil (Sato, 2014). In this method, depending on the depth of the topsoil, the default depth for sampling soil was set to 20 cm. However, since the determination of topsoil can represent a complex issue, and modern agricultural machines used in Japan are normally set to cultivate the soil at up to 15 cm, the depth of the soil for analysis was set to 15 cm.

The National Institute for Agro-Environmental Sciences (NIAES) released a guide (NIAES, 2011, see Supplementary material 1), describing in detail the sampling method for farmland soil in correlation between soil contamination and air dose rates.

Since the reference topsoil depth used in Japan is not similar to that assumed in other countries, it was suggested that the sampling method, as well as sampling depth, be re-investigated for use in case of future emergencies.

In 2011, two soil surveys for radioactivity were performed twice. The first soil examination (emergency survey) was performed from 31 March to 5 August (Table 1) by Fukushima Agricultural Technology Centre (FATC) for 371 sites, and the second sampling round was conducted from 24

October to 3 February 2012 (Table 2). The second soil survey was a detailed investigation, and 2,247 sites were carefully studied by NIAES, FATC, the Agriculture and Forestry Office of Fukushima prefecture, and two private companies contracted by NIAES. In this report, the sampling method was modified to obtain samples from depths of up to 15 cm. In some cases, the depth was less if there was some soil disturbance by rocks or other materials.

In Fukushima prefecture, soil sampling after the accident was divided into two phases.

- Before 27 April 2011, soil was obtained as a square shape sample from the soil profile. Soil was sampled as a 50 x 7 cm rectangle, with 15 cm in depth using a shovel.
- After 27 April 2011, modern soil samplers (Hand sampler HS-30, Fujiwara Seisakusyo) were received for soil sampling, and soil was sampled from a circle with a diameter of 5 cm with 15 cm in depth.
- Five samples were obtained from a field and well mixed. In total, 1 kg of soil was collected for the further analysis. The humidity of the soil was also measured.

Table 1 is about here

Table 2 is about here

3. Sampling and inspection in Fukushima after nuclear accident

3.1 Survey in emergency of agricultural product sampling

In 2011, Fukushima prefecture initiated inspections of brown rice for radioactivity for rice batches as follows: 1) early shipments of rice from 25 August to 9 September; and 2) normal shipments of rice from 8 September to 12 October, 2011 (Crop Production Division of Fukushima prefecture, 2013). In cases of early shipment of rice, the fields requiring examination were identified based on previous assessments, and radiocaesium concentrations in brown rice from each field were measured. The number of such inspections was 101, and the maximum radiocaesium concentration measured in the rice was 41 Bq kg^{-1}.

For normal shipments of rice, two-stage inspection was performed, namely, preliminary rice examination and main inspection. Selection for the preliminary inspections was based on environmental conditions, such as fields with relatively higher air dose rates and/or closer to the forest. If the measured concentration of radioactivity in brown rice was found to exceed 200 Bq kg^{-1} after the preliminary inspection, main inspections were performed.

During the preliminary rice examination, the inspection was performed based on 3 categories listed in Table 3 and Fig. 2, namely: 1) radiocaesium concentrations of agricultural fields more than 1,000 Bq kg^{-1}; b) air dose rates exceeding 0.1 μSv hr^{-1}; and c) selected cities, towns and villages in Fukushima prefecture. Representative samples were collected from the fields approximately one week before the harvest. The collected samples were threshed, dried, conditioned for the analysis and measured for radioactivity. Based on this inspection, 449 samples were analysed.

Table 3 is about here

If the radiocaesium in the samples exceeded 200 Bq kg^{-1}, the municipality that produced the rice was selected as a priority investigation zone. The data derived from the main inspections are summarised in Table 4. During the main inspection, 1,174 samples were investigated, and the maximum concentration of radiocaesium observed in the samples was 470 Bq kg^{-1}.

From the evaluation of 1,724 samples for radiocaesium concentrations in rice, Fukushima prefecture decided on 12 October 2011 that rice in all areas where production was allowed in 2011 could be shipped. However, on 16 November 2011, brown rice with radiocaesium greater than the Provisional Regulation Value was found in one settlement: Oguni village (one of the former municipalities in Fukushima, Fig. 2). On 16 November, Fukushima prefecture initiated a self-imposed restriction on the shipment of rice from the area indicated by NERH, which was declared for shipment termination on the next day.

Emergency inspection for rice started 3 months later (by 3 February 2012) and covered 129 areas in 29 municipalities. The target areas of inspection were: 1) former Oguni village; 2) the area including Specific Spots Recommended for Evacuation; and 3) where radiocaesium was detected during monitoring surveys. The shipment was claimed for self-restraint by the end of inspection. Based on the data of the emergency inspection, shipment suspension was declared for 9 former municipalities in 3 cities where concentrations of radiocaesium in the samples exceeded 500 Bq kg^{-1}. The areas where concentrations of radiocaesium in the samples exceeded 100 Bq kg^{-1} were requested to implement a self-imposed restriction on the shipment of contaminated produce. Through these measures, approximately 17,000 tons of contaminated brown rice were banned for shipment and disposed by specific waste disposal measures. In 2011, MAFF decided to restrict the production of brown rice based on the concept by NERH (2011), and the actual limitation zone was all of the Evacuation Zone, Planned Evacuation Zone and Emergency Evacuation Preparedness Zone (MAFF 2011a). However, based on the distribution of the area where the exceeding brown rice was produced in 2011, the limitation area was enlarged from 2011 to 2012, as shown in Fig. 3 (MAFF, 2013a).

Table 4 is about here

Fig. 2 is about here

Fig. 3 is about here

In Fukushima prefecture, a survey was conducted out by the Environmental Preservation Agriculture Division of Agriculture, Forestry and Fishery Department of the Fukushima Prefectural Government. Overall, monitoring of agriculture, forestry and fishery in emergency in areas affected by the Fukushima accident was based on the Prefectural Monitoring Implementation Policy (Fukushima prefecture, 2019a) and sampling standards according to approved documentation (Fukushima prefecture, 2019b). This monitoring was carried out for the purposes of: 1) verification of the effect of radioactive materials on the quality of agricultural products and confirmation of food safety for shipment and selling; and 2) obtaining precise information about the radiocaesium activity concentrations in the foodstuffs intended for consumers. For this purpose, the prefecture used 3 types of instruments as follows.

1) For monitoring investigations, Ge semiconductor detectors were used for:

1-1) Examination of shipment confirmation, performed on major foodstuffs produced in the Fukushima prefecture if the amount of production was large;

1-2) Inspection for cancellation of restrictions of agricultural products in an area; and

1-3) Examination of intensive monitoring data based on the directives of the director of Agriculture, Forestry and Fishery products division[2].

2) For preliminary confirmation of food safety, Ge semiconductors or simplified analytical instruments (e.g., NaI(Tl) scintillation spectrometers) were used.

2-1) Inspection for cancellation of food distribution restriction was performed before implementation of procedure 1-2) was performed by the prefecture.

2-2) Inspection for the cancellation of harvesting restriction was performed.

Simplified analytical instruments placed in prefectural organisations are listed in Table 5 (Fukushima prefecture, 2019a).

[2] Notice by director of Agriculture, Forestry and Fishery products division, 1 September 2014. Last revision was 28 May 2015). “About the reinforcement of the examination for radiological material of agriculture and forestry marine products when radiological scattering”

3) Quick inspection using simplified analytical instruments.

In the restricted areas under the control of the Agriculture and Forestry office and/or agricultural promotion sectors, where quick decisions for shipment of the food items were required, simplified analytical instruments were used instead of the procedure indicated in 1-1.

4) Voluntary inspections (Additional to prefectural monitoring)

The inspections performed out by municipalities and associations for agricultural products and food safety were identified as voluntary inspections using an implementation system for monitoring, as shown in Table 6 (Fukushima prefecture, 2019a). Fukushima prefecture cooperated with the organisations implementing these voluntary inspections and reported the results based on “Safe Confirmation System of Agriculture, Forestry and Fishery Products of Prefecture” (Notice by director of Agriculture, Forestry and Fishery products division, 14 September 2011. Last revision was 29 March 2016).

Table 5 is about here

Table 6 is about here

3.2 Implementation of food inspections

The Agriculture and Forestry Office, in addition to the municipal governments, were the main coordinators of systematic sampling for food inspections. The analyses for radioactivity were performed by the analytical division of FATC and by private organisations. For the effective confirmation of the safety of agricultural products in Fukushima prefecture, NaI(Tl) scintillators and germanium semiconductors were used for the initial confirmation inspections. Quick inspections using simplified analysis instruments were also performed near the production area.

3.2.1 Preparation of monitoring plans and sample collection

The annual monitoring plan was prepared by the Environmental Preservation Agriculture Division. Each section (i.e., Agriculture, Forestry and Fishery Department, Nature Conservation Division, and Food Safety and Environmental Hygiene Division) in the Fukushima prefecture prepared specific sampling plans for 3-month periods. The plans were prepared for March, June, September and December timeframes and considered emergency situations, as well as the ability of the sector involved to measure samples for radioactivity.

Special attention was paid to crop variety, cropping type, shipment time, and area planted as follows:

- Major agriculture, forestry and fishery product of the prefecture and large amounts of production and shipment;
- Products with long harvesting times, for which continuous analyses are required;
- Key products promoted as part of regional development efforts; and
- Products consumed in large quantities.

3.2.2 Sample collection and transport

Concerned sectors (e.g., Agriculture and Forestry office) defined the sampling places after coordinating the sampling plan with municipalities. Personnel (e.g., Agriculture and Forestry office staff) performing sampling and sample transportation for the radioanalytical laboratories were requested to make all prior arrangements to expedite sample delivery.

3.2.3 Sector analysing foodstuffs for radioactivity in Fukushima prefecture

As a rule, analyses for radioactivity were performed by the prefectural Agriculture, Forestry and Fishery department (mainly by FATC). In cases of large numbers of samples for inspection using the germanium semiconductor detector, a long time for analysis was expected, and there were other resource constraints. The samples were also measured in the Agricultural Technology Centre and by some private companies.

3.2.4 Evaluation of the analytical result

If the radiocaesium activity concentrations in the foodstuffs measured for shipment exceeded the regulation values, the prefecture requested the municipalities, producers, etc., for self-imposed restrictions on the shipment of the food items.

To respond to public concerns, the prefecture published the results of the monitoring of radioactivity in foodstuffs. These results are presented according to former municipality subdivision and for large villages. In the case of fisheries, the data are presented for sea areas and rivers.

If the concentration of radiocaesium in the foodstuffs was suspected of exceeding the Standard Limit during the quick inspection using an NaI(Tl) scintillation spectrometer, more precise measurements with the aid of a germanium semiconductor were obtained. The overall monitoring workflow provided by the Environmental Preservation Agriculture Division of Fukushima prefecture (Fukushima prefecture, 2018) is outlined in Fig. 4.

Fig. 4 is about here

3.3 Collection of monitoring data by independent municipality inspection

Those foodstuffs unrelated to the prefectural monitoring network examination were also carried out by the municipalities, the producer groups, and the Fukushima Association for Securing Safety of Agricultural Products, which was involved in the inspection of agricultural products for shipment and selling. The Fukushima prefecture collaborates with these organisations and refers to the data for further sampling and monitoring in the prefecture.

The schedule for sampling was identified in every quarter (March, June, September and December) based on the significance of the food items for Fukushima prefecture and for the development of the region. This schedule also considered whether continuous sampling is required because of long harvesting periods and the intake rates of individual products consumed by the population. Fukushima prefecture also accounted for the inspection data collected by municipalities themselves (not in the framework of environmental monitoring). For these measurements, NaI(Tl) scintillation counters were mostly used. If the radiocaesium concentrations exceeded 50 Bq kg^{-1}, the samples were sent for precise analysis.

The inspection centre for the whole prefecture is located in the Prefectural Consumer Empowerment Centre (Fukushima city). Local inspection sites were established in each of the 59 most contaminated municipalities. In these inspection sites, the target food items included self-produced garden vegetables, edible wild vegetables and mushrooms, field soil for self-produced vegetables, potable well water, etc. Non-destructive analytical techniques for the measurements were also used at some sites.

The results are being passed as reference data to the applicant. It was required that a preliminary reservation be made before applicants were able to submit samples for measurements (in case of the Prefectural Consumer Empowerment Centre). The inspection workflow is shown in Fig. 5a, b (Fukushima prefecture 2017b).

Fig. 5a, b is about here

3.4 Cost of sampling and analysis

One of the major food items monitored for radioactivity after the FDNPP accident is brown rice. Much of the brown rice produced in the affected regions was examined, and more than 10 million bags, weighing 30 kg each, have been inspected since 2012 with a total cost of approximately 6 billion JPY per year (Fukushima prefecture 2017c). Most of the cost (5.2 billion) was paid by the Tokyo Electric Power Company (TEPCO) and the remainder by the government (Fukushima prefecture 2017c). Two hundred three machines were placed in 173 inspection stations and handled by 1,700 operators across Fukushima prefecture (Agriculture, Forestry and Fishery Department, Fukushima prefecture, 2018).

The system to control implementation of food investigations for radioactivity has been established with the collaboration of Fukushima prefecture, municipalities, the JAC (Japan Agricultural Cooperatives), rice collectors, etc. At the prefecture level, the "Fukushima no megumi" safety measures meeting (prefecture meeting) was organized by Fukushima prefecture. Fukushima prefecture, the Fukushima agriculture promotion public cooperative (secretariat), the Japan Agricultural Cooperatives federation, ZEN-NOH Fukushima (a member of the JAC), the prefectural stage rice collector, and consumer organisations were members of this council. The role of "Fukushima no megumi" was:

- Development and operation of a unified data management system for foodstuffs contamination;
- Management of the homepage to visualize the examination results for radioactivity in the food items; and
- Compensation for damages requested, depending on additional expenses of the inspection.

The regional council of Fukushima municipalities, agriculture organisations, and collector organisations are members of 38 regional councils responsible for:

- Construction of local food inspection systems (decision of settlement places and groups for inspection system, adjustment of inspection equipment usage, etc.);
- Operation of inspections (operation of inspection places, publishing of barcodes, data uploading of inspection results, etc.); and
- Summary and allocation of additional expenses.

Based on information from the monitoring database in 2011, 10,100 ha were excluded from the rice cultivation scheme. This area included production regions in the restriction area (8,500 ha) and the self-imposed restriction area (1,600 ha) (Crop Production Division, Fukushima Prefecture, 2013).

The cost of sampling is mainly dependent on the economic condition of each country, so it is important to consider the resources needed, which is the number of persons involved multiplied by hours required for the sampling campaigns (IAEA, 2004). Sampling of soil and plants for Fukushima prefecture was mainly performed by the prefectural researchers in 2011 (belonging to FATC). After 2012, farmers were required to bring their products to either the inspection centre of each municipal or the Japan agricultural cooperatives.

3.5 Specific cases of monitoring

3.5.1 Brown rice: effects of exchangeable potassium

Regarding agricultural products, monitoring has been conducted for all food items produced in the prefecture, including brown rice as a staple crop (MHLW, 2011e). At first, brown rice inspection was performed by monitoring surveys, and more than 20,000 bags were monitored in 2011. Since there was no conventional measurement method for brown rice examination for radioactivity, the measurements were conducted mainly by the relevant agencies outside the prefecture.

By spring 2011, approval of brown rice production was provided based on the map of radiocaesium concentrations in the agricultural soils and expected transfer factor value[3] (ratio of the concentration of radiocaesium in rice to that in the soil). This methodology provided a rough indicator of areas where the radiocaesium concentrations in brown rice were less than the provisional regulation level (500 Bq kg^{-1} in 2011) (NERH, 2011).

Soon after the accident in FDNPP, the prefecture governor announced a food safety declaration in October 2011, and several bags of brown rice were reported to exceed the Provisional Regulation Value (MHLW, 2011f). The main cause of this excess was small amounts of exchangeable potassium in the paddy soil, which led to higher transfer factor values than expected.

3.5.2 Rice monitoring in Minamisoma city: effect of secondary contamination

In 2015, 27 bags of rice in Minamisoma city and 28 bags of rice in Fukushima were found to contain concentrations of radiocaesium exceeding the Standard Limits. Although the origin of contamination in the samples was not fully confirmed, based on the analysis of the ratio of ^{134}Cs to ^{137}Cs in soil and brown rice, it was shown that the origin of the radiocaesium contaminant did not come from the nearby soil. It was concluded that the reason was secondary contamination due to direct adhesion of radionuclides derived from FDNPP during the maturation stage (Matsunami et al. 2016). Since then, the radiocaesium concentrations in the leaves and ears of rice plants were included as objects of monitoring to assess this secondary effect. The changes in monitoring covered not only the southern

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[3] 0.1 (Bq kg^{-1}, dry rice)/(Bq kg^{-1}, dry soil)

area (where rice exceeding the Standard Limits was observed) but also broader areas around the city. The monitoring spatial structure is now under re-evaluation for more effective rice control. The latest monitoring results were presented in the MAFF report (2016).

3.5.3 Anpogaki (Dried persimmon)

Drying of fresh persimmon leads to an increase of the radiocaesium concentrations in the sample due to removal of water from fresh fruits. Anpogaki production is mainly located in the northern part of Fukushima prefecture (e.g., Date area), in a northeastern direction from the FDNPP, which was heavily contaminated after the accident. More than 1,500 tons of products were produced annually before the accident.

To restart the production of Anpogaki, the producers in the area collaborated with the Japanese agricultural cooperatives to perform monitoring of the process from start to finish. This process included sampling and measurement of products from the juvenile persimmon fruit to the dried Anpogaki at time of shipment. For the survey to estimate the concentration of persimmon fruits, more than 2.4 kg (approximately 160 pieces) of juvenile persimmon fruits were collected from each orchard (Date city, 2016). The sampling was performed in 5 locations around each orchard.

The regulation value of radiocaesium concentrations in juvenile persimmon fruit for Anpogaki production was 10 Bq kg^{-1}. If the radiocaesium concentration in the sample exceeded this value, the processing of Anpogaki from the orchard was subject to self-imposed restrictions. Even after clearance during the juvenile inspection stage, all of the samples were subjected to non-destructive inspections after processing. Inspected samples were discarded if the tray of fruit (8 pieces) or individual piece exceeded 50 Bq kg^{-1} (screening level) (Fukushima prefecture, 2017d)

Shipment of Anpogaki, whether individual or in trays, was dependent on the fulfilment of the following requirements:

• Radiocaesium did not exceed the screening level (50 Bq kg^{-1}) in samples obtained in the previous year; and

• The amount of the shipment was still less than 60% of the average amount shipped during 2010, compared to the amount shipped before the accident (2007) (MAFF, 2018).

For Anpogaki production, the Good Agricultural Practice (GAP) system for Anpogaki, based on Japanese GAP (JGAP, 2016), was introduced (Date city, 2014). Producers were requested to comply with the following:

- to confirm the allowed persimmon field for the process;
- to follow the detailed procedures to harvest;

- to clean the processing place and tools; and
- to implement Anpogaki inspection.

According to the Fukushima prefecture report (Fukushima prefecture, 2017e), even 6 years after the accident, the production of processed persimmon (Anpogaki and other dried persimmon products) was still restricted. The production of persimmon goods was not allowed in Naraha town, Tomioka town, Okuma town, Futaba town, Namie town, Katsurao village, and Iitate village because the inspection of Anpogaki was not organised in these areas. Self-imposed restrictions for processing of persimmon were requested in areas surrounding Fukushima city, Date city, Kori town, and Kunimi town except for the “Processing reopening model area”. In this “Processing reopening model area”, Anpogaki production is allowed for the fields and producers who comply with the appropriate inspection criteria. The selection of areas for agriculture production restraint is decided by Fukushima prefecture (Fukushima prefecture, 2017f).

If the production area falls within one of seven regions in Fukushima prefecture (Fig. 6) where self-imposed restrictions have been called for and/or if the radioactivity level of processed persimmon has exceeded the 50% of the Standard Limits within the past 3 years, then the area is asked to inspect for radiocaesium contamination. If a municipality (city, town or village) has never asked for self-imposed restrictions and never exceeded half of the Standard Limits, the inspection is not required (except for areas that have been under evacuation orders in previous years). The above scheme is summarised in Fig. 7.

Fig. 6 is about here

Fig. 7 is about here

3.5.4 Fodder

For the monitoring of radionuclides in fodder, MAFF provided manuals in 2011[4] (MAFF, 2011b), which considered:

1) Fresh forage (pasture grass) and soil measurements;

2) Concentrated feed (assorted feed, mixed feed, single feed, etc.) measurements; and

3) Forage crop measurements.

[4] The translation of the document is provided in the Supplementary material 2.

An improved manual including sampling of specific agricultural produce was subsequently published by MAFF in the following year (MAFF, 2012).

4. Measures to reduce radiocaesium in other agricultural products

One of the important differences between environmental samples and agricultural product samples is accounting for contamination of the samples by the adhered soil. Washing to remove contaminated soil from the edible parts of plants and avoiding the adhesion of surface soil are important steps in decreasing the radioactivity of samples. In the case of buckwheat, it is also recommended that the grain be polished after harvesting to reduce the radioactivity (Kubo et al., 2016). In several small-scale animal farms, edible plants are collected from the levee, slope area, etc. Since most of these places were not farmland and thus were not considered target areas for decontamination, a substantial amount of radioactive materials remains on the soil surface. Hence, sample collection must be performed conscientiously to ensure accurate reflection of the plant sample radioactivity (Harada, 2014).

Additional attention should be paid to avoiding cross-contamination before harvesting. When farmers started to harvest for the first time after the accident, it was recommended that the machine be "prewashed" with samples that were to be collected. In the case of rice, the procedure can be summarised as follows (MAFF, 2013b). Before the first usage of machines, such as hulling machines and sorting/weighing machines, after the accident, it was required to clean the contamination inside the machines using the same procedures for machines with the sample and then to discard the samples used for machine cleaning. This procedure is applied to avoid secondary contamination of cereals from contamination accumulating inside of the machine and vice versa to avoid unaccounted losses of radioactivity associated with the plants during processing inside of the machine if it is much cleaner than the grain.

In the case of hulling machines, a 50-kg sample is inserted into the machine and allowed to circulate for 3 minutes. Then, all of the contents used for machine washing are discarded. For the sorting/weighing machine, the first minimum weighing unit (5 or 10 kg) should be discarded. Other agricultural machines should also be thoroughly cleaned before usage. The procedure for cleaning the hulling machines using brown rice was published in 2013 (MAFF, 2013b). Attention was paid to all machines, equipment, or other materials for agricultural activity, as well as other products that might have been affected by direct depositions after the accident. For example, in the tobacco production process, it was a requirement that the drying area be cleaned by replacing the wind protection sheet, wiping pipes and all other materials that had been placed in the drying space before use. Similar recommendations were also provided for the cleaning of the drying space for Anpogaki (dried persimmon) (Sato et al. 2016).

5. Conclusion

Enormous efforts were undertaken at the national and local levels to ensure sustainable agricultural activities in areas affected by the FDNPP accident. The procedures for sampling and radioanalytical measurements implemented after the FDNPP accident were summarised in this paper based on the inspection guidelines for analysis of foodstuffs, as well as sampling and sample measurements performed in Japan in the 9 years after FDNPP. The important point was that monitoring and sampling for food safety should be considered differently from environmental monitoring because the purpose of monitoring for food safety is to provide safe food for the people, and the purpose of environmental monitoring is to investigate how the environment has been contaminated. The experiences gained and the lessons learned after the Fukushima accident can be useful in the improvement of emergency responses in many other countries.

In the case of the FDNPP accident, it was clearly demonstrated that preparedness for the nuclear accident should have existed before the accident. When the accident occurred, the shortage of machines, specialists, and manuals to handle the situation made the situation worse. The problems to deal with a tremendous number of plant and soil samples under emergent condition was also discussed in another paper by Hachinohe and Shinano (2020). It is recommended to prepare the plan for monitoring food in early and successive phases after the accident, and the procedures should be more flexible based on the situation of the disaster: more efficient but less precise information is required for urgent surveys of food, and precise determinations can be backup for these monitoring systems. However, such issues as how detailed the examination of the foodstuffs should be, where and how the inspections should be continued and where, when and how these activities are to be terminated remain challenges in areas affected by the Fukushima accident even 9 years after the nuclear power plant accident in Japan.

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Appendix A. Supplementary data

Supplementary data to this article can be found online.

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