Journal of Environmental Radioactivity 223-224 (2020) 106373

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: http://www.elsevier.com/locate/jenvrad

# Soil and vegetation sampling during the early stage of Fukushima Daiichi Nuclear Power Plant accident and the implication for the emergency preparedness for agricultural systems

Yuichi Onda [a,*], Lieve Sweeck [b], Takuro Shinano [c], Gerd Dercon [d], A. Lee Zhi Yi [d], H. Kato [a]

[a] Center for Research in Isotopes and Environmental Dynamics, University of Tsukuba, Tsukuba, 305-8572, Japan
[b] Biosphere Impact Studies, Environment, Health and Safety Institute, Belgian Nuclear Research Centre, Boeretang 200, 2400, Mol, Belgium
[c] Research Faculty of Agriculture, Hokkaido University, N9 W9, Kitaku, Sapporo, Hokkaido, 060-8589, Japan
[d] Joint FAO/IAEA Division of Nuclear Techniques in Food and Agriculture, International Atomic Energy Agency, IAEA, Department of Nuclear Sciences and Applications, Wagramerstrasse 5, A-1400, Vienna, Austria

ARTICLE INFO

*Keywords:*
Nuclear emergency
Soil and vegetation sampling
Fukushima nuclear power plant accident
$^{137}$Cs
$^{131}$I

ABSTRACT

After the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, immediate soil and vegetation sampling were conducted according to the action plan of nuclear emergency monitoring; however, analysing the monitoring dataset was difficult because the sampling protocols were not standardised. In this study, the sampling protocols applied just after the FDNPP accident were reviewed, and the monitoring data were analysed. The detailed protocols and results can provide a sound basis for guidelines of soil and vegetation sampling for nuclear emergency monitoring. The activity concentrations of $^{137}$Cs and $^{131}$I in weed samples measured immediately after the FDNPP accident were related to the air dose rate at 1 m. Consequently, vegetation sampling is recommended when the additional dose rate (above background) is higher than 0.1 μSv/h. To enhance the efficiency of a protective response in the case of a nuclear accident, predetermined sampling points for soil and vegetation sampling should be considered in the preparedness plan for nuclear emergencies. Furthermore, sampling and analytical measurement capacities (time, people, cost) during the early phase after nuclear emergencies need to be considered in the preparedness and action plan, and sampling and measurement exercises are highly recommended.

## 1. Introduction

After a nuclear emergency, to protect the public from external and internal radiation, performing immediate sampling and radioactivity measurements to determine the contamination levels of the soil and vegetation is advisable (IAEA, 1999). Possible consumption restrictions of agricultural products based on measurement results can be implemented. For example, short-lived $^{131}$I is radiotoxic to humans because it may accumulate in the thyroid and cause thyroid cancer (Baverstock et al., 1992). Therefore, it is essential to measure $^{131}$I within a few days. If the activity concentration is expected to exceed the intervention criteria for food consumption, the agricultural products should be banned immediately. For this purpose, it is necessary to establish emergency sampling protocols and sample analysis capacity within a limited time frame. For quick monitoring in case of an accident, it is necessary to establish well-designed emergency monitoring systems.

As part of a regulatory protocol, environmental monitoring networks have been established in most countries with nuclear facilities. Such monitoring networks often combine off-line sampling and analysis programs (measurements of radioactivity) with an on-line gamma dose rate and air concentration network, which can act as an early warning system in the case of an accident. The online monitoring network is one of the primary data sources for decision-making during the radioactivity release phase of an accident. Meanwhile, sampling and measurement programs that commence just after the release would provide valuable supplementary information. International agencies, such as the IAEA, have issued guidelines (IAEA, 1999) on sampling and analysis in the case of a nuclear emergency. Thus far, there have been very few actual events to test the emergency response of the monitoring networks. In addition, modification of sampling methodologies may be required over time,

* Corresponding author. Center for Research in Isotopes and Environmental Dynamics, University of Tsukuba, A405 Integrative Research Building, 1-1-1 Tennodai, Tsukuba, 305, Japan.
*E-mail address:* onda@geoenv.tsukuba.ac.jp (Y. Onda).

https://doi.org/10.1016/j.jenvrad.2020.106373
Received 28 January 2020; Received in revised form 13 July 2020; Accepted 27 July 2020
Available online 29 August 2020
0265-931X/© 2020 Elsevier Ltd. All rights reserved.

from days and weeks to months because the monitoring objectives change. Therefore, improving and optimising the sampling and monitoring of the soil and food while creating general guidelines for monitoring radioactivity in food and agriculture is necessary (Dercon et al., 2020). In this study, the focus is directed toward the early transition phase of a nuclear emergency, starting from a few days to a few weeks after release. During the first stage of a nuclear accident, direct deposition to vegetables is the main contamination pathway when crops are in the field during the release (Pröhl, 2009), whereas root uptake will become the main pathway in the following year. The prognosis of the contamination of food, amount of radionuclides, and their spectra in the soil requires soil and vegetation monitoring data.

In Japan, after the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, the Emergency Operation Center (EOC) performed operations at the Environmental Radioactivity Monitoring Center of Fukushima (ERMF), collecting soil and vegetation samples (MEXT, 2011a,b). The measurement results were published one day after the major release on March 15 (Katata et al., 2015). Although these data were essential for immediately estimating the radioactivity contamination, interpreting the deposition density of $^{134}$Cs, $^{137}$Cs, and $^{131}$I in the soils (given in Bq/m$^2$) was challenging because the data are provided in the units of (Bq/kg). For the vegetation sampling results, detailed protocols have not been established; thus, it is difficult to properly analyse the dataset. The use of standardised protocols, analytical methods, and communication is essential for applying the lessons learned from the Fukushima accident.

The selection of sampling locations is also essential in the implementation of nuclear emergency monitoring. For vegetation sampling, it is necessary to select optimal locations for sampling and apply appropriate methods for reporting and visualising sample locations in further data analysis. In the case of the Japanese emergency monitoring plan, no map-based dataset of food and vegetables has been developed; only the names of the cities, towns, etc. have been recorded. For example, in Fukushima, the areas of these municipalities range from 16 to 1272 km$^2$ (Kato and Onda, 2018); thus, estimating the degree of radioactivity contamination after a nuclear emergency based on this geographical information is difficult. Meanwhile, other countries, such as Belgium, have well-defined sampling protocols. By combining the lessons learned from the FDNPP accident with protocols of other countries, we can improve and optimise the guidelines for monitoring radioactivity during an emergency period.

In this study, based on the experience of emergency monitoring immediately after the FDNPP accident in Fukushima and the present monitoring programme in the Belgian emergency preparedness and action plan, the method and applicability of emergency protocols are discussed for the radioactivity monitoring of foods and agricultural products during a nuclear emergency period.

## 2. Emergency monitoring

### 2.1. Case study after the FDNPP accident

#### 2.1.1. Emergency vegetation sampling conducted in the FDNPP accident

Initially, the Fukushima Off-Site Center was established in Okuma Town, within 10 km of the FDNPP. The off-site centre was located within the evacuation zone and then moved to Fukushima city on March 14, 2011. The emergency plan contained pre-determined sampling points within a 10 km radius; however, there were no predetermined sampling points outside of 10 km from the FDNPP. Consequently, sampling points outside of the 10 km radius from the FDNPP had to be selected on March 15, comprising mainly public facilities (e.g., city halls, city administrative offices, public parks). The actual sampling could only start the next day, on March 16.

Fig. 1 indicates the location of emergency sampling sites near the FDNPP; the sampling started on March 16 and 17. A total of 13 sampling sites were chosen, and among them, 11 points were used as monitoring points until June 30, 2011 (Table 1) for the dose rate, vegetation samples, and soil samples (MEXT, 2011a,b). The following sampling protocols for vegetation were established by the Fukushima Prefectural Government (2008).

1) Collection of $^{131}$I measurement samples

Approximately 400 g of $^{131}$I was filled in a V-5 styrene container (AS ONE, Tokyo, Japan; inner diameter of 135 mm, height of 56 mm, capacity of approximately 900 mL) and compressed as much as possible. The collected V-5 type styrene container was placed in a plastic bag, and the name of the sampling place, date, and time were written on the plastic bag.

2) Collection of samples for measurement of other radionuclides

Approximately 3 kg of the sample was collected in a plastic bag. In addition, when the sample was collected, the plant was covered with a vinyl bag, such that radioactive substances adhering to leaf vegetables do not splash; then, the part above the plant was cut from the root and packed. On the plastic bag, the collection point name, date, and time were noted.

Although this sampling method is easy, the large sample volume requires much time for sampling, sample preparation, and analysis. In addition, because the Fukushima accident occurred in the early spring when there was no new growth, it was difficult to apply the sampling protocol; thus, a simplified method, described below, was developed on March 15, 2011.

*2.1.1.1. Simplified method for vegetation sampling.* Because the FDNPP accident occurred in early spring, it was impossible to collect large volumes of vegetation. As a result, ERMF was used to simplify the sampling protocols and collect the samples of the rosette plants that were present, such as *Rumex obtusifolius*, dandelion, plantain, goldenrod, and *Papaver dubium* (Fig. 2a and b) (Y. Abe, personal comm.). This simplified protocol can be used as an excellent analogue for leafy vegetables, because the chosen vegetation can efficiently capture the fallout radionuclides. The simplified method also made much quicker measurements possible. The dose rate measurements and collection of soil and vegetation samples at each location were performed within 10 min with a small number of people.

The simplified method is as follows (Fig. 2):

1) List the name of the sampling point, collection date, and time in the collection container (U-8 container; AS ONE, Tokyo, Japan; 50 mm inner diameter and 60 mm height). Take pictures of the soil sampling place (nearby and distant views including the sampler).
2) Duplicate the collector's rubber gloves to avoid cross-contamination. In addition, cover the outside of the container with a plastic bag.
3) Place the pinch bag in the hand and grab to pick up and extract the grass, as shown in Fig. 2b and c.
4) Turn over the plastic bag, place it on the hand, shape it, and push it into the container until the container is filled (Fig. 2d).

#### 2.1.2. Soil sampling protocol

To determine the migration depths of $^{137}$Cs and $^{131}$I due to the initial fallout, it is essential to elucidate the vertical distributions of $^{137}$Cs and $^{131}$I in the soil column. The best method to obtain the depth distributions of $^{137}$Cs and $^{131}$I is the application of the scraper plate, which scrapes the soil from the surface typically at a 5 mm interval, as summarised by the CRP of IAEA "Assess the effectiveness of soil conservation techniques for sustainable watershed management using fallout radionuclides" (D1.50.08; Zapata, 2002; Dercon et al., 2012). In the case of the FDNPP accident, we found that 99% of the radiocaesium and $^{131}$I can be collected in the upper 5 cm of the soil column (Kato et al., 2012), and an average of 98.6% of $^{137}$Cs can be found in the top 5 cm. In total, eight

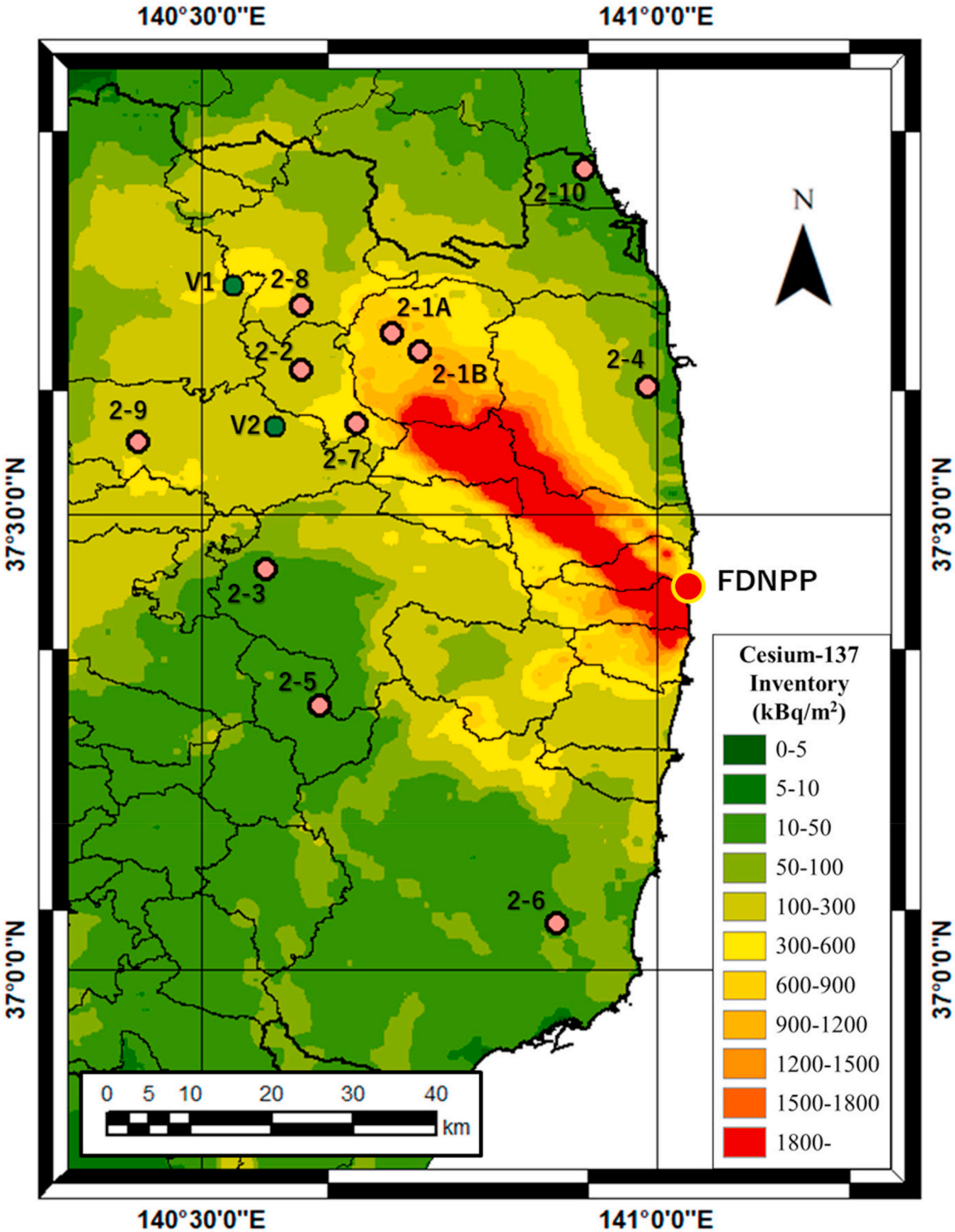



**Fig. 1.** Location of the early stage sampling site. The pink circles are regular sampling points, and the green circles are one-time sampling points. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

**Table 1**
Early-stage soil sampling data near FDNPP.

| Site ID | Municipality | Latitude | Longitude | $^{137}$Cs initial deposition (kBq/m$^2$) | Wet deposition ratio | $^{137}$Cs concentration in soil | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | Mean (Bq/kg) | Geomean | Median | SD | CV | n |
| 2-1A | Iitate Village | 37.70 | 140.71 | 961 | 0.96 | 29,737 | 16,716** | 20,072 | 34,155 | 1.15 | 98 |
| 2-1B | Iitate Village | 37.68 | 140.74 | 798 | 0.95 | 19,598 | 13,744 | 20,062 | 13,197 | 0.67 | 90 |
| 2–2 | Kawamata Town | 37.66 | 140.61 | 168 | 0.92 | 4764 | 3828 | 4520 | 2884 | 0.61 | 98 |
| 2–3 | Tamura City | 37.44 | 140.57 | 20 | 0.85 | 723 | 510 | 641 | 555 | 0.77 | 99 |
| 2–4 | Minami-Soma | 37.64 | 140.99 | 67 | 0.85 | 2157 | 1624 | 1805 | 1595 | 0.74 | 104 |
| 2–5 | Ono Town | 37.29 | 140.63 | 24 | 0.48 | 812 | 583 | 692 | 660 | 0.81 | 99 |
| 2–6 | Iwaki City | 37.05 | 140.89 | 28 | 0.91 | 941 | 648 | 851 | 627 | 0.67 | 95 |
| 2–7 | Kawamata Town | 37.60 | 140.67 | 459 | 0.80 | 14,113 | 10,522 | 14,055 | 8656 | 0.61 | 91 |
| 2–8 | Date City | 37.73 | 140.61 | 212 | 0.98 | 4495* | 3581 | 4575 | 2395 | 0.53 | 92 |
| 2–9 | Nihonmatsu City | 37.58 | 140.43 | 214 | 1.00 | 12,191 | 9883 | 11,043 | 6813 | 0.56 | 91 |
| 2–10 | Shinchi Town | 37.88 | 140.92 | 50 | 0.76 | 210 | – | – | – | – | 1 |
| V1 | Fukushima City | 37.75 | 140.54 | 341 | 1.00 | 18,099 | – | – | – | – | 1 |
| V2 | Nihonmatsu City | 37.60 | 140.58 | 177 | 0.98 | 6264 | – | – | – | – | 1 |

SD: Standard deviation, CV: Coefficient of variation, * Normal distribution ($p < 0.05$), ** Log-normal distribution ($p < 0.05$).
Reference date of $^{137}$Cs concentration in soil was March 12, 2011. Data was by Kato et al. (2019).

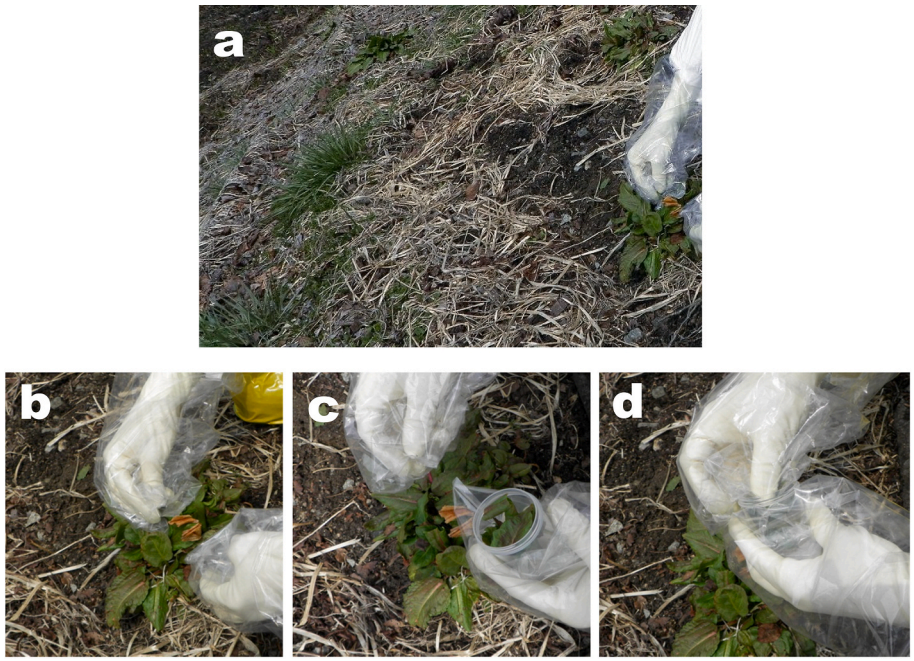



**Fig. 2.** Emergency Vegetation sampling (weed; in this case *Rumex obtusifolius*) by ERNF Fukushima Prefecture. Photo taken by Environmental Radioactivity Monitoring Center of Fukushima.

**Fig. 3.** Emergency Soil sampling protocol used as the emergency sampling by ERNF Fukushima Prefecture. (Photo taken by Environmental Radioactivity Monitoring Center of Fukushima).

scraper sampling datasets were collected from different land uses from June 21 to July 2, 2011 (Takahashi et al., 2015).

Again because of time restrictions, the ERMF established an emergency protocol as follows (Fig. 3) on March 15, 2011.

1) Duplicate the collector's rubber gloves, put the plastic bag in the hand, hold the sampling container from below, and open the lid of the U-8 container. Insert the container into the ground and fill approximately 80% of the container with the soil to a height of 5 cm (Fig. 3a).
2) Turn the container open upwards and shake it to shape the sample (Fig. 3b).
3) Close the lid, flip the plastic bag over the hand, tie the container, and tie the plastic container (Fig. 3c).
4) When collecting for $^{90}$Sr analysis, use a shovel and take approximately 1 kg of the 5 cm sample in a plastic bag, as a low $^{90}$Sr activity concentration is expected.

The soil core samples were taken at the same locations every day for more than one month. Each sampling location can be easily detected by the "hole" left after sampling (Fig. 4). In this case, the photo was taken on June 11, 2011.

Although the EOC continues to sample soil at the same locations with a single soil sample, the activity concentrations of $^{137}$Cs in the soil samples varied significantly. Fig. 5 shows a histogram of the activity concentrations of $^{137}$Cs in the soil at each sampling site. The detailed data are shown in Table 1. At each location, 91–104 soil samples were typically taken to be 25 m$^2$. The coefficient of variation ranges from 0.53 to 1.15, depending on the sampling location. The frequency distribution at the 2-1A location shows a log-normal pattern, whereas that at the 2–8 location show a normal distribution ($p < 0.05$); however, no distribution can be fitted for the other sites. This substantial variation can be caused by the sampling method because of spatial heterogeneity. Owing to time restrictions, no representative sampling was performed. Preferably, the places where no vegetation was found were selected for each site. In each of these places, the U8 container was inserted. Because the depth of the sample container was 5 cm, the upper 5 cm of soil was collected. The small sampling area (i.e., the opening of the container is only 19.6 cm$^2$) may partly explain the high amount of scattering in the data. Khomutinin et al. (in this issue) and Barnekov et al. (2019) recommend a sampling area of at least 50 cm$^2$.

**Fig. 4.** Situation of soil sampling location after several sampling by ERNF Fukushima Prefecture. (photo taken June 11, 2011 by Environmental Radioactivity Monitoring Center of Fukushima).

In addition, the sample mixture is another critical issue. Onda et al. (2015) demonstrated that the measured values of the activity concentrations of $^{137}$Cs in the soil samples varied depending on the degree of mixing in the soil sample. The activity concentration changes substantially if the soil sample is not homogeneously mixed. During the large-scale government sampling programme (Saito et al., 2015), the EOC protocol was modified to ensure the quality and reproducibility of the soil sample data (Onda et al., 2015). For the detailed sample preparation protocol, we refer to Onda et al. (2015). Here, we propose a modified sampling method.

1) In a square of 9 m$^2$, five easily accessible points were selected from which to collect soil samples. The preferred method is to collect soil samples from points as evenly spaced as possible (Fig. 6a); it is not ideal but in case the sampling person cannot find the bare soil, they will collect soils with small vegetation. In this case, small root system layer was also collected together as a soil sample the when small weeds grow in the soil sampling position, sampling is conducted, including weeds. The root system layer was also collected together as a soil sample.
2) The samples were then mixed by pouring into a plastic bag (Fig. 6b).
3) The sample in the plastic bag was then firmly mixed by hand (Fig. 6c).
4) The soil samples were returned to the container (Fig. 6d).

With this additional processing, the sample is better homogenised, and the variability in the results is significantly reduced. During large-scale government sampling, the average coefficient of variation (CV) for $^{137}$Cs concentrations among the five samples was 0.36 (Saito et al., 2015). These data clearly suggest that the large CV observed during the emergency sampling (Fig. 5, Table 1) may be due to the mixture effect. The significant fallout of the radionuclides in the FDNPP accident occurred in March 2011. During this season in Japan, it is easy to find a bare soil surface; however, if the atmospheric release of radionuclides occurs during the plant-growing season, both vegetation and soil need to be carefully sampled separately at the same time and mixed well, as described in 2).

### 2.1.3. *Sample analysis after sampling*

In the case of a nuclear emergency, such as Fukushima, which was initiated by a natural disaster, many of the equipment did not work. In EOC Fukushima, one Ge-detector out of three in the gamma spectrometer was seriously damaged by the earthquake. Thus, such a monitoring plan needs to consider that all of the equipment may not be operable in the case of a nuclear emergency. Moreover, in the emergency period, high activity concentrations of radionuclides in the samples need to be measured, which makes sample preparation and measurement time the bottlenecks of the analysis process. Consequently, protocols used for routine releases (MEXT, 1983) should be changed to reduce these steps in the sample analysis.

The collected sample is measured as it is fresh weight without any sample preparation. In addition, drying in the case of $^{131}$I is not recommended to prevent loss by volatilisation. The radioactive contamination (Bq/sampling area) should be converted to contamination per square meter of land (Bq/m$^2$). In some cases, because of the high activity concentration of $^{134}$Cs, the cascade of rays can cause an underestimation of the activity concentration of $^{134}$Cs due to the summing effect. This effect can be corrected by increasing the distances (e.g., 30 cm) between the detector and soil samples in the case of highly contaminated soil samples (Yamamoto et al., 2020).

The radioactivity measurement is conducted for all five subsamples, in which the five subsamples or five subsamples mixed to create one sample are measured to represent one location. The method can be selected depending on the bottlenecks of sample preparation or measurement time. In addition, the contamination of the detector always needs to be carefully maintained.

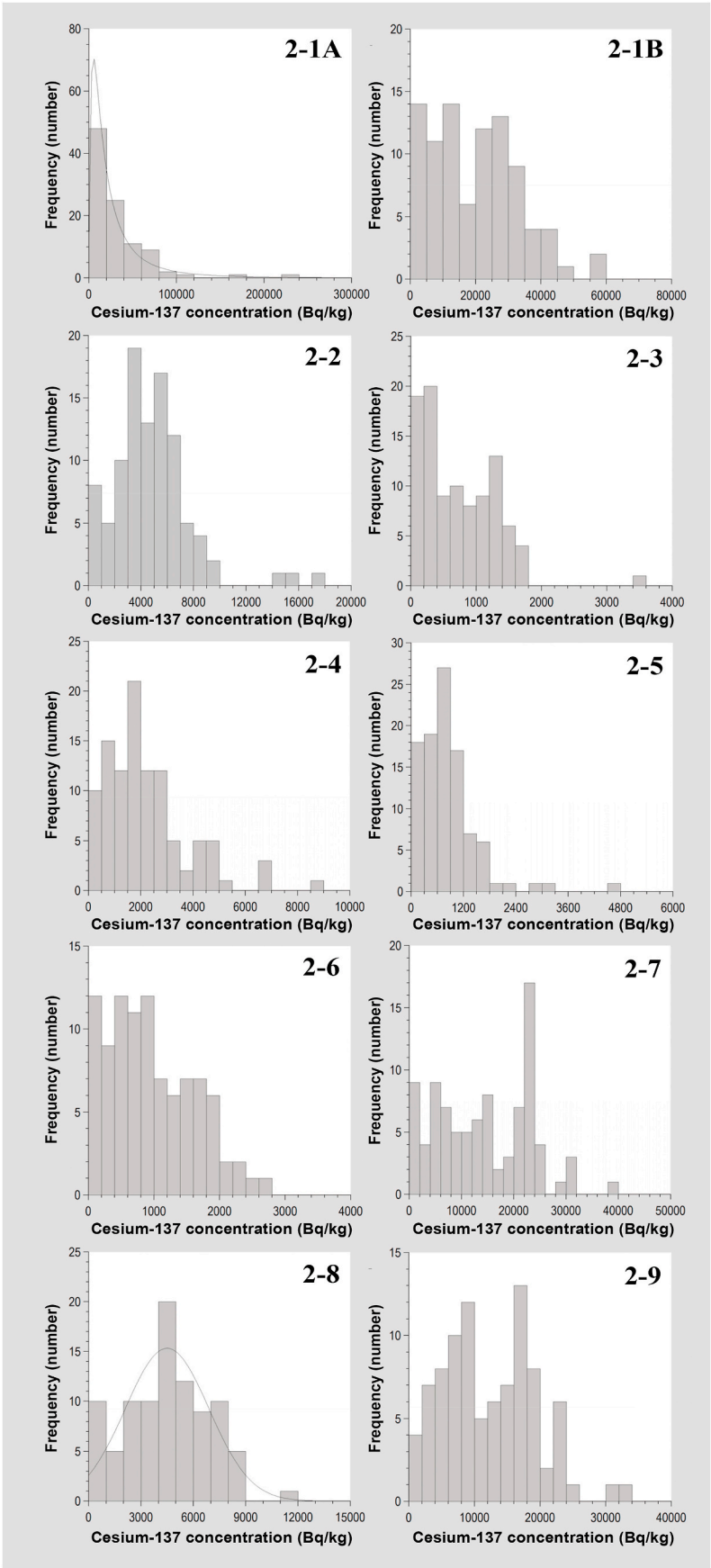



**Fig. 5.** Histogram of the activity concentrations of $^{137}$Cs in the soil samples taken from March 16 to June 30.

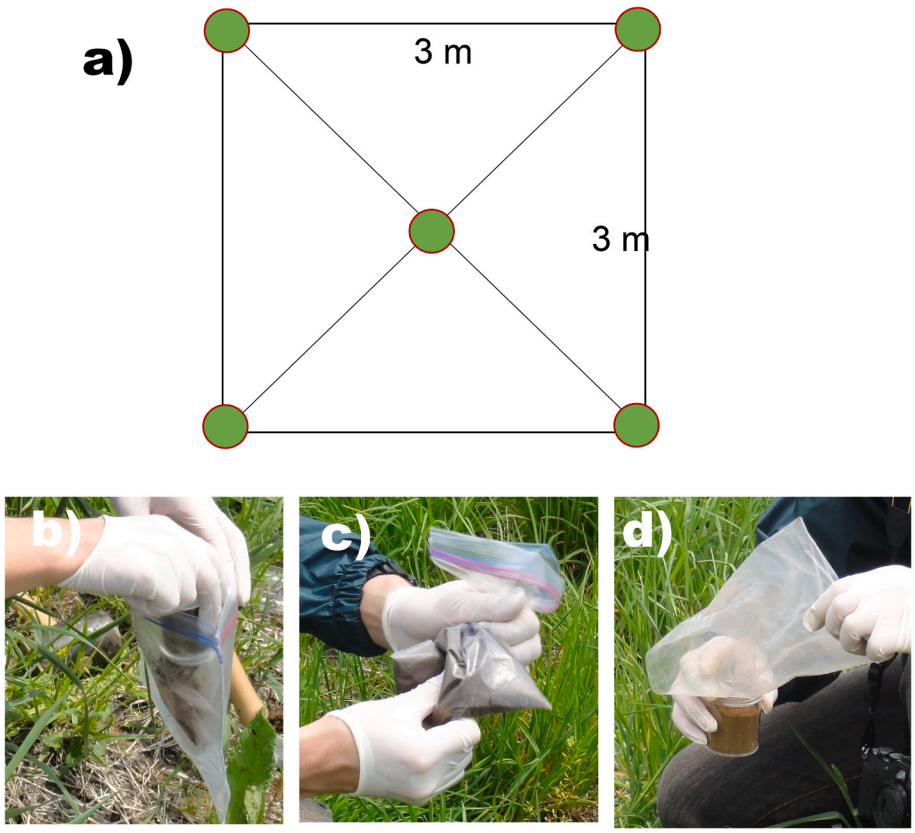



**Fig. 6.** Emergency sampling points (a) and the procedure for the investigation of radioactive contamination of the soil using the U-8 container (b,c,d).

*2.1.4. Data released to the public*

After the FDNPP, the Japanese government and local government have published data on leafy vegetables, which are highly likely to be affected by the radionuclide fallout. Fig. 7 represents the activity concentration data of $^{131}$I for spinach in Ibaraki Prefecture (Ibaraki Prefecture, 2011). These data have been mapped by our group because, as shown before, government data are recorded only at the city–town level. As shown in Fig. 4, the activity concentrations of $^{131}$I (more than 54 kBq/kg of fresh weight) in the spinach samples have been reported on March 18, 2011. This data caused significant public anxiety because the temporal consumption criterion (2000 Bq/kg) was exceeded. In the following week, the measured values were greater than the restrictive consumption level of $^{131}$I. Eventually, the, reporting of field data regarding vegetables by the local government had ceased. In March 2011, other data on vegetables, except that of open-culture vegetables, were published. These data demonstrated much lower values than those recorded on March 18 and 19 because they involved the activity concentration data of spinach sold on the market. Most of the spinach on the market was not open culture but grew in greenhouses that shielded the plants from direct radioactive fallout deposits but can have affected by the fresh air from the outside by ventilation.

Although a significant fallout of $^{137}$Cs and $^{131}$I occurred in this area (Katata et al., 2015) on March 21, no data were published until March 31, and all of the published data were of products cultivated in greenhouses. Consequently, no published field data from Ibaraki Prefecture were available immediately after the most contamination-driven event that has occurred in Ibaraki Prefecture.

## 2.2. Emergency sampling protocol in Belgium

*2.2.1. Establishing predetermined sites*

The Belgian radiological monitoring programme provides a radiological profile of the Belgian territory and contributes to the increasing need to quantify the actual situation as explicitly as possible, implementing, lessons learned from the Chernobyl experience. Besides the field sampling programme in which air, water, soil food products, and aquatic bio-indicators are sampled on a regular basis at the same locations spread over the entire country, the Belgian monitoring programme also entails the TELERAD network. The TELERAD network consists of 250 stations measuring the overall radioactivity of air (ambient radiation), aerosols, and river water. It operates fully automatically, and an important function of the TELERAD network is that it can be used as an early warning network. However, the field sample campaign of the national monitoring programme is only designed for routine releases.

To increase the rapid and effective execution of field measures, predefined sampling points were added to the measuring programme of the Belgian emergency response programme in 2010 (IBZ, 2010). For each of the main nuclear sites located in the Belgian territory and foreign nuclear sites located near the Belgian borders, a set of measuring points was defined. The area near the nuclear site is divided into 30° sectors, and one measuring point is taken at 1, 3, 5, 10, 15, and 20 km for each sector (starting from 0°) and 1 measuring point at 17.5 km for each 30° sector (starting from 15°), as illustrated in Fig. 8 for the nuclear site in Tihange.

At each predefined measuring point, soil, grass, and/or one of the other sample matrices are available for sampling. If this was not the case, the nearest location to this point was considered, and the coordinates were adapted. Of each point, a description was made using the template given in Fig. 9. The name of each point is an abbreviation indicating that it is a pre-defined measurement location, including the area of interest (e.g., T = Tihange) and a sequencing number (e.g., 01). The label of each measuring point exists by the identification of the site or area of interest (first letter) + angle (°) + distance (km).

*2.2.2. Sampling procedures for emergency situations*

Sampling procedures in case of emergency have been defined by

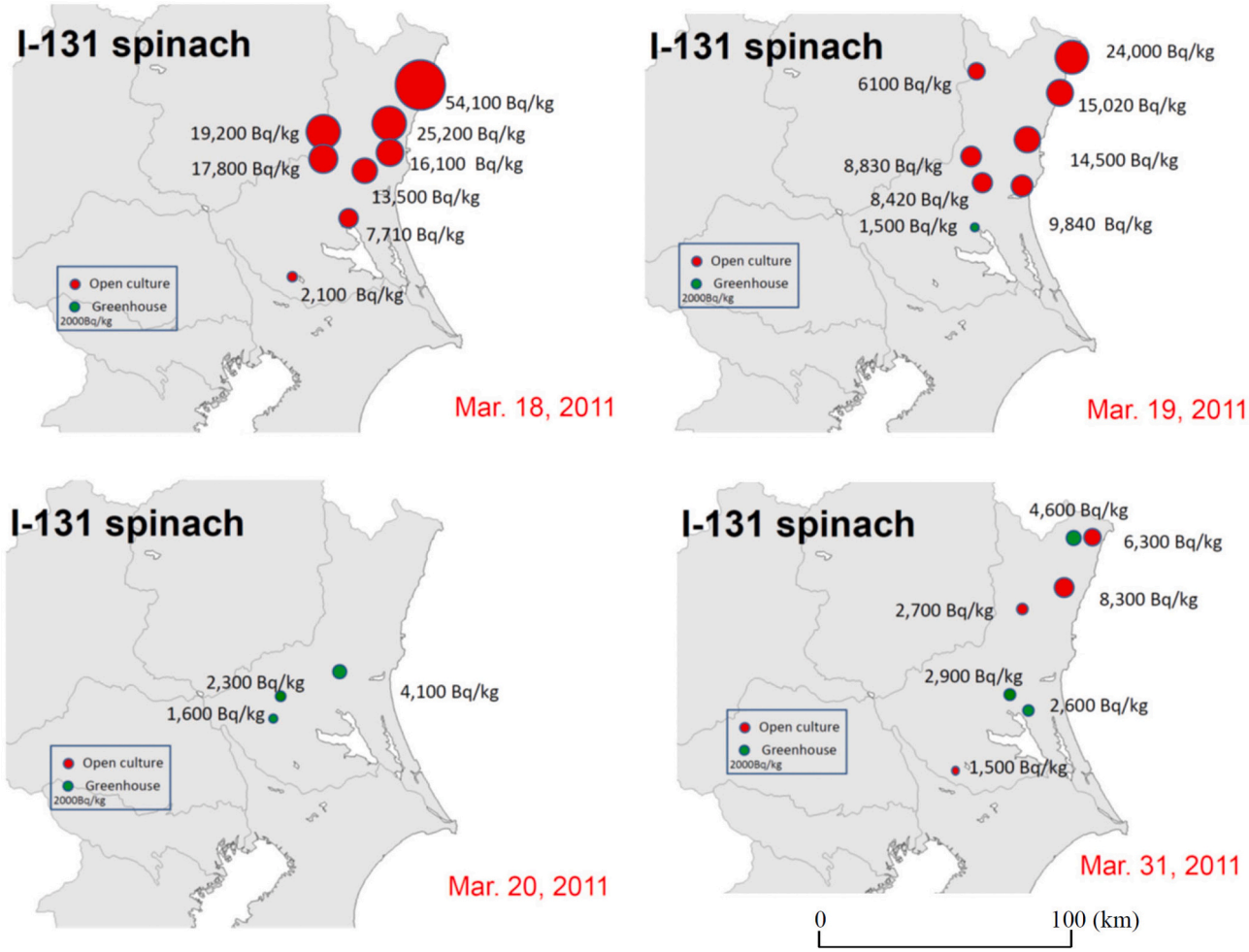



**Fig. 7.** Geographical distributions of $^{131}$I activity concentrations in the Japanese mustard spinach in Ibaraki Prefecture (published data).

FANC, IRE-ELit, and SCK•CEN. Procedures were set up for the sampling of grass, air, water (rain, sea), milk, and vegetables. The procedures highlight the main points to follow for a harmonised operation to be used by the different teams in the field. As an example, the procedure for sampling grass is reproduced here:

1) Delineate a zone of 1 m$^2$ with a frame, rope, or other means of undisturbed grass.
2) Cut the grass at approximately 4 cm above ground, avoiding as much loss from fallout as possible.
3) The minimum weight of the sample was 250 g. If after cutting 1 m$^2$ a weight below 250 g is obtained, then an additional square meter must be included.
4) The sampled grass was collected in a plastic bag. The grass was weighed in the bag using a dynamometer. The sample bag was properly labelled and placed together with the sampling fill in the form of a second bag.

Sampling exercises following the corresponding procedures are part of the drills used during the different training sessions organised for the sampling teams.

## 3. Suggestions for improving emergency response

### 3.1. Dose rate and activity concentration of $^{131}$I in the vegetation samples

During the first days after the Fukushima accident, short-lived radioisotopes, such as $^{132}$Te and $^{132}$I, accounted for nearly 49% of the deposit activity (Champion et al., 2013). Afterwards, $^{134}$Cs and $^{131}$I became the primary source of radiation. For grass samples, the direct deposition of the fallout on the vegetation can result in the relatively high activity concentrations of these radionuclides in feed and foods (Pröhl, 2009).

The activity concentrations of $^{137}$Cs and $^{131}$I in vegetation versus the air dose rate are plotted in Fig. 10. In addition, the data for spinach from Tsukuba were plotted on the same diagram (Ohse et al., 2015). Fig. 10a shows the results measured 1–3 d after the significant fallout (March 15). It is evident that good relationships between the air dose rate and the activity concentrations of $^{137}$Cs and $^{131}$I were obtained. For $^{131}$I, the activity concentrations in weeds, where $^{131}$I/$^{137}$Cs activity ratios are expected to be high (locations 2–3 and 2–6; Saito et al., 2015) due to the plumes of the high dry fallout ratios, would be even higher than the values expected from the regression line. However, in this case, the $^{137}$Cs data did not show such a tendency. If the regression line toward lower dose rates is extended, we saw that at an air dose rate of 0.1 μSv/h at the time of the measurement, the activity concentration of $^{137}$Cs would be approximately 1500 Bq/kg, except sites 2–3 and 2–6, and 4000 Bq/kg for $^{131}$I. After 10 d of significant fallout (Katata et al., 2015), again, good relationships between the air dose rate and $^{137}$Cs and $^{131}$I were found, as shown in Fig. 10b. We did not find any higher intercept values for $^{137}$Cs and $^{131}$I in Iwaki and Tamura. At an air dose rate of 0.1 μSv/h at the time of the measurement, the activity concentrations of $^{137}$Cs and $^{131}$I on the vegetation samples were well over the100Bq/kg. The background air dose rate at Tsukuba of the spinach sampling point was 0.027 μSv/h, and Fukushima prefecture was 0.034 μSv/h (Yoshimura et al., 2020). Therefore, when we assume the background dose at 0.03 μSv/h, the

**Fig. 8.** Predefined measuring point around the nuclear power plant of Tihange, Belgium.

activity concentrations of $^{137}$Cs and $^{131}$I on the vegetation samples were estimated as 1700 Bq/kg and 4300 Bq/kg, respectively, in the area of additional 0.1 μSv/h were observed (see Fig. 11).

If these vegetation samples can be used as analogues for leafy vegetables, the activity concentrations of both $^{137}$Cs and $^{131}$I are well above the allowance level in Japan ($^{137}$Cs for 500 Bq/kg and $^{131}$I for 2000 Bq/kg), and IAEA Operational Intervention Levels for Reactor Emergencies (OIL) 7 criteria ($^{137}$Cs for 200 Bq/kg and $^{131}$I for 1000 Bq/kg) (IAEA, 2017). Spinach data were collected at Tsukuba. The good relationship between dose rate and activity levels in the vegetation samples in the early emergency phase can facilitate decision-making concerning the possible limitations of vegetable consumption just after nuclear emergencies.

Globally, population-weighted absorbed dose rates in air average are 59 nGy/h (≈0.059 μSv/h) A typical range of variability for measured absorbed dose rates in air is from 10 to 200 nGy (UNSCEAR, 2000). Due to the background dose rate varies between places, the dose rate should be monitored to prepare the nuclear emergency. Also these variable background doses demonstrate that this will be above variation in background at sites and that the approach is going to viable worldwide. Bearing mind above constraints, the measurement of the air dose rates can be used to determine the activity concentration of leafy vegetables, or the temporarily forbidden harvest, in the case of an additional dose rate above background exceeding 0.1 μSv/h.

After a few weeks, the deposited $^{131}$I decayed and was gradually washed away. Fesenko et al. (in this issue) estimated that the $T_{eff}$ of grass weed was at 3.7 d for $^{131}$I and 6.9 d for $^{137}$Cs. Ohse et al. (2015) measured $^{137}$Cs and $^{131}$I in spinach and Japanese mustard in Tsukuba and estimated similar effective half-lives for $^{137}$Cs and $^{131}$I. Hirose (2012) summarised that the half-life of the atmospheric deposition of $^{137}$Cs was approximately 12 d, which suggests that the subsequent wash off may be important for $T_{eff}$ on grass weed.

Renaud et al. (2012) also used the same dataset to simulate the washout from the leaves in Japanese vegetation sampling data using the SYMBIOSE model and Fesenko et al. (in this issue).

### 3.2. Proposed soil/vegetation sampling

In the initial days after a nuclear accident, the crops present in the field are mainly contaminated by the radionuclides deposited on the plant leaves. As such, it is not necessary to sample "real" vegetables. The contamination level may be different depending on the size of the crops; however, weed or grass can be a good indicator of the contamination level of leafy vegetables. Since the time between the end of the release and sampling, the campaign is limited to establishing predetermined sampling points, as shown in Fig. 9, as is strongly recommended. In addition, the locations at which the grass (to use as an analogue for leafy vegetables) is regularly trimmed or kept short by grazing animals, would be recommended for use.

In the initial 2–3 days after the nuclear emergency, vegetation and soil with determined protocols target $^{131}$I; thus, it will be essential to determine the fallout radionuclide ratio (e.g., $^{131}$I/$^{137}$Cs ratio). The data presented in this paper can provide insight into the determination of OIL and sampling protocols.

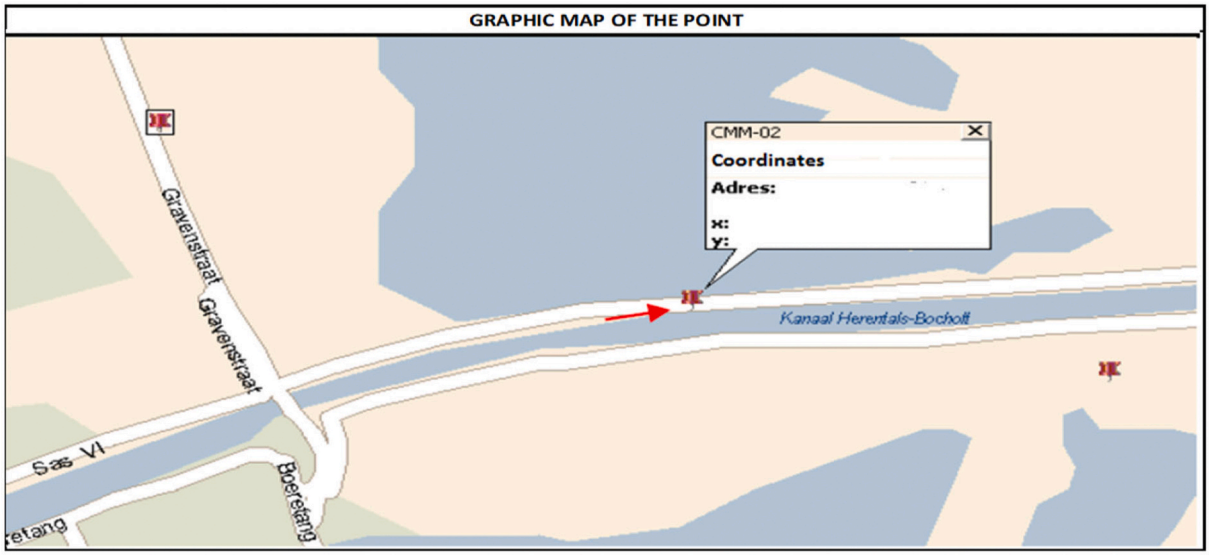
IAEA (2011) estimates that the OIL 3 criterion for vegetation sampling/monitoring sets the air dose rate to <1 μSv/h. Recently, IAEA has established a new criterion, that is, OIL 7, for the allowance level of food (IAEA, 2017). The OIL 7 establishes an activity concentration of 200 Bq/kg for $^{137}$Cs and 1000 Bq/kg for $^{131}$I; however, no criteria was issued for the air dose rate for food consumption. In Japan, OIL 6 is the allowance level for the air dose rate, set at 0.5 μSv/h (NRA, 2017). However, based on Fig. 10, the activity concentration of $^{131}$I in leafy vegetables under the condition of 0.5 μSv/h can be as much as 10,000 Bq/kg, which is 10 times higher than the OIL 7 criterion (IAEA, 2017)

| ROAD BOOK | Name of the measuring point | | Label | |
|---|---|---|---|---|
| Nuclear site | | | | |
| COORDINZATES | LATITUDE (NORD) | | LONGITUDE (EAST) | |

| | | | | |
|---|---|---|---|---|
| ADRESS | | | | |
| GEOGRAPHICAL SITUATION | | | | |
| COORDONNEES | Lambert X (m) | | Lambert Y (m) | |

PICTURE OF MEASURING POINT

GRAPHIC MAP OF THE POINT




| REMARKS | | |
|---|---|---|
| If the coordinates cannot be respectéé due to difficult access, take the nearby measuring point and mark the coordinates together with the results. | | |
| Difficult access in winter: | ☐ | Others |
| Difficult connection to communication network | ☐ | ..... |
| Dirt road | ☐ | ..... |

**Fig. 9.** Example of a description of a predefined measuring point.

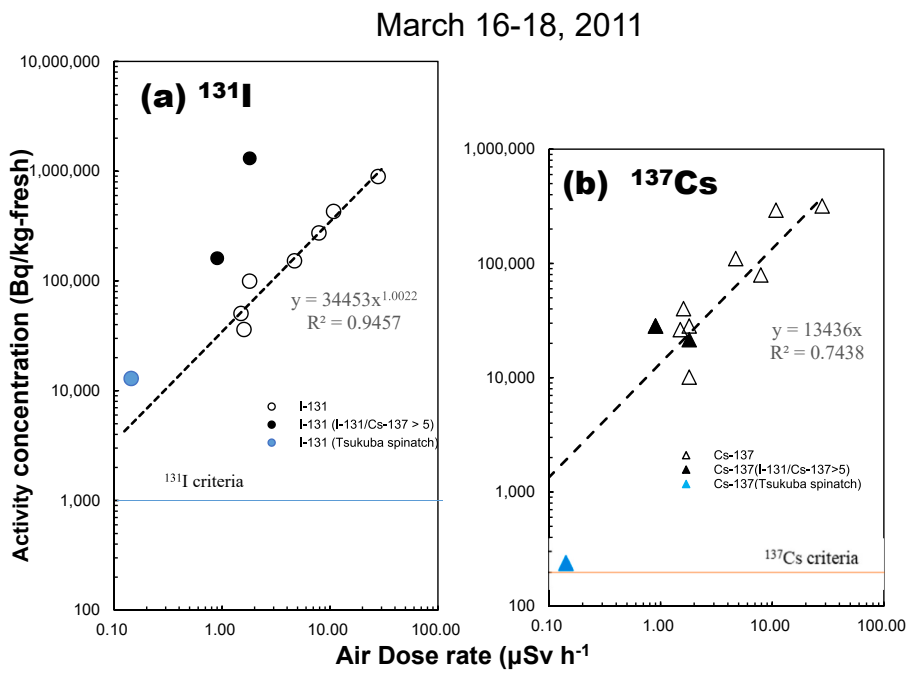



**Fig. 10.** Activity concentrations of $^{137}$Cs and $^{131}$I in grass weeds. A: The weed samples were collected on March 16–18, 2011 (Decay corrected on March 16), B: The weed samples were collected on March 24–25, 2011 (Decay corrected on March 24). The line of criteria is based on IAEA OIL 7.

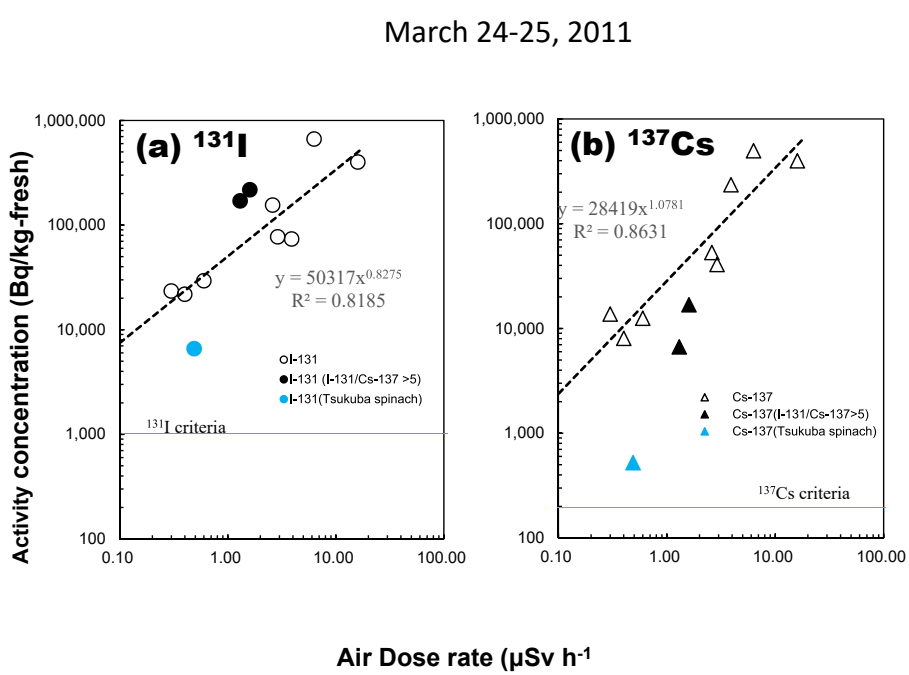



**Fig. 11.** 11 activity concentration of vegetation of $^{131}$I (a) and $^{137}$Cs (b) on March 24–25 (Decay corrected on March 24). The line of criteria is based on IAEA OIL 7.

Therefore, based on the experience of the Fukushima fallout, the $^{137}$Cs and $^{131}$I levels in the early phase of the nuclear accident can be higher than expected in such an air dose rate, especially where the dry-fallout is dominant (Katata et al., 2015). Sy et al. (2015) mentioned that the interception rate $f$ becomes constant at a specific biomass by dry deposition, and leafy vegetables can capture the fallout radionuclides very effectively. Therefore, in the immediate period, the direct deposition of the radionuclides to the vegetation is the dominant pathway; therefore, the $T_f$-*based* estimation of the initial contamination of leafy vegetation (Kirchner and Wirth, 2008) may be underestimated. Although this is just an example of the Fukushima outcome, for safety, vegetation sampling is recommended when the additional dose rate above background is higher than 0.1 μSv/h.

To be prepared for accidents, regular field exercises are essential. From vegetation and soil sampling to the sample measurements, workers need to learn how to collect samples in highly contaminated areas, carry out in situ dose rate measurements, and deal with limited measurement capacity. At all times, safety precautions need to be followed.

The lesson learned from the above discussion and the dataset of weed samples in Fukushima is that we may need to measure the vegetation

sample for food safety if, at the sampling location, the dose rate is above 0.1 μSv/h. Because the detection of airborne $^{131}$I is still not easy (Torii et al., 2013), soil/vegetation sampling remains very important. Further discussion may be needed to define the OIL criteria based on the updated data set.

## 4. Conclusion

Soil sampling approaches for analysing the radioactivity concentration in the soil and fallout on the farmland and the soil and vegetation sampling methods recommended during a nuclear emergency response are described. After the FDNPP accident, immediate soil and vegetation sampling were conducted; however, the detailed protocols were not shown, which made it difficult to analyse the monitoring dataset. In this study, the sampling protocols applied just after the FDNPP were reviewed, and data were analysed. This detailed protocol and the lessons learned from the Fukushima accident can be useful in establishing a possible guideline for soil and vegetation sampling. The data measured immediately after the FDNPP accident were analysed. The activity concentrations of $^{137}$Cs and $^{131}$I for the weed sample were found to be related to the air dose rate at 1 m, which means that the possible contamination of vegetables can be estimated by only measuring the air dose rate. With this analysis, vegetation sampling is recommended where the additional dose rate is above 0.1 μSv/h. For soil and vegetation sampling, the establishment of predetermined sampling points is recommended, as it increases the efficiency of the sampling campaign and data gathering. Analytical capacities, time, people, and cost during the short time after the nuclear emergencies need to be considered, and large-scale exercises are highly recommended. We propose measuring the radionuclides in vegetation for food safety if the additional dose rate exceeds 0.1 μSv/h.

## Declaration of competing interest

No conflict of Interest of this paper.

## Acknowledgment

This paper is supported by the IAEA CRP D15015.

## References

Barnekov, U., Fesenko, S., Kashparov, V., Kis-Benedek, G., Matissov, G., Onda, Y., Sanzharova, N., Tarjan, S., Tyler, A., Varga, V., 2019. Guidelines on Soil and Vegetation Sampling for Radiological Monitoring Purposes. IAEA TRS, p. 247, 486.

Baverstock, K., Egloff, B., Pinchera, A., Ruchti, C., Williams, D., 1992. Thyroid cancer after Chernobyl. Nature 359, 21–22.

Champion, D., Korsakissok, I., Didier, D., Mathieu, A., Quelo, D., Groell, J., Quentric, E., Tombette, M., Benoit, J.-P., Saunier, O., Parache, V., Simon-Cornu, M., Gonze, M.A., Renaud, P.H., Cessac, B., Navarro, E., Servant-Perrier, A.-C., 2013. The IRSN's earliest assessments of the Fukushima accident's consequences for the terrestrial environment in Japan. Radioprotection 48, 493–509.

Dercon, G., Mabit, L., Hancock, G., Nguyen, M.L., Dornhofer, P., Bacchi, O.O.S., Benmansour, M., Bernard, C., Froehlich, W., Golosov, V.N., Haciyakupoglu, S., Hai, P.S., Klik, A., Li, Y., Lobb, D.A., Onda, Y., Popam, N., Rafiq, M., Ritchie, J.C., Schuller, P., Shakhashiro, A., Wallbrink, P., Walling, D.E., Zapata, F., Zhang, X., 2012. Fallout radionuclide-based techniques for assessing the impact of soil conservation measures on erosion control and soil quality: an overview of the main lessons learnt under an FAO/IAEA Coordinated Research Project. J. Environ. Radioact. 107, 78–85.

Dercon, G., Blackburn, C., Onda, Y., Shinano, T., Sweeck, L., Lee Zhi Yi, A., Fesenko, S., 2020. Sampling, analysis and modelling technologies for large-scale nuclear emergencies affecting food and agriculture. J. Environ. Radioact. 106174. https://doi.org/10.1016/j.jenvrad.2020.106174.

Fesenko, S., Onda, Y., Shinano, T., Dercon, G. (in this issue) Comparative analysis of the dynamics of radionuclide activity concentrations in plants and external dose rate in the early phase after the Fukushima Daiichi accident. J. Environ.Radioact. (submitted).

Hirose, K., 2012. 2011 Fukushima Dai-ichi nuclear power plant accident: summary of regional radioactive deposition monitoring results. J. Environ. Radioact. 111, 13–17.

IAEA, 1999. Generic Procedures for Monitoring in a Nuclear or Radiological Emergency. IAEA-TECDOC-1092.

IAEA, 2011. Criteria for Use in Preparedness and Response for a Nuclear or Radiological Emergency IAEA Safety Standards Series No. GS-G-2. IAEA pub 1467.

IAEA, 2017. Operational Intervention Levels for Reactor Emergencies. EPR-NPP-OILs, 2017. https://www-pub.iaea.org/MTCD/Publications/PDF/EPR_NPP_OILs_2017_web.pdf.

Ibaraki Prefecture, 2011. Information on the great east Japan earthquake March 2011. https://www.pref.ibaraki.jp/earthquake/bugai/koho/kenmin/important2/20110311eq/2011/03/index.html (accessed April 5, 2020).

IBZ, 2010. Plan Particulier d'Urgence et d'Intervention Provincial – Problématique des zones d'intervention Sectorisation et Vectorisation : Principes Directeurs.

Katata, G., Chino, M., Kobayashi, T., Terada, H., Ota, M., Nagai, H., Kajino, M., Draxler, R., Hort, M.C., Malo, A., Torii, T., Sanada, Y., 2015. Detailed source term estimation of the atmospheric release for the Fukushima Daiichi Nuclear Power Station accident by coupling simulations of an atmospheric dispersion model with an improved deposition scheme and oceanic dispersion model. Atmos. Chem. Phys. 15, 1029–1070.

Kato, H., Onda, Y., 2018. Determining the initial Fukushima reactor accident-derived cesium-137 fallout in forested areas of municipalities in Fukushima Prefecture. J. For. Res. 23, 73–84. https://doi.org/10.1080/13416979.2018.1448566.

Kato, H., Onda, Y., Teramage, M., 2012. Depth distribution of $^{137}$Cs, $^{134}$Cs, and $^{131}$I in soil profile after Fukushima Daiichi nuclear power plant accident. J. Environ. Radioact. 111, 59–64.

Kato, H., Onda, Y., Gao, X., Sanada, Y., Saito, K., 2019. Reconstruction of a Fukushima accident-derived radiocesium fallout map for environmental transfer studies. J. Environ. Radioact. 210, 105996. https://doi.org/10.1016/j.jenvrad.2019.105996.

Khomutinin, Y.V., Fesenko, S., Levchuk, S., Kashparov, V. (in this issue) Optimising soil sampling strategies for emergency response: 1. Soil. J. Environ. Radioact. (submitted).

Kirchner, G., Wirth, E., 2008. Operational intervention levels (OILs)–A tool to overcome differences in intervention levels? Proceeding of the International Congress of the International Radiation Protection Association (IRPA) 19–24.

MEXT, 1983. Environmental sample collection method: radioactivity measurement method series 16 (in Japanese). https://www.kankyo-hoshano.go.jp/series/lib/No16.pdf (accessed May 20, 2020), 142.

MEXT, 2011a. Readings of Environmental Monitoring Samples (Weed). MEXT (2011a). http://radioactivity.nsr.go.jp/ja/contents/7000/6416/24/Readings_of_environmental_monitoring_sample_June1,2011-%20December31,2011_20121101.pdf (accessed May 20, 2020).

MEXT, 2011b. Readings of early stage of the soil and environmental monitoring sample. http://emdb.jaea.go.jp/emdb/assets/site_data/ja/associated/1020302000/ref_rel_20110317.pdf (accessed April 5, 2020).

NRA, 2017. Nuclear emergency response guideline (in Japanese). http://www.nsr.go.jp/data/000024441.pdf (accessed April 5, 2020).

Ohse, K., Kihou, N., Kurishima, K., Inoue, T., Taniyama, I., 2015. Changes in concentrations of I-131, Cs-134 and Cs-137 in leafy vegetables, soil and precipitation in Tsukuba City, Ibaraki, Japan, in the first 4 months after the Fukushima Daiichi nuclear power plant accident. Soil Sci. Plant Nutr. 61, 225–229.

Onda, Y., Kato, H., Hoshi, M., Takahashi, Y., Nguyen, M., 2015. Soil sampling and analytical strategies for mapping fallout in nuclear emergencies based on the Fukushima Dai-ichi Nuclear Power Plant accident. J. Environ. Radioact. 139, 300–307.

Pröhl, G., 2009. Interception of dry and wet deposited radionuclides by vegetation. J. Environ. Radioact. 100, 675–682.

Renaud, P.H., Simon-Cornu, M., Gonze, M.-A., Morlon, C., Parache, V., Orsakissok, I., Navarro, E., Cessac, B., Rannou, A., Champion, D., 2012. Contamination of Japanese foodstuffs of terrestrial origin after the Fukushima nuclear accident and related dose assessments Part 1: foodstuff contamination. Radioprotection 48, 493–509.

Saito, K., Tanihata, I., Fujiwara, M., Saito, T., Shimoura, S., Otsuka, T., Onda, Y., Hoshi, M., Ikeuchi, Y., Takahashi, F., Kinouchi, N., Saegusa, J., Seki, A., Takemiya, H., Shibata, T., 2015. Detailed deposition density maps constructed by large-scale soil sampling for gamma-ray emitting radioactive nuclides from the Fukushima Daiichi Nuclear Power Plant accident. J. Environ. Radioact. 139, 308–319.

Sy, M.M., Ancelet, S., Henner, P., Hurtevent, P., Simon-Cornu, M., 2015. Foliar interception of radionuclides in dry conditions: a meta-analysis using a Bayesian modeling approach. J. Environ. Radioact. 147, 63–75.

Takahashi, J., Tamura, K., Suda, T., Matsumura, R., Onda, Y., 2015. Vertical distribution and temporal changes of $^{137}$Cs in soil profiles under various land uses after Fukushima Dai-ichi Nuclear Power Plant Accident. J. Environ. Radioact. 139, 351–361.

Torii, T., Sugita, T., Okada, C., Reed, M., Blumenthal, D., 2013. Enhanced analysis methods to derive the spatial distribution of $^{131}$I deposition on the ground by airborne surveys at an early stage after the Fukushima Daiichi Nuclear Power Plant accident. Health Phys. 105, 192–200.

UNSCEAR, 2000. Annex A: dose assessment methodologies. Sources Eff. Ioniz. Radiat.

Yamamoto, M., Takada, T., Nagao, S., Koike, T., Shimada, K., Hoshi, M., Zhumadilov, K., Shima, T., Yoshimura, K., Saegusa, J., Sanada, Y., 2020. Initial decrease in the ambient dose equivalent rate after the Fukushima accident and its difference from Chernobyl. Sci. Rep. 10, 1–9. https://doi.org/10.1038/s41598-020-60847-0.

Yoshimura, K., Saegusa, J., Sanada, Y., 2020. Initial decrease in the ambient dose equivalent rate after the Fukushima accident and its difference from Chernobyl. Sci. Rep. 10, 3859 https://doi.org/10.1038/s41598-020-60847-0.

Zapata, F., 2002. Handbook for the Assessment of Soil Erosion and Sediment Using Environmental Radionuclides. Kluwer, p. 219.