

The Series of Environmental Radioactivity Measuring Methods

AN-I-E
(No.15)

Radioiodine Analysis in Emergencies

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Radiation Monitoring Division
Radiation Protection Department
The Secretariat of
Nuclear Regulation Authority

This document was written in Japanese and translated into English.
The Japanese version is the original and the English version was written as a reference.

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Chapter 1 Introduction

No.15 “Radioiodine Analysis in Emergencies (AN-I-E)” (Secretariat of Nuclear Regulation Authority, published as Science and Technology Agency, 1977) was established as a rapid measurement method to estimate the radioiodine concentration in environmental samples in the vicinity of nuclear facilities when an abnormal situation occurs and a large amount of radioiodine is or may be released outside the facility. The document was revised in 2002 in line with latest changes, such as the improvement of measurement instruments, changes in units, and revisions in indicators for food and drink intake restrictions in disaster prevention guidelines (“Regulatory Guide: Emergency Preparedness for Nuclear Facilities” (Nuclear Safety Commission decision, June 1980, revised in June 2001)).

The 2002 revision stipulates that a simple measurement (in-situ measurement) shall be performed at the collection site using a NaI(Tl) scintillation survey meter for screening collected samples, which are then taken to a monitoring center or other analysis laboratory to perform a more accurate measurement (laboratory measurement) using a germanium semiconductor detector. In addition, the types of samples were limited to four—air, drinking water, bovine milk, and leafy greens—considering inhalation exposure and intake restrictions.

However, the accident at the TEPCO Fukushima Daiichi Nuclear Power Plant caused by the Great East Japan Earthquake on March 11, 2011 (hereinafter referred to as “1F accident”) led to the establishment of the Guideline for Emergency Preparedness and Response (Nuclear Regulation Authority, October 2012; partially revised on July 6, 2022)¹⁾, under which emergency monitoring shall be conducted. In situ measurements were not actually conducted during emergency monitoring at the time of the 1F accident because preparation for monitoring takes time due to factors, such as the need to use an I-131 standard solution to calibrate the NaI (Tl) scintillation survey meter. Therefore, it was not mentioned in the “Emergency Monitoring (Supplementary Reference Material for The Guideline for Emergency Preparedness and Response)”. (Radiation Monitoring Division, Secretariat of Nuclear Regulation Authority; partially revised on December 21, 2021)²⁾ (hereinafter referred to as “Emergency Supplementary Reference Material”), which was developed afterward and wherein the details of emergency monitoring were stipulated.

In light of the above, the description of this measurement method was revised to implement emergency monitoring, in accordance with descriptions in the Emergency Supplementary Reference Material.

The main changes in this revised document are listed below.

- Regarding nuclides to be measured, it was stated that only I-131 should be evaluated even when measurement was performed using the germanium semiconductor detector. However, I-132 and I-133 were observed during the 1F accident, and because they were also listed as target nuclides in the Emergency Supplementary Reference Material, it was decided to list them in this document as they can be evaluated at the same time as I-131.
- The types of samples included previously were air, drinking water, leafy vegetables, and bovine milk, with the objective of considering internal exposure through inhalation and ingestion; nevertheless, soil and fallout (rainwater) were also mentioned, with the aim of understanding the extent of the release and range of effects.
- Although no priority was set for sample types, priorities were clearly stated because in initial emergency monitoring, the sampling of air, drinking water, and soil has to be given priority as the situation progresses.
- For air sampling, only transportable air samplers were mentioned, but the Emergency Supplementary Reference Material stipulates the installation of iodine samplers with auto sample changers that enable automatic sampling via remote control and air monitors.
- On-site measurements using a NaI (Tl) scintillation survey meter were described for screening samples to determine whether or not laboratory measurements using a germanium semiconductor detector should be performed. This step was not implemented at the time of the 1F accident due to the time required for measurement preparation, and there were concerns that long-term outdoor tasks, such as sample collection,

in an emergency could expose workers to radiation and contaminate equipment. This step was also not included in the current Emergency Supplementary Reference Material. Therefore, this has been deleted, with the basic rule being that collected samples should be brought back and measured using a germanium semiconductor detector.

- In the case of atmospheric measurements using a germanium semiconductor detector, the only description given was that the measurement sample was a combination of a glass fiber filter that collects particulate iodine and an activated carbon cartridge that collects gaseous iodine. However, the Emergency Supplementary Reference Material states that the glass fiber filter paper and activated carbon cartridge are to be measured separately to determine the ratio of particulate iodine to gaseous iodine. Therefore, the method of separate measurement was mainly described. Considering the tight machine time of the germanium semiconductor detector in emergency situations, a combined measurement method was also described.

The structure of this document is as follows: Chapter 1 presents an introduction, Chapter 2 describes sample collection and sample preparation methods for measurements using a germanium semiconductor detector for each sample type, and Chapter 3 explains measurements using a germanium semiconductor detector. In addition, as references, a study on measurement and analysis using a germanium semiconductor detector, a case study of radioiodine detection at the 1F accident, and the estimation of the I-131 radioactivity concentration using iodine isotope ratios were described.

Although for the measurement method presented in this document, a series of operations for collecting and measuring radioiodine was described, please refer to the Series of Environmental Radioactivity Measuring Methods for details on each operation.

- Sampling:
 - No. 16 “Method for the sampling of Environmental Materials (AS)”³⁾
 - No. 35 “Generic Procedures for Environmental Sampling in Emergencies (AS-E)”⁴⁾
 - No. 36 “Method for the Measurement of Radioactive Materials in Air (AD-N·E)”⁵⁾
- Preparation of samples for measurements using a germanium semiconductor detector:
 - No. 24 “Preparation of Samples for Gamma-ray Spectrometry in Emergencies (AP-E)”⁶⁾
- Measurements using a germanium semiconductor detector:
 - No. 7 “Gamma-ray Spectrometry using a Germanium Semiconductor Detector (AI-Ge)”⁷⁾
 - No. 29 “Gamma-ray Spectral Analysis Using a Germanium Detector in Emergencies (AI-Ge-E)”⁸⁾

Chapter 2 Collection of Environmental Samples and Preparation of Samples for Measurements

2.1 Types of Samples

Initial monitoring during an emergency is divided into the following phases depending on how far the emergency situation has progressed: alert, facility-site emergency, and full-scale emergency (i.e., before, during, and after the release of radioactive materials into the atmosphere). The purpose of monitoring conducted in each phase is to collect information on the environmental radiation level due to a nuclear disaster, to provide a reference for making decisions on the implementation of protective measures based on the OIL*, and to provide a basis for evaluating the radiation impact on residents and the environment due to the nuclear disaster.

The initial monitoring flow is shown in Figure 2-1. Initially, in the event of an emergency at a facility site, measurements of radioactive iodine in air are ordered (measurement preparation in the case of alert), and monitoring is conducted continuously thereafter. Subsequently, in the event of a full-scale emergency, in which radioactive materials begin to be released into the environment, drinking water will be collected and analyzed. Following the release of radioactive materials into the environment, soil, food and beverages produced in the area, as well as drinking water will be sampled and analyzed. Soil sampling and analysis are performed to confirm the spread of radioactive materials.

The sampling and analysis of air, drinking water, and soil are prioritized based on the nature of initial monitoring conducted during the emergency. In addition, the Emergency Supplementary Reference Material states that other environmental samples are to be collected as well. Therefore, various food samples (bovine milk; leafy greens; and other agricultural, livestock, and marine products) were also described in this method.

As for fallout (rainwater), when the Liaison Conference on Counter-measures for Radiation established by the Cabinet instructs to strengthen monitoring at the time of a nuclear-related event, samples collected by local governments and related organizations nationwide will be analyzed using γ -ray spectrometry; therefore, this measurement method was also included.

* OIL: Operational intervention level, expressed in measurable values such as the air radiation dose rate and radioactive material concentrations in environmental samples.

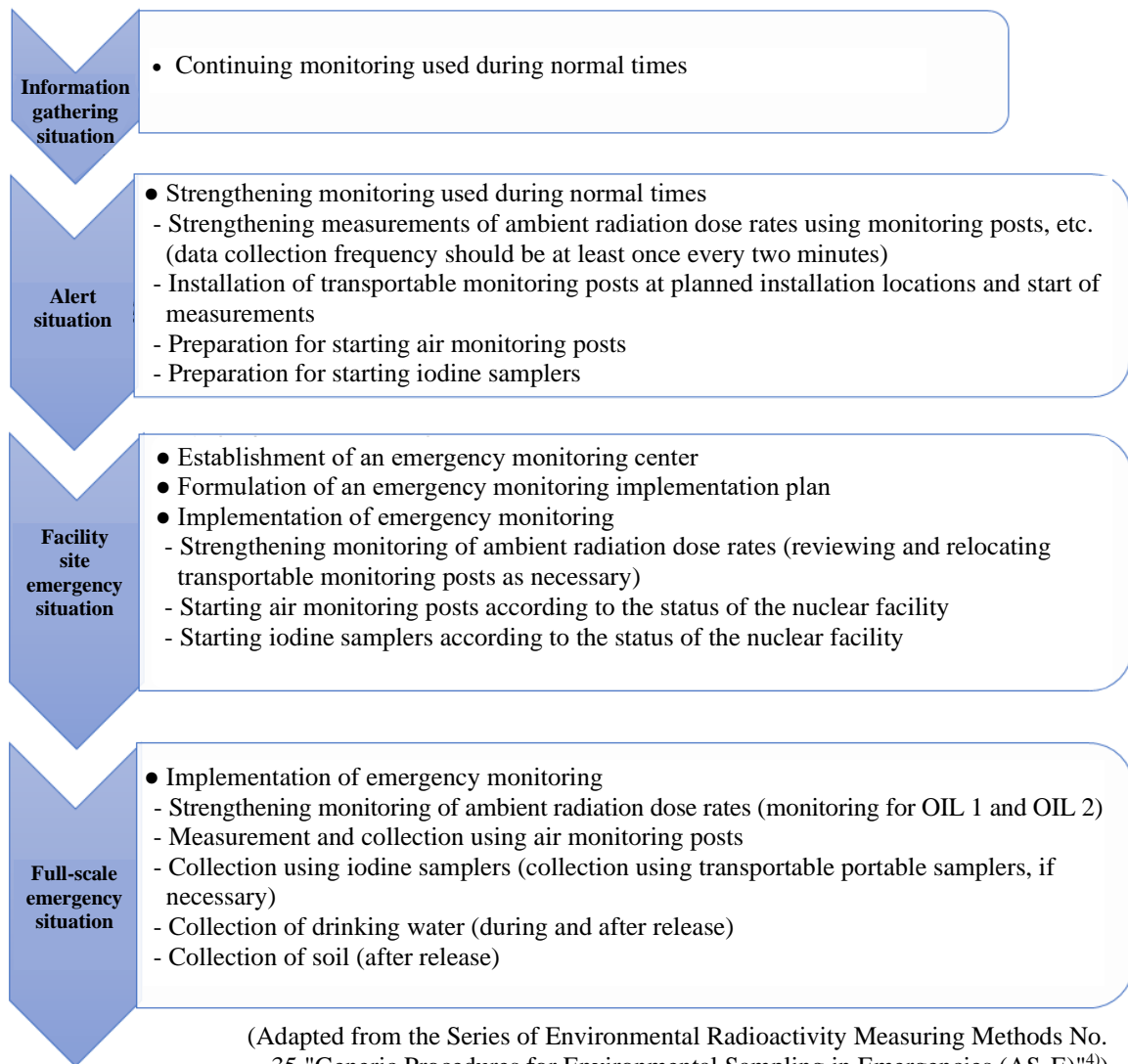


Figure 2-1 Initial monitoring flow

2.2 Collection and Preparation of Samples for Measurements

This section describes the procedure for steps from sample collection to the preparation of the measurement sample for γ -ray spectrometry for each sample type. Refer to The Series of Environmental Radioactivity Measuring Methods No. 35 “Generic Procedures for Environmental Sampling in Emergencies (AS-E)”⁴⁾ and No. 36 “Method for Measurement of Radioactive Materials in the Air (AD-N·E)”⁵⁾ for details on sample collection and to No. 24 “Preparation of Samples for Gamma-ray Spectrometry in Emergencies (AP-E)”⁶⁾ for explanations on the preparation of measurement samples.

In addition to methods used for collecting and preparing samples for measurements, a method that can be employed for storing samples after measurements is described here. Short-term storage is utilized for temporary storage for re-measurement preparation, while long-term storage is employed for the analysis of other radioactive nuclides as radioiodine has a short half life (e.g., the half life of I-131 is 8.0 days) and cannot be detected after decay. Furthermore, methods have been developed to estimate I-131 radioactivity in air and soil by analyzing the I-129 concentration (a half life of 1.57×10^7 years) after the decay of short-lived radioiodine (see Information E).

2.2.1 Air

The main purpose of measuring the radioiodine concentration in air is to collect information on environmental radiation due to a nuclear disaster and to provide a basis for evaluating the radiological impact of the nuclear disaster on residents and the environment. Air sampling can be divided into two main categories: fixed type (an air monitor and an iodine sampler with an auto sample changer) that allows automated sampling via remote control and transportable type that enables sampling at any location.

Radioiodine released into the atmosphere has particulate and gaseous components⁹⁾. Therefore, in air sampling, a glass fiber filter paper collects particulate components, while an activated carbon cartridge gathers gaseous components¹⁰⁾. Note that when air is sampled using only the filter paper (e.g., glass fiber filter paper) for air monitoring, only particulate components will be evaluated.

2.2.1.1 Air sampling by an iodine sampler with an auto sample changer

To determine the temporal change in the radioiodine concentration in air, an iodine sampler equipped with the glass fiber filter paper and activated carbon cartridge is used with an auto sample changer, which continuously samples air and changes absorbents at regular intervals.

[Equipment overview]

The iodine sampler equipped with the glass fiber filter paper and activated carbon cartridge is used to continuously collect gaseous and particulate radioiodine. The auto sample changer can automatically change absorbents at set intervals.

Assuming the occurrence of a power failure, the iodine sampler must be able to operate continuously without refueling or re-energizing for at least three days after startup, even if the power supply is interrupted, using, for example, an emergency generator or a battery.

Note: For the glass fiber filter paper, the side without the lattice structure is the front side, which is the surface that collects dust¹¹⁾.

Note: The activated carbon cartridge comprises 50-mesh activated carbon packed in a polyvinyl chloride or stainless steel container, which is covered with a nonwoven fabric or stainless steel mesh on the front and back. To increase iodine collection efficiency, activated carbon is coated with 10% triethylenediamine (TEDA). Potassium iodide is sometimes used as an additive¹¹⁾.

[Collection conditions]

- In general, one sampling station shall be located in every one or two of 16 azimuths centered on the target facility, within a radius of 5–30 km. However, the actual conditions of the population distribution and natural environment shall be taken into consideration when placing sampling stations.

- Height of the sampling port: Approx. 1 m (but may be more than 1 m considering weather and other conditions)
- Suction flow rate: Approx. 50 L/min
- To avoid the influence of condensation on the absorbent, the sample air is heated using a heater controlled at approximately 50°C to reduce relative humidity.
- Air is collected for a fixed period of time (~6 h), and absorbents (the glass fiber filter paper and activated carbon cartridge) are automatically replaced at regular intervals by the auto sample changer.

Note: The auto sample changer shall be designed to have a sufficient filling volume to allow the automatic replacement of 20 or more absorbents as samples must be collected for at least 5 days after the start of operation.

Note: Because absorbents are set in place during normal operation, the adsorption capacity of the activated carbon cartridge, in particular, may deteriorate due to moisture absorption and other factors. Therefore, they need to be replaced periodically (usually about every three months), even if they are not in use. In addition, models with a simple airtight or depressurized storage area for absorbents are commercially available and are designed to withstand long-term storage.

Note: Example of sample volume: $50 \text{ L/min} \times 360 \text{ min} = 18,000 \text{ L} = 18 \text{ m}^3$

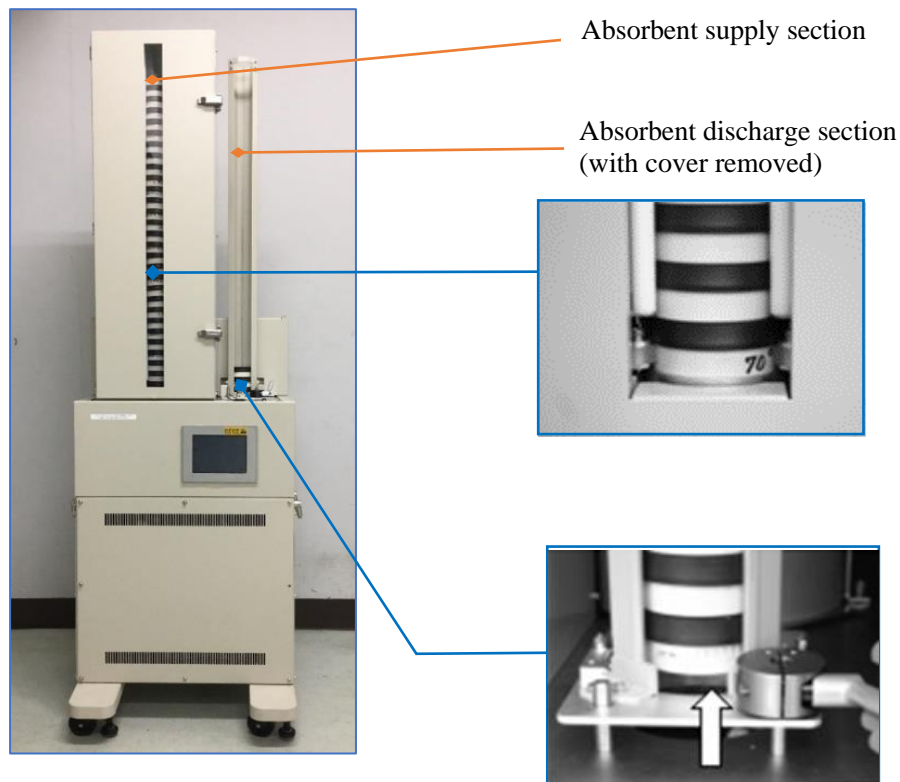


Photo 2-1 Example of an iodine sampler with an auto sample changer

[Sampling]

- 1) As a rule, equipment shall be operated using remote control. During a facility-site emergency, radioiodine in air will be monitored in accordance with the emergency monitoring implementation plan, depending on the status of the facility as the accident progresses. The operation of equipment will be initiated from a monitoring center, etc.
- 2) Retrieve absorbents after collection, while taking care to prevent contamination. Retrieve absorbents for several days of operation or, if instructions are issued, for the period indicated in the instructions.
- 3) Pair retrieved absorbents (the glass fiber filter paper and activated carbon cartridge) by collection time, and

place them in a polyethylene bag.

4) Attach a label with an identification code (or write the code on the bag).

5) Set new absorbents and resume collection. Check the air flow path and suction volume, and confirm that equipment is operating properly.

2.2.1.2 Air sampling with an air monitor

An air monitor draws air into the filter paper and determines the presence or absence of a radioactive plume in the vicinity of the air-monitor installation location by monitoring changes in the concentration of radioactive materials in air over time. In addition, by analyzing the filter paper that has absorbed air using a germanium semiconductor detector, detailed changes in the concentration of radioactive materials in air and their nuclide compositions can be estimated. Because the only absorbent used in the air monitor is the filter paper, the only radioiodine that can be evaluated is particulate iodine.

This method presents a technique for evaluating particulate radioiodine by analyzing the filter paper retrieved from the air monitor using a germanium semiconductor detector. For details on measurement while collecting dust, refer to The Series of Environmental Radioactivity Measuring Methods No. 36 “Method for Measurement of Radioactive Materials in the Air (AD-N·E)”⁵⁾.

[Equipment overview]

Using a continuous dust collection and measurement device equipped with the filter paper, particles are sucked in with air by a pump and collected on the filter paper, and either total alpha or total beta radioactivity or both are measured in the radioactivity detection unit. A glass fiber filter paper or a membrane filter paper is used for collection. A long roll paper is utilized because the filter paper is automatically fed at regular intervals (~1 h).

In the case of a power failure, the system must be able to operate continuously without refueling or re-energizing for at least three days after the startup of the air monitor using an emergency generator, batteries, etc., even if the power supply is interrupted.

[Collection conditions]

- In general, one sampling station shall be located for each of 16 azimuths centered on the target facility, with a radius of 5–10, 10–20, and 20–30 km. However, when installing monitors, consideration shall be given to the actual conditions of the population distribution and natural environment; additionally, care shall be taken to ensure that air monitors are not placed on the same straight line in each direction.
- Height of the sampling port: Approx. 1 m (but may be more than 1 m considering weather and other conditions)
- Suction flow rate: Approx. 50 L/min
- To avoid the influence of condensation on the absorbent, the sample air is heated using a heater controlled at approximately 50°C to reduce relative humidity.
- Air is collected for a certain period of time (about 1 h), and the filter paper is automatically fed.

Note: Example of sample volume: $50 \text{ L/min} \times 60 \text{ min} = 3,000 \text{ L} = 3 \text{ m}^3$



Photo 2-2 Example of an air monitor

[Sampling]

The filter paper is collected via the same procedure as that used by the iodine sampler with an auto sample changer; however, because the filter paper for the air monitor is a long roll paper, the entire roll is collected or only the used portion is cut off for collection.

2.2.1.3 Air sampling with a transportable sampler

This section describes a transportable sampler that can be used to collect air samples at any location. There are two types of transportable samplers: a high-volume air sampler and a low-volume air sampler. However, for radioiodine, a low-volume air sampler is used.

[Equipment overview]

Equipped with a glass fiber filter and an activated carbon cartridge, the sampler allows continuous collection for several days. The suction volume is adjustable at 20–100 L/min.

[Sampling]

- 1) Install the sampler at the collection point so that the air flow is not impeded. When installing the sampler, it shall be adjusted so that the sampler suction port is approximately 1 m above the ground.

Note: Because the collection performance of the glass fiber filter paper and activated carbon cartridge will be degraded if raindrops are sucked in, collection shall be conducted in a roofed area in case of rain so that raindrops do not get into the sampler. If a location with a roof around is not available, hold an umbrella or the like to avoid sucking in raindrops by the adsorbents.

Note: When a transportable generator is used as a power source or multiple samplers are utilized together, care shall be taken to ensure that their exhaust does not affect each other.

Note: Because it affects the total volume, the criteria for the direction of installation shall be determined in advance, such as which direction to face with respect to wind or to face the emission source regardless of the wind direction.

- 2) Set the glass fiber filter paper and activated carbon cartridge in the suction port. For the glass fiber filter paper, the side without the lattice structure is the front side, which is the surface that collects dust.
- 3) Once sampling begins, record the time and sampling flow rate and collect air until the desired total volume is reached.

Note: The flow rate of the dust collector shall be set to approximately 50 L/min. If the flow rate is 50 L/min, the total volume will be 1,000 L (= 1 m³) after 20 min of sampling. The sampling flow rate and total

volume shall be in accordance with the instructions, if any.

- 4) When the desired total volume is reached, stop the dust collector and record the time.
- 5) Remove the glass fiber filter paper and activated carbon cartridge from the dust collector, and store them in a polyethylene bag.



Photo 2-3 Example of a low-volume air sampler

2.2.1.4 Preparation of measurement samples

This section describes the preparation of measurement samples using glass fiber filter papers and activated carbon cartridges used to collect air. Although each sample contains a set of the glass fiber filter paper and activated carbon cartridge, the basic method is to measure each adsorbent separately to determine the ratio of particulate to gaseous matter. However, if the machine time is tight due to the number of samples to be measured, they may be measured together as a single sample. Even in such cases, samples to be measured separately shall be determined beforehand, and samples at other points shall be measured together, so as to calculate the ratio and reduce the number of samples measured.

A method for preparing the filter paper from the air monitor as a measurement sample is described as this filter paper is also analyzed with the germanium semiconductor detector. Note that only particulate radioiodine is targeted as the air monitor uses only the filter paper for sample collection.

If there is a time between the collection of air samples and preparation and measurement of measurement samples, it is recommended to store the collected samples in a desiccator to prevent moisture absorption.

[Preparation of measurement samples from the glass fiber filter paper]

- 1) Write or attach a number or other data identifying the sample on a small container, and then measure and record the tare weight of the small container.
- 2) Press the glass fiber filter paper firmly to the bottom of the small container, with the air suction side (dust adhesion side) down, so that gaps are not formed. (Photo 2-4 and Photo 2-5)
- 3) Cover the small container, and measure and record the height of the sample.
- 4) After thoroughly wiping the outside of the container with a paper towel moistened with pure water, ethanol, etc., weigh the small container and subtract the previously obtained tare weight to determine and record the weight of the measurement sample.
- 5) To prevent the contamination of the measuring instrument, cover the small container with a polyethylene bag, remove air, and seal the bag by tying it at the top to ensure that there are no wrinkles at the bottom of the container. Use this as the measurement sample. (Photo 2-6)



Photo 2-4 Sample placed inside the small container

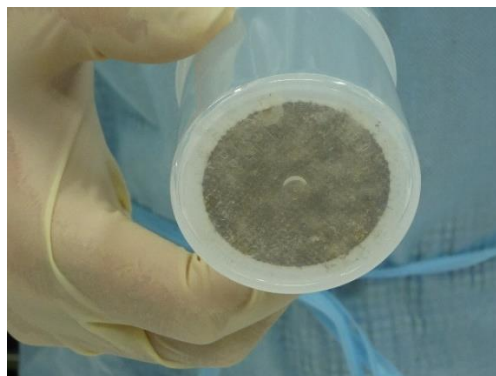


Photo 2-5 Sample placed inside the container, with the suction surface facing the bottom



Photo 2-6 Small containers covered with a polyethylene bag

[Sample preparation for activated carbon cartridges]

In an emergency, to prevent contamination from the absorbent, it is generally not removed from the activated carbon cartridge, and measurements are carried out as is. For analysis method when the efficiency of the germanium semiconductor detector does not correspond to the shape of the activated carbon cartridge, refer to Section 3.1 Adjustment and Calibration of Instrument. For measurements, the activated carbon cartridge should be installed with the air suction side (upstream side where air is introduced) facing the detector to ensure accurate evaluation, considering internal nonuniformity.

- 1) To prevent the contamination of the measuring instrument, cover the activated carbon cartridge with a polyethylene bag, remove air, and seal the bag by tying it at the top to ensure that there are no wrinkles at the bottom on the detector side. Use this as the measurement sample.

When measuring the glass fiber filter paper and activated carbon cartridge together, insert the filter paper into the suction side (detector side) of the cartridge and seal them together in a polyethylene bag.

[Sample preparation for analyzing the filter paper collected from the air monitor]

The filter paper used in the air monitor is a long roll paper. This section describes the procedure for cutting out a six-hour sample according to the sampling interval of the iodine sampler with an automatic sample changer and placing it into a small container. Measurements may be taken at even smaller time intervals. For details, see The Series of Environmental Radioactivity Measuring Methods No. 36 “Method for Measurement of Radioactive Materials in the Air (AD-N·E)”⁵⁾.

- 1) Write or attach a number or other data identifying the sample on a small container, and then measure and record the tare weight of the small container.

- 2) The cut filter paper is folded and placed inside a small container so that the collecting surface is close to the bottom of the small container to ensure the evaluation of the safe side (Photo 2-7).

Note: To prevent cross contamination between dust collection surfaces, it is recommended to protect the cut filter paper with a polyethylene film, etc.

Note: The filter paper may be cut into one-hour portions and placed in layers. In this case, put it in such a way that the dust collection surface faces down.

- 3) Cover the small container, and measure and record the height of the sample.
- 4) After thoroughly wiping the outside of the container with a paper towel moistened with pure water, ethanol, etc., weigh the small container and subtract the previously obtained tare weight to determine and record the weight of the measurement sample.
- 5) To prevent the contamination of the measuring instrument, cover the small container with a polyethylene bag, remove air, and seal the bag by tying it at the top to ensure that there are no wrinkles at the bottom of the container. Use this as the measurement sample.



Photo 2-7 Example of placing a long roll paper cut into six-hour portions
(Example of a roll packed into a small container and folded to make a long strip so that the dust collecting surface is close to the bottom)

[Storage of samples after measurements (short and long terms)]

- Samples after measurements may be subjected to radiochemical or other analysis as necessary and shall be stored with care to avoid contamination.
- After completing measurement, store the sample in its bag in another polyethylene bag or other container.

2.2.2 Drinking water

Radioactive substances that have migrated into drinking water are transferred to the human body through the ingestion of water; therefore, understanding radioactivity is important for evaluating radiation exposure doses. Sampling and analysis of drinking water are conducted to collect information on the environmental radiation situation due to a nuclear disaster and to provide a basis for evaluating radiation effects on residents and the environment as well as for making decisions on the implementation of protective measures. Drinking water sampling will be a priority in the initial monitoring.

2.2.2.1 Drinking water collection

To ascertain the effect of radioactive materials on drinking water, when a release of radioactive materials is confirmed, drinking water supplied from public waterworks and small water supply systems located within the PAZ* and UPZ** that are at the risk of being contaminated will be promptly sampled and analyzed.

* PAZ: Precautionary action zone

**UPZ: Urgent protective action planning zone

Candidate sampling points and priorities shall be determined in advance, considering local conditions in the water catchment area and water distribution system. Furthermore, if the air radiation dose rate around the water source is high, intensive monitoring shall be conducted by increasing the frequency of sampling and number of target points using that particular water source.

For the safety of monitoring personnel during the release of radioactive materials, samples shall be taken from indoor faucets.

[Drinking water collection]

- 1) Open the faucet or allow water intake at the collection site wide open and discharge water for at least 1 min.
- 2) Wash the collection vessel with the water to be collected. Dispose wastewater from washing in a place where it will not affect collection.
- 3) Collect at least 2 L directly from the faucet or intake into the vessel.

Note: For measurement using the germanium semiconductor detector, a small container or a Marinelli beaker shall be used, and a total of approximately 2 L shall be collected. In addition, when performing radiochemical and other analysis, it is necessary to increase the volume of collected samples, considering the volume of each sample to be analyzed.

Note: No additives are added in initial monitoring as rapidity is a priority. If there is an addition instruction because it will take several days to measure the sample, add 80–100 mg sodium thiosulfate per 1 L to prevent radioiodine from adhering to the inner walls of the container (it is recommended to collect samples separately for radioiodine and for other nuclides)⁴).

- 4) Cover the collection vessel, and wipe around the vessel with a paper towel.
- 5) Record the time and conditions of collection.
- 6) Attach a label with an identification code to the vessel containing the sample (or clearly write the code on the vessel).
- 7) Seal the lid with a plastic tape, etc., and place the vessel in a polyethylene bag (inner bag). Further pack the sample in another bag (outer bag).

Table 2-1 shows the collection of drinking water according to the status of radioactive material release.

Table 2-1 Drinking water sampling for initial monitoring

| | During the release | After the release (immediately after it subsided) |
|------------------------|---|--|
| Location of collection | <ul style="list-style-type: none"> Collect purified water from a water purification plant or tap water from a public facility (such as a town hall or a branch office) whose water supply comes from a water source within the UPZ (including the PAZ), which is easily contaminated by radioactive materials, such as surface water. The target areas are areas where collection can be done without going outdoors. | <ul style="list-style-type: none"> Collect purified water (or raw water if it is difficult to collect purified water) from all water treatment plants that take water from a water source within the UPZ (including the PAZ), which is easily contaminated by radioactive materials, such as surface water. |
| Sampling frequency | At least once a day | At least once a day |
| Sample volume | As a rule, 2 L or more | As a rule, 2 L or more |
| Remarks | <ul style="list-style-type: none"> To avoid the exposure of persons collecting samples, in principle, samples shall be collected by persons working at the sampling locations (water purification plant, etc.). Select approximately one water purification plant for each municipality. If the area is large due to the mergers of municipalities, multiple locations shall be selected. | <ul style="list-style-type: none"> Establish a system from where the facility manager of the water treatment plant or other appropriate persons can collect samples. If it is difficult to collect samples from all water treatment plants, priority shall be given to water treatment plants that serve a large population and that draw water from sources with high air radiation levels in the surrounding area. |

Source: *About emergency monitoring*(Supplementary Reference Material for The Guideline for Emergency Preparedness and Response²⁾)

Because drinking water comes from various sources (river water, groundwater, etc.) and purification processes exist in large water supply systems, tap water may be a sample that has been decontaminated to some extent.

The faucet or intake from which drinking water will be collected during emergency monitoring shall be determined in advance and clearly indicated on drawings or photographs.

2.2.2.2 Preparation of samples for drinking water measurements

This section describes a method for preparing and storing drinking water as a sample for measurements in emergency situations. The cases where Marinelli beakers or small containers are used as measurement containers are described separately.

[When using a Marinelli beaker]

- 1) Place the inner bag inside the Marinelli beaker without gaps (use a disposable bag to avoid the contamination of the inner walls of the measurement container).
- 2) Write or attach a number or other data identifying the sample on the Marinelli beaker, and then measure and record the tare weight of the Marinelli beaker, lid, and inner bag.
- 3) Mix the sample thoroughly, and transfer it to the Marinelli beaker prepared in step 2), aligning it with the marked line.

A piston pipette can be used when aligning with the marked line.

- 4) Weigh the Marinelli beaker and lid together with the sample, subtract the tare weight to obtain the weight of the measurement sample, and record it.
- 5) Seal the top of the inner bag of the Marinelli beaker using a plastic tape. A rubber band or a tying band may be used in place of the plastic tape (Photo 2-9).
- 6) Place the lid on the Marinelli beaker, and seal the gap between the body and lid with a plastic tape (Photo 2-

10 and Photo 2-11).

- 7) Wipe the outside of the Marinelli beaker thoroughly with a paper towel moistened with pure water or ethanol.
- 8) To prevent the contamination of the measuring instrument, place the Marinelli beaker in a polyethylene bag, remove air, and seal the bag by tying it at the top. Use this as the measurement sample. (Photo 2-12).



Photo 2-8 Filling a sample into the Marinelli beaker



Photo 2-9 Closing the inner bag with a plastic tape



Photo 2-10 Fixing the lid with a plastic tape



Photo 2-11 Fixing the lid with a plastic tape



Photo 2-12 Marinelli beaker covered with a polyethylene bag

[When using a small container]

- 1) Wrap a sealing tape around the threaded portion of the small container to prevent leakage (Photo 2-13).
- 2) Write or attach a number or other data identifying the sample on the small container, and then measure and record the tare weight of the small container.
- 3) Mix the sample thoroughly, and transfer ~80 g of it into the small container prepared in step 2) (Photo 2-14).
A piston pipette can be used to adjust the liquid volume.
- 4) Cover the small container, and measure and record the height of the sample.

- 5) Wipe the outside of the container thoroughly with a paper towel moistened with pure water or ethanol.
- 6) Weigh the small container, subtract the tare weight to obtain the weight of the measurement sample, and record it.
- 7) To prevent the contamination of the measuring instrument, cover the small container with a polyethylene bag, remove air, and seal the bag by tying it at the top to ensure that there are no wrinkles at the bottom of the container. Use this as the measurement sample. (Photo 2-15).



Photo 2-13 Wrapping a sealing tape around the threaded portion of the small container



Photo 2-14 Small container containing the sample



Photo 2-15 Small container covered with a polyethylene bag

[Storage of samples after measurement]

• Short-term

- 1) The measured sample in the Marinelli beaker shall be kept in the inner bag and put in another polyethylene bag or container for storage.
- 2) The measured sample in the small container shall be kept in the container and put in another polyethylene bag or container for storage.
- 3) It is recommended to store samples at low temperatures to prevent decomposition. Therefore, they shall be stored in a refrigerator or a cool, dark place.

• Long-term

- 1) It is recommended to store the samples at low temperatures to prevent decomposition. Therefore, they shall be stored in a refrigerator or a cool, dark place.

2.2.3 Soil

Soil sampling in initial monitoring is conducted after the release of radioactive materials into the atmospheric environment to collect and measure potentially contaminated samples and to obtain information on the spread and nuclide compositions of radioactive materials deposited on the ground as a result of nuclear disaster. Understanding the radionuclide composition may enable to obtain relevant information for protective measures and to predict and evaluate the decay of dose rates based on the half life and other factors.

2.2.3.1 Soil sampling

The collection points for soil samples should be selected as described below, taking into consideration the measurement points for air radiation dose rates and atmospheric radioactive material concentrations.

- Locations where OIL2 criteria have exceeded among locations where fixed stations or electronic dosimeters were installed.
- Locations where the concentrations of radioactive materials in air are measured, such as the sites where air monitors are installed.
- Locations where in situ measurements are performed (analysis of radionuclides emitting α - and β -rays shall be performed as necessary).

The sampling frequency should be as follows.

First sampling: Immediately after the release of radioactive materials stops and radioactive materials are deposited on the ground.

→ Second sampling: Approximately one week after the first sampling.

→ Third and later sampling: Based on the measurement results of the sample taken at the second sampling, the sampling plan shall be reviewed and implemented.

When performing soil sampling, it is important to record the area and depth of sampling. In particular, an area-specific inventory of radionuclides is required to confirm the relationship between the air radiation dose rates and radionuclides deposited on the ground surface during an emergency. Another advantage is that conversion coefficients from the IAEA and other sources can be used to determine the relationship between the dose rate and radionuclide concentration. In the 1F accident, the deposition of radioactive materials on the ground surface immediately after their release was generally less than 5 cm on the soil surface in the case of uncultivated soil¹²⁾. Therefore, if surface soil is sampled, the inventory of deposited radionuclides can be almost completely determined, along with the initial contamination distribution. Thus, as radionuclides are likely to be deposited on the soil surface in the early deposition stage, radioactivity per sample weight (Bq/kg) tends to be a smaller value at deeper sampling depths due to dilution by soil in lower layers. Therefore, if the purpose is to determine the initial contamination distribution, it is appropriate to use the surface layer as the sampling depth.

The following is an example of soil sample collection using a small container used in γ -ray spectrometry. For details on the number of samples to be collected per site when taking into consideration representativeness and the method of using sampling tools such as soil collectors, refer to the Series of Environmental Radioactivity Measuring Methods No. 35 “Generic Procedures for Environmental Sampling in Emergencies (AS-E)”⁴⁾.

[Soil sampling]

- 1) Determine the area of sampling so that the specified area can be secured.

Note: The sampling area shall be at least 1 m².

- 2) Check the condition of the soil to be sampled.

Note: If the ground surface is covered with grass, weeds, snow, etc., they shall be sampled separately from soil. Devise a method to identify them, such as assigning a branch number (such as -2) of the number of the soil sample at that location, to ensure that the related measurement results can be evaluated together.

- 3) Place the collection device (a small container with a clear bottom) perpendicular to the ground surface,

- and push it in with a hand or a foot to a depth of 5 cm (if the soil is hard, aim for 2 cm).
- 4) Use a disposable spoon or a similar device to scoop up the soil in the collection device.
 - 5) Wipe off any soil adhering to the screw cup with paper towels, cover the collection container, wipe the surrounding area with a paper towel, and keep the container aside.
 - 6) Record the time and conditions of collection.
 - 7) Attach a label with an identification code to the container with the sample (or clearly write the code on the container).
 - 8) Seal the lid with a plastic tape, and place the container in a polyethylene bag (inner bag). Further pack the sample in another bag (outer bag).

2.2.3.2 Preparation of measurement samples

In initial monitoring, samples are generally taken in small containers and measured as is without pretreatment, but this section describes some precautions if pretreatment is used (measurements may be made after pretreatment if the goal is to achieve more accurate quantification). Drying is not performed to prevent iodine volatilization and contamination. Wet soil in a small measurement container is used as the measurement sample.

[Preparation of soil samples for measurements]

- 1) Remove large plants, roots, stones, and gravels that are mixed in (Photo 2-16).
- 2) Stir the bag containing the sample from the top, and mix it thoroughly (Photo 2-17).
- 3) Measure the tare weight of the measuring container.
- 4) Place an appropriate amount of wet soil in the measuring container. The remaining sample is used to determine the dry soil fraction.
- 5) Lightly compress the top surface of the sample to form a cylindrical shape, place the lid on the measuring container, and measure the filling height to prepare the measurement sample (Photo 2-18).
- 6) Wipe the outside of the container thoroughly with a paper towel moistened with pure water or ethanol.
- 7) Weigh the small container, subtract the tare weight to obtain the weight of the measurement sample, and record it.
- 8) To prevent the contamination of the measuring instrument, cover the small container with a polyethylene bag, remove air, and seal the bag by tying it at the top to ensure that there are no wrinkles at the bottom of the container. Use this as the measurement sample.



Photo 2-16 Removal of foreign matter



Photo 2-17 Stirring and mixing of sample

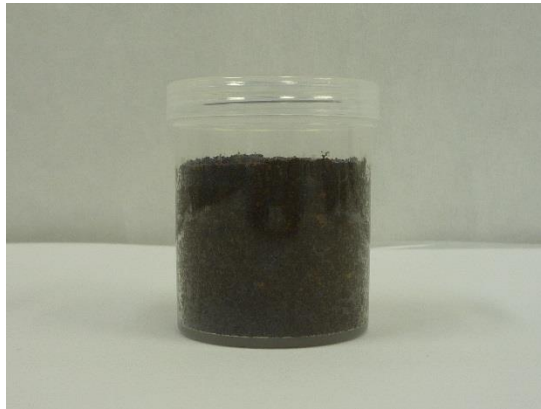


Photo 2-18 Filling small containers with samples

[Storage of samples after measurement]

- Short-term

- 1) Store the sample in a small container placed in another polyethylene bag or container.

- Long-term

- 1) Store the sample in a small container in another polyethylene bag or container, or dry the sample. However, because iodine volatilizes when exposed to heat, if there is a possibility that the I-129 concentration will be analyzed, soil should be stored in a cool, dark place while still wet or dried at around 70°C¹³⁾⁻¹⁵⁾.
- 2) When storing dried samples, place them in a polyethylene bag or container and store them in a low-humidity environment, such as in a desiccator or take measures to prevent contact with air as much as possible.

2.2.4 Various food samples (bovine milk; leafy greens; and other agricultural, livestock, and marine products)

The Emergency Supplementary Reference Material²⁾ describes the measurement of the concentrations of radioactive materials in environmental samples and classifies such samples into precipitation, soil (including land water, seawater, river bottom soil, lake bottom soil, sea bottom soil, and indicator organisms, in addition to soil), and food and drink.

Food and drink are those that are primarily served for eating and drinking or act as a substitute for leafy greens, considering local food culture and other factors. Among environmental samples, priority is given to the collection of soil and drinking water for providing a reference for making decisions on protective measures. However, other environmental samples shall also be collected and analyzed for providing a basis for evaluating radiation effects on residents and the environment and collecting information on the environmental radiation situation.

Bovine milk, leafy greens, and other agricultural, livestock, and fishery products that are collected and analyzed when necessary as environmental samples, other than soil and drinking water, are described below.

A. Bovine milk

Among “milk” samples, bovine milk is representative because of the well-known exposure pathway of grass–cow–milk–humans. In an emergency, the contamination of milk with radioactive iodine is important because it will be the main route of exposure to the public.

The sampling and analysis of bovine milk will be conducted as necessary to provide a basis for making decisions on the implementation of protective measures and for evaluating the radiological impact of a nuclear disaster on residents and the environment.

A.1 Collection of bovine milk

This section describes the procedure for bovine milk collection by monitoring personnel at a ranch or other location. Bovine milk shall be collected from dairy cows that have consumed grass or water from the area surrounding the collection point, not from cows fed with stored feed. Another bovine milk collection involves collecting milk from the combined milk of dairy cows (a mixture of 4–5 cows’ raw milk as it is milked from dairy cows).

The consent and cooperation of the producer (dairy farmer) are required when bovine milk is collected. When developing an emergency monitoring plan, it is recommended that producers selected as candidates for sampling be asked for permission and cooperation in preparation for emergency monitoring during normal times.

[Collection of bovine milk]

- 1) Wash the collection vessel with the milk to be collected (if necessary). Dispose of wastes from washing in a place where it will not affect collection.
- 2) Collect ~2 L of bovine milk in the vessel.
- 3) Cover the collection vessel, and wipe the surrounding area with a paper towel.
- 4) Record the time and conditions of collection.

Note: If the milking date is different from the sampling date, check and record the milking date.

- 5) Attach a label with an identification code to the container with the sample (or clearly write the code on the container).
- 6) Seal the lid with a plastic tape, and place the container in a polyethylene bag (inner bag).
- 7) Place the sample container inside another bag (i.e., outer bag).

Note: Avoid vibration and shock as much as possible when transporting samples as they may cause the separation of water and the fat content.

- 8) Verify packaging, records, and photographs.
- 9) Place the sample in a cool container.

A.2 Preparation of bovine milk samples

Refer to 2.2.2.2 Preparation of samples for drinking water measurements for information on pretreatment and storage methods used to prepare samples for the measurement of raw milk and commercial milk that have been pasteurized, homogenized, or otherwise processed.

[Storage of samples after measurement]

If drying and ashing is performed for long-term storage, care shall be taken to avoid contamination, and ashed samples shall be stored in the same manner as dried soil.

B. Other food samples (leafy greens and other agricultural, livestock, and marine products)

When collecting and analyzing leafy greens, etc., to determine the initial contamination status, care shall be taken to ensure that outer leaves are not removed and that radioactive materials deposited on the leaf surface do not spill out or touch the surrounding area in the process. In addition, when performing analysis to evaluate internal exposure when ingested as food, sampling shall be conducted according to the form of ingestion. Other agricultural, livestock, and marine products (grains, beans, mushrooms, meat, eggs, dairy products, fish and shellfish, etc.) shall also be collected and analyzed as needed to obtain extensive information necessary to assess the actual radiation dose received by nearby residents and grasp the status of radioactive materials and radiation released into the environment.

Sample collection includes collecting samples from fields, requesting samples from producers, and purchasing samples at the market. When collecting samples from fields by monitoring personnel, refer to The Series of Environmental Radioactivity Measuring Methods No. 16 “Method for sampling of Environmental Materials (AS)”³⁾ and No. 35 “Generic Procedures for Environmental Sampling in Emergencies (AS-E)”⁴⁾. This section describes how to obtain samples by purchasing them at the market.

B.1 Collection of other food samples

Crops that are ready to be shipped shall be selected for sampling, and the planting status and storage/shipping methods of crops at the sample collection site as well as for land use in the area shall be recorded. For meat samples, it is advisable to confirm the distribution route with the producer (it is also acceptable to obtain samples at slaughterhouses). When purchasing samples at the market, purchase samples that meet the conditions described in terms of the type, quantity, place of collection, date of collection, etc.

[Collection of other food samples]

- 1) Obtain the sample of the specified type, quantity, and collection site, and place it in the container.

Note: Obtain the sample in such a way that the edible portion is approximately 1 kg. For specific examples of edible portions, see The Series of Environmental Radioactivity Measuring Methods No. 24 “Preparation of Samples for Gamma-ray Spectrometry in Emergencies (AP-E)”⁶⁾.

- .2) Record information regarding the sample (sample name, collection site, collection date, whether or not the surface was cleaned, etc.).
- 3) Attach a label with an identification code to the container containing the sample (or clearly write the code on the container).
- 4) Seal the lid with a plastic tape, etc., and place the sample in a polyethylene bag (inner bag).
- 5) Pack the sample in another bag (i.e., outer bag).
- 6) Verify packaging, records, and transportation conditions.
- 7) Samples that require refrigeration (meat, dairy products, etc.) should be placed in a cool container.

[Precautions for sample selection and collection]

- Select crops that are harvested in the respective season. As a rule, open-field crops are collected from fields

and other areas that may be contaminated. To assess internal exposure from food that is actually consumed, measurements may not be limited to open-field crops.

- Samples shall be collected continuously under the same conditions.
- The amount of the target part (edible part) and other parts (bones, etc.) shall be determined by considering the ratio of the target part to the whole weight.

B.2 Preparation of other food samples for measurements

A method that uses Marinelli beakers or small containers as measuring containers is described here. This section contains only a representative example. For details, refer to The Series of Environmental Radioactivity Measuring Methods No. 24 “Preparation of Samples for Gamma-ray Spectrometry in Emergencies (AP-E)”⁶⁾.

[When using a Marinelli beaker]

- 1) Remove any parts of the sample that are not to be used for the intended measurement, and cut the sample into pieces of 1–2 cm using a cutter, etc. (Photo 2-19 and Photo 2-20).
- 2) Cover the inside of the Marinelli beaker with a plastic bag without gaps (use a disposable bag to avoid the contamination of the inner walls of the measurement container).
- 3) Write or attach a number or other data identifying the sample on the Marinelli beaker, and then measure and record the tare weight of the Marinelli beaker, lid, and inner bag.
- 4) Pack the sample tightly inside the Marinelli beaker up to the mark, while ensuring homogeneity (Photo 2-21).
- 5) Lightly compress the sample surface, and make it level with the marked line. Subsequently, weigh the sample together with the Marinelli beaker and lid, and subtract the tare weight to obtain and record the weight of the measurement sample.
- 6) Seal the top of the inner bag inside the Marinelli beaker using a plastic tape. Rubber bands or cable ties may be used instead of a plastic tape.
- 7) Place the lid on the Marinelli beaker, and seal the gap between the body and lid with a plastic tape (Photo 2-22).
- 8) Wipe the outside of the Marinelli beaker thoroughly with a paper towel moistened with pure water or ethanol.
- 9) To prevent the contamination of the measuring instrument, place the Marinelli beaker in a polyethylene bag, remove air, and seal the bag by tying it at the top. Use this as the measurement sample.

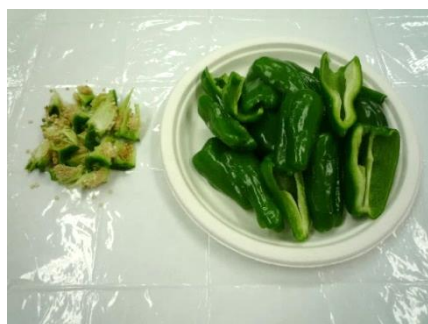


Photo 2-19 Example of the edible part of the sample (green bell pepper)



Photo 2-20 Example of the edible part of the sample (horse mackerel)



Photo 2-21 Sample packed inside
the Marinelli beaker
(Shiitake mushrooms)



Photo 2-22 Fixing the lid with
a plastic tape
(Milled rice)

[When using a small container]

- 1) For samples with a high moisture content, it is recommended to wrap a sealing tape around the threads of the small container to prevent leakage.
- 2) Write or attach a number or other data identifying the sample on the small container, and then measure and record the tare weight of the small container.
- 3) Using a laboratory spatula, pack the sample inside the small container without gaps, while ensuring homogeneity.
- 4) Lightly compress the sample surface, and level it horizontally (Photo 2-23 and Photo 2-24).
- 5) Cover the small container, and then weigh and record the height of the sample.
- 6) Wipe the outside of the container thoroughly with a paper towel moistened with pure water or ethanol.
- 7) Weigh the small container, and subtract the tare weight to obtain and record the weight of the measurement sample.
- 8) To prevent the contamination of the measuring instrument, cover the small container with a polyethylene bag, remove air, and seal the bag by tying it at the top to ensure that there are no wrinkles at the bottom of the container. Use this as the measurement sample.



Photo 2-23 Sample packed into a small container
(Asparagus)



Photo 2-24 Sample packed into a small container
(Butter)

[Storage of samples after measurement]

- A. Store in the same way as bovine milk.

2.2.5 Fallout (rainwater)

Fallout (rainwater) is an important sample for determining the amount of radioactive materials falling to the ground surface (MBq/km²). There is no mention of fallout (rainwater) in the Emergency Supplementary Reference Material. However, if an accident occurs in the country or overseas that poses the risk of the radioactive contamination of the environment and exposure to radiation and the Liaison Conference on Radiation Countermeasures or other such organizations instruct local governments and related organizations to strengthen monitoring, the temporary collection of the fixed-point samples of fallout (rainwater) shall be carried out. At that time, to grasp fluctuations in radiation levels in the environment and obtain information that helps decide on measures to be taken for public safety as well as assists in estimating the public radiation exposure dose, the continuous monitoring of ambient radiation dose rates will be strengthened and nuclide analysis of airborne dust will be conducted, along with the nuclide analysis of fallout (rainwater).

2.2.5.1 Collection of fallout (rainwater)

A precipitation sampling device (model 70A-H, water collection area of ~500 cm²) that is primarily used to collect precipitation samples for total beta analysis or a water collection funnel (diameter of ~25 cm) in combination with a water storage bottle (diameter of 20 cm and depth of ~10 cm) is used for fallout collection. If these are not available, a tray, a tub, or other container with a known opening area may be used instead.

[Fallout (rainwater) collection]

- 1) Place a collection container at the collection site, and record the collection start date and time as well as collection area of the container.

Note: The collection container shall be placed on a flat area unaffected by trees or buildings.

- 2) After the prescribed time has elapsed, transfer the entire sample into a polyethylene container.

Note: If no precipitation is observed, wash the inside of the collection container with pure water and collect as a dry fallout sample (Photo 2-25).

If there is snow on the collection container, snow shall also be collected along with water. If snow is collected or water is frozen, the total volume of the sample shall be measured after it has been transported indoors and completely melted at room temperature.

- 3) Record the end time of collection and other conditions.
- 4) Attach a label with an identification code onto the sample containing container (or clearly write the code on the container).
- 5) Seal the lid with a plastic tape, and place the sample in a polyethylene bag (inner bag). Further pack the sample in another bag (outer bag).



Photo 2-25 Example of collection in the absence of precipitation

2.2.5.2 Preparation of the fallout (rainwater) sample for measurements

For fallout (rainwater) collected using the precipitation sampling device, the measurement container and sample volume shall be selected according to the volume of the sample collected.

[Preparation of fallout (rainwater) measurement sample]

1) Transfer the sample to a measuring cylinder, and measure the collected sample volume.

Note: Because the liquid sample contains dust, etc., it should be thoroughly mixed and homogenized before transferring to the measuring cylinder.

2) Select an appropriate measurement container based on the collected volume.

3) Transfer the sample to the measurement container. If the sample volume is small, transfer the entire amount to a small container.

4) Prepare a measurement sample in a Marinelli beaker or a small container by referring to 2.2.2.2 Preparation of samples for drinking water measurements.

5) Calculate the fractionation ratio from the amount of sample taken and amount of test sample as follows.

The fractionation ratio is used in radioactivity calculation.

$$\text{Fractionation ratio(\%)} = \frac{\text{Amount of test sample}}{\text{Amount of sample taken}} \times 100$$

[Storage of samples after measurement]

Store the sample together with the portion that did not fit into the measuring container in the same manner as described in 2.2.2.2 Preparation of samples for drinking water measurements.

Chapter 3. Measurements Using a Germanium Semiconductor Detector

Radioactive iodine nuclides analyzed using a germanium semiconductor detector are I-131, I-132, and I-133. The half lives of these nuclides are generally short: 8.0 days for I-131, 2.3 h for I-132, and 21 h for I-133. Hence, the rule is to measure their half lives as soon as possible after sampling. However, during emergency monitoring, a large number and variety of samples may be generated; therefore, it is necessary to determine the order of priority and perform measurements.

This chapter describes the minimum measurement procedures and points to note when measuring radioactive iodine in an emergency. For details on the adjustment, calibration, and measurement procedures for a germanium semiconductor detector, refer to The Series of Environmental Radioactivity Measuring Methods No. 7 “Gamma-ray Spectrometry using a Germanium Semiconductor Detector (AI-Ge)”⁷⁾ and No. 29 “Gamma-ray Spectral Analysis Using a Germanium Detector in Emergencies (AI-Ge-E)”⁸⁾.

3.1 Adjustment and Calibration of the Instrument

The adjustment and calibration of the germanium semiconductor detector include energy and peak efficiency calibrations and peak to total (P/T) ratio calibration. There is no distinction between the normal and emergency calibration methods, but peak efficiency calibration shall be performed in advance for containers that are expected to be used in an emergency.

Containers used for measuring radioiodine in an emergency include small containers, Marinelli beakers, and activated carbon cartridges. To accurately perform peak efficiency calibration, it is necessary to use national standards and traceable sources that have the same shape as containers used and contain multiple radionuclides in known quantities. Because peak efficiency calibration is performed using this method for small containers and Marinelli beakers used in normal times, the peak efficiency calibration formula remains the same in an emergency when using these containers.

However, the above calibration formula cannot be used as such for activated carbon cartridges used to collect radioiodine in air. Efficiency calibration in this case is described below.

[Peak Efficiency Calibration for Activated Carbon Cartridges]

I) Method using a radioactive standard volume source

To obtain the accurate peak efficiency of an activated carbon cartridge, it is necessary to prepare an activated carbon cartridge containing known amounts of multiple radioactive nuclides using the activated carbon cartridge that will actually be used and perform peak efficiency calibration as a radioactive standard source. This method is the most appropriate method for peak efficiency calibration in γ -ray spectrometry. Because a sealed source comprising an activated carbon cartridge-shaped container filled with alumina containing known amounts of multiple nuclides can be obtained from a calibration laboratory, peak efficiency calibration can be performed using a radioactive standard volume source that satisfies the traceability of the national standard. Peak efficiency calibration is performed using this source in the same manner as that done for a small container and a Marinelli beaker.

If the source used for peak efficiency calibration is self-made by adding a known amount of unsealed radioactivity standard solution to activated carbon in an activated carbon cartridge actually used, it is desirable to ensure traceability by requesting the calibration service provider to measure the source. However, numerous difficulties are encountered when implementing this method because of the complicated work involved in purchasing and managing unsealed radioactivity standard solutions, handling them, and managing the prepared sources as well as the skill required of the operator to ensure quality.

If peak efficiency calibration cannot be performed using this method due to the unavailability of the radioactivity standard volume source in the form of an activated carbon cartridge, peak efficiency calibration shall be performed via alternative methods described in II) or III) below.

II) Method using an I-131 simulated source

This method uses an I-131 simulated source (Mock Iodine-131)*, which is similar in shape to an activated carbon cartridge, and as it is commercially available as a sealed source, it is easy to obtain and manage. However, due to the presence of a tin (Sn) filter at the bottom, peak efficiency is lower than that of a true activated carbon cartridge. Although the degree of self-absorption differs due to the difference in materials between the source and activated carbon cartridge, their shapes are similar, and the added radionuclide can provide peak efficiency, with energy close to that of I-131.

*I-131 simulated source

A sealed source containing Ba-133 and Cs-137 with long half lives was employed for the calibration of an NaI(Tl) scintillation detector. It exhibits a γ -ray spectrum that is almost similar to that of I-131 when measured with an NaI(Tl) scintillation detector. The radioactive material is adsorbed and fixed on ion-exchange resins and then sealed in a duralumin capsule. A 1-mm-thick tin (Sn) filter is inserted at the bottom to block the 81-keV γ -rays of Ba-133.

Figure 3-1 shows the γ -ray spectrum of the I-131 simulated source measured with a germanium semiconductor detector. Similar to the peak efficiency calibration of a small container, peak efficiency is calculated based on the net counting rate of each peak in the spectrum and radioactivity of the added nuclide listed in the certificate.

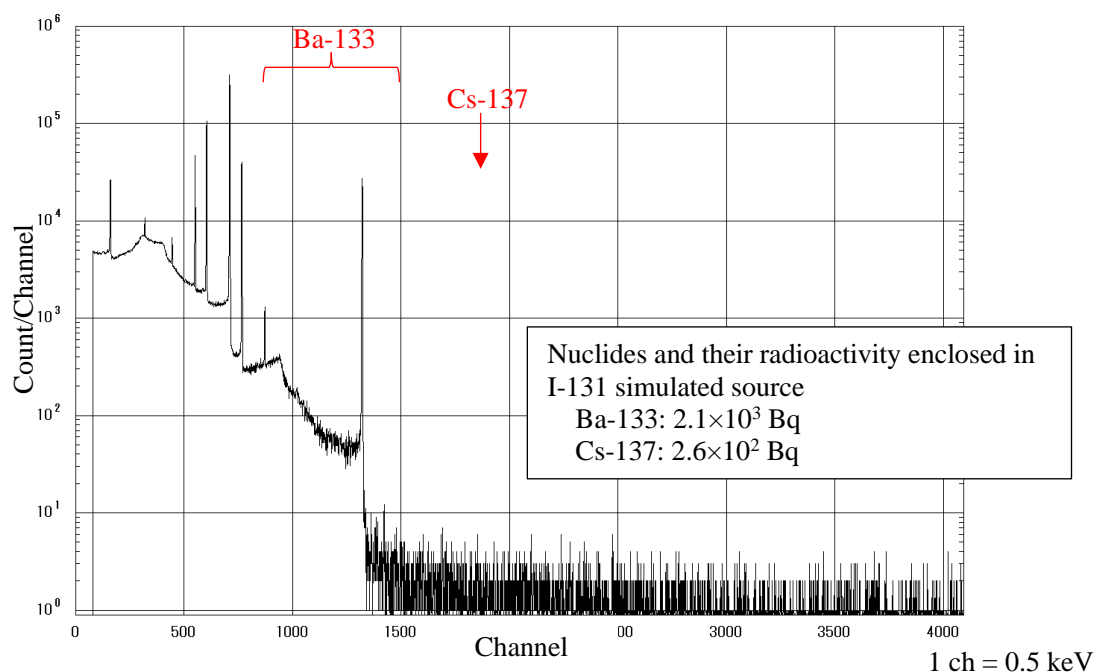


Figure 3-1 γ -ray spectra of an I-131 simulated source measured with a germanium semiconductor detector
(Relative efficiency of Ge: 28% and measurement time: 15,000 s)

Note that when calculating peak efficiency for the I-131 simulated source, the summing effect correction can be neglected because the 81-keV γ -rays of Ba-133, which cause the summing effect, are cut off. By drawing the obtained approximate curve of peak efficiency for each energy, the peak efficiency curve shown in Figure 3-2 can be obtained. For details, refer to The Series of Environmental Radioactivity Measuring Methods No. 7

“Gamma-ray Spectrometry using a Germanium Semiconductor Detector (AI-Ge)”⁷⁾.

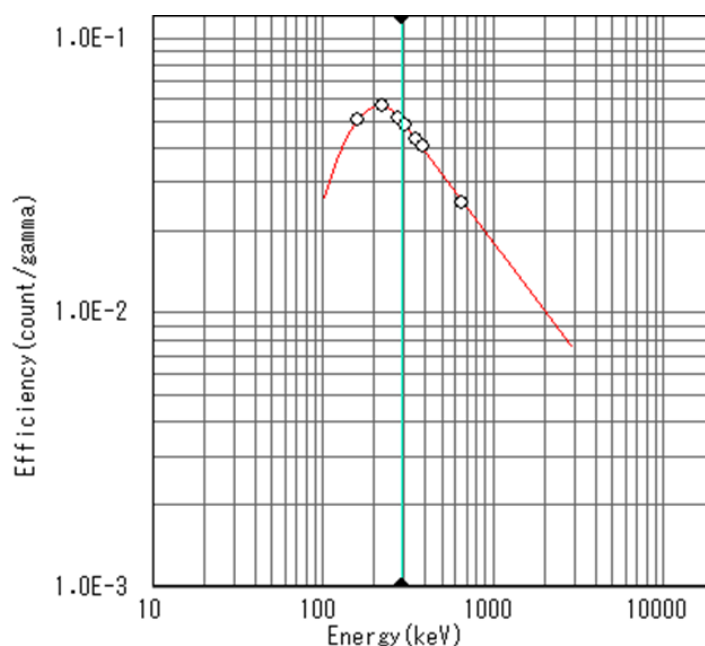


Figure 3-2 Peak efficiency curve obtained from an I-131 simulated source

III) Method using peak efficiency conversion

This method is used for calculating the peak efficiency of an activated carbon cartridge-shaped source through simulations based on the existing peak efficiency of a small container, etc. Because the existing efficiency value is used, efficiencies can be obtained without preparing a cartridge-shaped source. It shall be noted, however, that the validity of the peak efficiency conversion program to be used must be verified in advance. If settings are appropriate, the most accurate peak efficiency can be obtained. However, as it is necessary to master the calculation method and confirm its validity, it is a difficult method to be used to obtain peak efficiency. For details, refer to The Series of Environmental Radioactivity Measuring Methods No. 7 “Gamma-ray Spectrometry using a Germanium Semiconductor Detector (AI-Ge)”⁷⁾.

In an emergency, when none of the aforementioned methods, i.e., I) to III), can be used, the only method that can be used is utilizing the efficiency of small containers that are normally used as it is.

In this method, the thickness of the activated carbon cartridge is treated as the filling height of the sample, and the cartridge is analyzed as a small container. However, as the shapes of the standard radiation source and sample are considerably different, this method deviates from the principles of peak efficiency calibration for a germanium semiconductor detector. Therefore, if a significant value is detected, the obtained peak efficiency shall be reanalyzed at a later date using any of the methods of I) to III).

When using this method, it is advisable to use a container with a smaller peak efficiency than the activated carbon cartridge to avoid underestimation and to ensure safe-side evaluation. Usually, if the peak efficiency of a small container with features such as a raised bottom, a thicker bottom, or a larger diameter than the cartridge is used, then evaluation is to be performed on the safe side.

Comparative examples of peak efficiencies obtained with alternative methods described above are shown in Table 3-1.

Table 3-1 Simple calibration method for measuring the peak efficiency of an activated carbon cartridge and an example of the obtained efficiency

| Peak efficiency simple calibration method | Peak efficiency (%) | | |
|---|----------------------|----------------------|----------------------|
| | I-131 (364.5 keV) | I-133 (529.9 keV) | I-132 (667.7 keV) |
| Method with an I-131 simulated source | 4.17 | 3.10 | 2.59 |
| Method using peak efficiency conversion* | 4.98 | 3.57 | 2.90 |
| Methods using the efficiency*** of small containers*** | 4.55 | 3.28 | 2.68 |

* Conversions are performed in EFFTRAN based on a volumetric source in a small container** with a filling height of 30 mm.

** The small container has a diameter of ~50 mm, a height of ~50 mm, and a capacity of ~100 mL.

*** Calculated with a sample density of 1.096 g/cm³ and a filling height of 20 mm.

3.2 Nuclear Data

In an emergency, not only radioactive iodine, but also many artificial radionuclides are released into the environment; therefore, a nuclear data library is necessary for emergencies. Because the creation of the nuclear data library is a complicated process and there will not be enough time to create it in an emergency, it is necessary to create it in advance. For the creation of the nuclear data library for emergency, see The Series of Environmental Radioactivity Measuring Methods No. 29 “Gamma-ray Spectral Analysis Using Germanium Detector in Emergencies (AI-Ge-E)”⁸⁾.

Nuclear data (half life, γ -ray energy, and emission rate) used in γ -ray spectrometry obtained utilizing a germanium semiconductor detector can be obtained from nuclear data collections in databases or literature. Some of these collections have been published on the Internet in recent years and are listed below.

- Evaluated Nuclear Structure Data File (ENSDF)
<https://www.nndc.bnl.gov/ensdf/>
- Decay Data Evaluation Project (DDEP)
<http://www.lnhb.fr/nuclear-data/>
- Evaluated Nuclear Data File (ENDF)
<https://www-nds.iaea.org/exfor/endl.htm>
- Joint Evaluated Fission and Fusion (JEFF) Library
<http://www.oecd-nea.org/dbdata/>

Nuclear data may have different values in various nuclear data collections, and even in the same nuclear data collection, values may differ due to data updates. This is because methods used for evaluating nuclear data and the data referenced by nuclear data collections are different. As a result, it is necessary to verify nuclear data because the calculated results will vary depending on the nuclear data used.

In γ -ray spectrometry, analysis is performed as follows:

- (1) γ -ray intensity (γ/s) is obtained from the standard source test value (Bq) using the nuclear data, and peak efficiency is acquired from the peak counting rate (cps).
- (2) γ -ray intensity (γ/s) is obtained from the peak counting rate (cps) of the sample measurement results using peak efficiency, and radioactivity (Bq) is acquired using the nuclear data.

Nuclear data are used to convert radioactivity to γ -ray intensity (or vice versa). Therefore, the nuclear data used in (1) and (2) shall be taken from the same nuclear data collection. If they are taken from the same nuclear data collection, there is no need to be particular about the latest data as no differences will occur during conversion. Additionally, note that when updating nuclear data for analysis from these perspectives, peak efficiencies must be recalculated.

The nuclear data for radioiodine are shown in Table 3-2. The nuclear data in Table 3-2 are listed according to the following:

- 1) Source of nuclear data is ENSDF (as of September 2022).
- 2) Values are assigned for items with an emission rate of 1% or more.
- 3) For the unit of half life, D indicates day and H denotes hour.

Table 3-2 Nuclear data of radioiodine

| Nuclide name | Half life | Standard uncertainty of half life* | Half life unit | γ -ray energy (keV) | Standard uncertainty of γ -ray energy* (keV) | Emission rate (%) | Standard uncertainty of emission rate* (%)** | Main formation reaction |
|------------------|-----------|------------------------------------|----------------|----------------------------|---|-------------------|--|---|
| ^{131}I | 8.0252 | 0.0006 | D | 80.1850 | 0.0020 | 2.62 | 0.04 | Fission $^{132}\text{Xe}(\text{n}, \text{p})$ |
| | | | | 284.305 | 0.005 | 6.12 | 0.07 | |
| | | | | 364.489 | 0.005 | 81.5 | 0.8 | |
| | | | | 636.989 | 0.004 | 7.16 | 0.10 | |
| | | | | 722.911 | 0.005 | 1.77 | 0.03 | |
| ^{132}I | 2.295 | 0.013 | H | 262.90 | 0.10 | 1.28 | 0.10 | $^{132}\text{Te} \rightarrow$ Fission $^{132}\text{Xe}(\text{n}, \text{p})$ |
| | | | | 505.79 | 0.03 | 4.94 | 0.20 | |
| | | | | 522.65 | 0.09 | 16.0 | 0.5 | |
| | | | | 547.20 | 0.20 | 1.14 | 0.08 | |
| | | | | 621.2 | 0.3 | 1.58 | 0.20 | |
| | | | | 630.190 | 0.020 | 13.3 | 0.04 | |
| | | | | 650.50 | 0.20 | 2.57 | 0.20 | |
| | | | | 667.7141 | 0.0020 | 98.70 | | |
| | | | | 669.80 | 0.20 | 4.6 | 0.6 | |
| | | | | 671.40 | 0.20 | 3.5 | 1.0 | |
| | | | | 727.0 | 0.3 | 2.2 | 0.6 | |
| | | | | 727.2 | 0.3 | 3.2 | 0.6 | |
| | | | | 728.40 | 0.20 | 1.6 | 0.4 | |
| | | | | 772.600 | 0.010 | 75.6 | 1.3 | |
| | | | | 780.00 | 0.20 | 1.18 | 0.04 | |
| | | | | 809.50 | 0.20 | 2.6 | 0.3 | |
| | | | | 812.00 | 0.20 | 5.5 | 0.4 | |
| | | | | 876.60 | 0.20 | 1.04 | 0.04 | |
| | | | | 954.55 | 0.09 | 17.6 | 0.5 | |
| | | | | 1136.000 | 0.020 | 3.01 | 0.14 | |
| | | | | 1143.30 | 0.20 | 1.35 | 0.06 | |
| | | | | 1172.90 | 0.20 | 1.09 | 0.07 | |
| | | | | 1290.80 | 0.20 | 1.13 | 0.05 | |
| | | | | 1295.10 | 0.20 | 1.88 | 0.07 | |
| | | | | 1372.07 | 0.13 | 2.47 | 0.10 | |
| | | | | 1398.57 | 0.10 | 7.01 | 0.20 | |
| | | | | 1442.56 | 0.10 | 1.40 | 0.05 | |
| | | | | 1921.08 | 0.12 | 1.23 | 0.06 | |
| ^{133}I | 20.83 | 0.08 | H | 510.530 | 0.004 | 1.83 | 0.06 | Fission |
| | | | | 529.872 | 0.003 | 87.0 | 2.3 | |
| | | | | 706.578 | 0.008 | 1.51 | 0.05 | |
| | | | | 856.278 | 0.007 | 1.24 | 0.04 | |
| | | | | 875.329 | 0.005 | 4.51 | 0.13 | |
| | | | | 1236.441 | 0.006 | 1.51 | 0.05 | |
| | | | | 1298.223 | 0.005 | 2.35 | 0.07 | |

* Standard uncertainty refers to the uncertainty of the inclusion factor $k = 1$.

** Taking the emission rate of ^{131}I at 80.1850 keV as an example, an emission rate of 2.62% with the standard uncertainty of the emission rate of 0.04% gives the emission rate of 2.62 ± 0.04 (%).

3.3 Measurement and Analysis

The measurement sample prepared in Chapter 2 is analyzed with the germanium semiconductor detector. In an emergency, unlike normal times, pay attention to the contamination of the detector and increased dead time during measurements and to the complex γ -ray spectra and misidentification of nuclides during analysis.

In the initial phase of an accident (when multiple nuclides are detected), the measurement time shall be at least 10 min. If the sample has a large dead time (more than 10%), the dead time shall be reduced by decreasing the sample volume or measuring the sample away from the detector. However, if the measurement sample is moved away from the detector, peak efficiency shall be obtained at that position beforehand.

[Measurement time and quantifiable level]

The measurement time and quantifiable levels are shown in Tables 3-3 and 3-4. However, these values are merely guidelines and may vary significantly depending on the type and concentration of radionuclides in measurement samples. Note that I-131, I-132, and I-133 have short half lives ranging from few hours to eight days. Therefore, the time elapsed between sample collection and measurement should be noted.

The quantifiable levels shown in Tables 3-3 and 3-4 are excerpts taken from The Series of Environmental Radioactivity Measuring Methods No. 24 “Preparation of Samples for Gamma-ray Spectrometry in Emergencies (AP-E).”⁶⁾ To calculate quantifiable levels, the spectra of environmental samples taken after the 1F accident using Marinelli beakers or small containers are utilized to obtain the detection limit at each measurement time. The relative efficiency of the germanium semiconductor detector is assumed to be a commonly used value of ~30%.

For food and drink, the OIL6 standard (Table 3-5), which serves as a guideline for determining intake limits, has been established, and the measurement conditions shall be determined so that quantifiable levels are lower than this standard. If drinking water or bovine milk is measured in a small container for 10 min, its quantifiable level is 350 Bq/kg, and it is not possible to determine whether the OIL6 standard value (300 Bq/kg) has been exceeded. Therefore, the sample volume or measurement time must be increased. However, as quantifiable levels shown in Tables 3-3 and 3-4 contain many radioactive nuclides whose spectra on which calculations are based, detection limits in actual samples may be lower than these. Details regarding the measurement time and quantifiable level are provided in Information A-1.

Table 3-3 Relationship between the measurement time and quantifiable level when using a Marinelli beaker (2 L)

| Sample name | Test quantity | I-131 quantifiable level | | | | Unit |
|-------------------------------|---------------|--------------------------|--------|------|-------|-------|
| | | Measurement Time | | | | |
| | | 10 min | 30 min | 1 hr | 10 hr | |
| Fallout Precipitation | 2000 g | 110 | 70 | 50 | 20 | Bq/kg |
| Drinking water Bovine milk | 2000 g | 110 | 70 | 50 | 20 | |
| Soil | 3100 g | 80 | 50 | 30 | 10 | |
| Vegetables | 1000 g | 200 | 120 | 80 | 30 | |
| Meat Egg Seafood | 1900 g | 120 | 70 | 50 | 20 | |

Table 3-4 Relationship between the measurement time and quantifiable level when using a small container (50 mmφ × 50 mm)

| Sample name | Test quantity | I-131 quantifiable level | | | | Unit |
|-------------------------------|---------------------|--------------------------|--------|-------|--------|-------------------|
| | | Measurement Time | | | | |
| | | 10 min | 30 min | 1 hr | 10 hr | |
| Air | 1 m ³ | 6 | 4 | 3 | 0.8 | Bq/m ³ |
| | 10 m ³ | 0.6 | 0.4 | 0.3 | 0.08 | |
| | 1000 m ³ | 0.006 | 0.004 | 0.003 | 0.0008 | |
| Fallout Precipitation | 89 g | 350 | 200 | 150 | 50 | Bq/kg |
| Drinking water Bovine milk | 89 g | 350 | 200 | 150 | 50 | |
| Soil | 140 g | 240 | 140 | 100 | 30 | |
| Vegetables | 47 g | 610 | 350 | 250 | 80 | |
| Meat Egg Seafood | 86 g | 360 | 210 | 150 | 50 | |

Table 3-5 OIL6 (Default setting values)

| Nuclide | Drinking water, bovine milk, dairy products | Vegetables, grains, meat, egg, fish, etc. |
|-------------|---|---|
| Radioiodine | 300 Bq/kg | 2,000 Bq/kg |

Source: Guideline for Emergency Preparedness and Response¹⁾

3.3.1 Sample measurements

In an emergency, measurement using a germanium semiconductor detector is performed according to the same procedures as employed during normal times, but care must be taken to avoid contamination inside the detector and shielding.

The measurement procedure is shown as a flow diagram in Figure 3-3. For details, see The Series of Environmental Radioactivity Measuring Methods No. 29 “Gamma-ray Spectral Analysis Using Germanium Detector in Emergencies (AI-Ge-E)”⁸⁾.

[Measurement of air samples]

When air samples are collected using an iodine sampler with an automatic sample changer, a glass fiber filter paper (particulate iodine) and activated carbon cartridges (gaseous iodine) are used as measurement samples. The retrieved glass fiber filter paper and activated carbon cartridges are measured separately. The radioactivity concentrations of particulate iodine and gaseous iodine are determined, and then, the two concentration values are added together to calculate the total radioactive iodine concentration for each nuclide. The composition ratio of the particulate and gaseous forms can be obtained by conducting separate measurements. This ratio is used to calculate the concentration of gaseous radioiodine from the measurement results collected only using the filter paper employed in the air monitoring post, and the total radioiodine concentration for each nuclide can be estimated.

In an emergency, it is not necessary to measure all samples separately as the machine time may become tight due to an increase in the number of samples to be measured. However, keep in mind that samples measured separately should be determined so that the ratio of particulate to gaseous matter can be continuously monitored.

When measuring air samples, the glass fiber filter paper is packed into small containers and activated carbon cartridges are used as the measurement sample as such, according to the method described in 2.2.1.4. The activated carbon cartridge is measured with the suction side (detector side) facing down. Because the distribution of radioactive iodine adsorbed by activated carbon inside the cartridge is not uniform, it will be more accurate to measure both sides of the activated carbon cartridge and average the results. However, measurements on both sides are not performed because the process is more complicated to perform in an emergency, and because the measurement results are larger when the suction side is on the detector side, resulting in a safer side evaluation. The details regarding the measurement surface and measurement values of the cartridge are provided in Information A-2.

[Measurement of other samples]

For samples other than air, all measurements are performed in Marinelli beakers or small containers. Therefore, the normal measurement procedures are employed. For soil samples, it is important to record the area and depth of sampling so that the relationship between them and measurement results can be evaluated later.

Measurement

- ← **(1) Confirm the identification of the sample to be measured**
 - Check the sample container against the collection record sheet, ledger, etc. to confirm that there is no mistake.
 - To prevent sample mix-ups, verify with the person in charge, as necessary.
- ← **(2) Place the measurement sample on the detector end cap**
 - Verify that the geometry is identical to that of the standard source for efficiency calibration.
 - Be careful not to contaminate the detector.
- ← **(3) Erase the previously measured γ -ray spectrum (after confirming that it has been saved)**
- ← **(4) Set the measurement time (preset time) (10 min to 1 h is the standard)**
- ← **(5) Start the measurement**
 - Record the start time of measurement.
 - Confirm the measurement start time and actual time on the multichannel analyzer (or PC).
 - Confirm that the measurement time (live time) is increasing.
 - Check the dead time.
 - If it is more than 10%, take measures such as reducing the sample amount, as necessary.
 - Confirm that no drift of the focus peak has occurred.
 - Recheck again at time intervals of about every 10–30 min.
 - Verify that there are no broad peaks in the low-energy region.
 - If peaks corresponding to artificial radionuclides are observed within a short period of time, alert the sample preparation personnel regarding the possibility of cross contamination.
- ← **(6) Record the measurement information**
- ← **(7) End of measurement**
 - Stop measurement (if preset time is not set).
 - Confirm that measurement has been completed (if preset time is set).
 - Record the live time and measurement end time, as needed.
 - Save the measured γ -ray spectra.
- ← **(8) Take out the sample to be measured**
 - Check the geometry (positional relationship between the sample to be measured and detector).
 - Check the taken sample against records.
- ← **(9) Storage of measured samples**
 - Store in a designated location.
 - If possible, samples exhibiting high radioactivity shall be stored separately.
 - Check the detector for contamination, as necessary.
 - After measuring a sample with high radioactivity, perform a background measurement to check for contamination.

Analysis

Figure 3-3 Sample measurement procedure flow

(Adapted from The Series of Environmental Radioactivity Measuring Methods No. 29
“Gamma-ray Spectral Analysis Using Germanium Detector in Emergencies (AI-Ge-E)”⁸⁾)

3.3.2 Spectrum analysis

Recent spectrum analysis using a commercial software allows users to perform simple operations such as peak search, nuclide identification, quantification, and the output of analysis reports for measured γ -ray spectra.

The commercial software can be used during normal times and in emergency, but in γ -ray spectra taken in an emergency, γ -ray peaks derived from a large number of artificial radionuclides are observed. It shall be noted that some peaks may be treated as unknown peaks because they do not match with the nuclear data library for

the analysis used, and γ -ray peaks and sum peaks with small emission rates may also be observed. Additionally, analytical detections may occur due to statistical fluctuations of the baseline.

In general, in γ -ray spectrum analysis during an emergency, it is important to visually check the γ -ray spectrum, rather than simply reporting the output report automatically generated using the commercial analysis software.

The analysis procedure is shown as a flow diagram in Figure 3-4. For details, see The Series of Environmental Radioactivity Measuring Methods No. 29 “Gamma-ray Spectral Analysis Using Germanium Detector in Emergencies (AI-Ge-E)”⁸⁾.

[Radioiodine analysis]

The I-131, I-132 and I-133 peaks are identified and quantified from the γ -ray spectrum. The peak used for quantification is basically the peak corresponding to the highest emission rate. Table 3-6 lists the main quantitative peaks for each nuclide.

Table 3-6 Main quantitative peaks of radioactive iodine

| Nuclide | Half life | Energy (keV) | Emission rate (%) |
|---------|-------------|--------------|-------------------|
| I-131 | 8.0252 days | 364.489 | 81.5 |
| I-132 | 2.295 h | 667.7141 | 98.70 |
| | | 772.600 | 75.6 |
| I-133 | 20.83 h | 529.872 | 87.0 |

The quantitative peak of I-131 has a lower energy than other artificial radionuclides. Care must be taken if there is a large amount of Cs-137 (661.7 keV) or I-132, which are on the higher-energy side than I-131, because the baseline of the γ -ray spectrum will be high due to the effect of the Compton scattering of γ -rays emitted by these radionuclides, and I-131 may not be detected.

γ -ray spectrum

← **(1) Registration of measurement sample information**

- Register sample name, sample type, sampling location, sampling start and end dates and times, measurement container, sample volume (including units), sample height, sample density, material of the measurement sample, name of the person in charge of measurement, etc.

← **(2) Registration of various calibration files**

- Register the energy calibration file (including the relationship between the half width and γ -ray energy).
- Register the relative efficiency or P/T ratio file and cascade file.
- Register the efficiency calibration file.

← **(3) Setting of analysis conditions**

- Select a nuclear data library for analysis.
- Select peak search sensitivity (usually has a value of 3).
- Select the decay correction conditions.
Select one of the following: the start or end date and time of sampling, the middle of sampling period, and no decay correction.
- Select the method to be used for calculating the peak area.
Choose from the Covell method or function fitting.
- Register background correction files.
Select the background spectrum for correction.

← **(4) Execution of analysis**

← **(5) Output of analysis results as a report**

- Output peak search results, peak quantification results, γ -ray spectral plots, etc.

← **(6) Confirmation of analysis results**

- Check the information in the analysis result form.
- Confirm that the peak of interest (K-40, Cs-137, etc.) in the γ -ray spectrum has not drifted.
- Check the detected artificial radionuclides.
Magnify the γ -ray spectrum to confirm the shape of the γ -ray peak.
(Check to see if the statistical variation in the baseline counts affects detection.)
- Check for γ -ray peaks with small emission rates to improve the accuracy of identification.
- In the case of nuclides that decay sequentially, check for the γ -ray peaks of parental and descendant nuclides.
- Check for the γ -ray peaks of other radioisotopes (e.g., radioiodine).
- Identify unknown peaks by referring to the nuclear data sorted based on energy.
If necessary, register the nuclear data of identified nuclides in the nuclear data library for analysis, and repeat the analysis.

Evaluation

Figure 3-4 Analysis procedure for measuring a γ -ray spectrum
(Adapted from The Series of Environmental Radioactivity Measuring Methods No. 29 “Gamma-ray Spectral Analysis Using Germanium Detector in Emergencies (AI-Ge-E)”⁸⁾)

For I-132, as shown in 3.2 Nuclear data, as it is a nuclide that emits multiple γ -rays and produces a sum peak, pay attention to the sum effect correction and unknown peaks derived from the sum effect. Further details regarding the I-132 sum peak are provided in Information A-3.

I-132 has a half life of 2.3 h, the shortest of the three nuclides listed in Table 3-6. However, it is also a descendant of Te-132 (half life of 3.2 days), which is produced as a fission product, and the two are in a transient equilibrium (Figure 3-5 and Figure 3-6). Therefore, when radioactive materials are released into the environment, they exist for a longer period of time than I-133, which has a half life of 21 h. Furthermore, I-132 is a nuclide in a transient equilibrium, and the radioactivity ratio at the time of measurement is not necessarily that in the transient equilibrium state. Therefore, simply applying decay correction may result in overestimation or underestimation⁸⁾. Therefore, the basic rule is to report the radioactivity concentration at the time of measurement without decay correction. If the correction is necessary, a decay-corrected value shall be reported. In either case, the reported values shall include information on whether or not decay correction was performed, and if so, the reason and method utilized for decay correction (such as the measurement time and correction destination time) are provided.

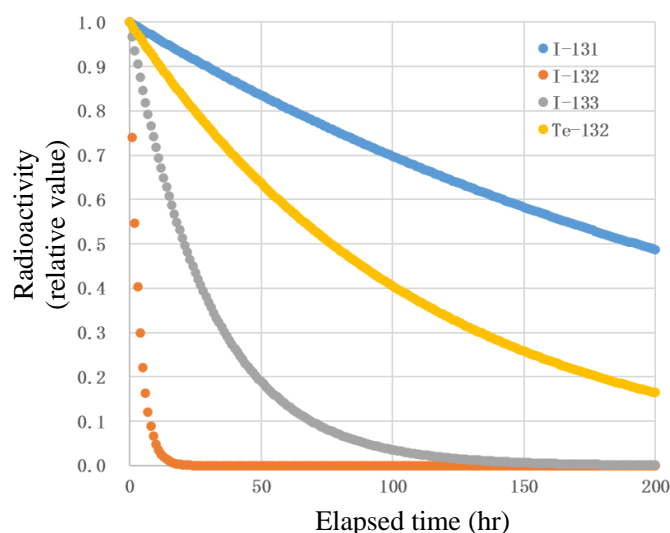


Figure 3-5 Change in radioactivity over time when each radionuclide decays according to its half life

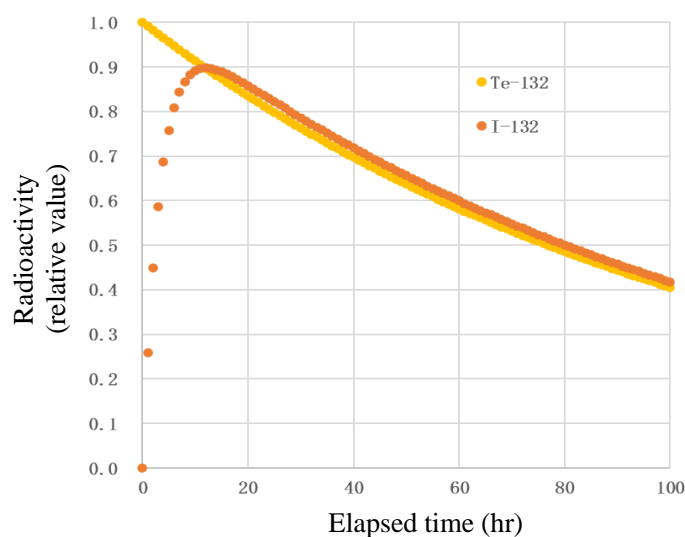


Figure 3-6 Change in the radio activities of Te-132 and I-132 over time (transient equilibrium)

3.4 Data Evaluation

[Air samples]

Air samples are collected using an iodine sampler with an automatic sample changer, and particulate iodine is acquired on a glass fiber filter, while gaseous iodine is collected in an activated carbon cartridge. The collected glass fiber filter and activated carbon cartridge are then measured with the germanium semiconductor detector to obtain the concentrations of particulate and gaseous radioactive iodine. By adding up the two concentration values, the total radioactive iodine concentration for each nuclide is estimated and used to assess the radiation exposure dose.

Furthermore, air monitoring posts only use a filter paper to collect air; hence, although it can capture particulate iodine, it cannot capture gaseous iodine. Therefore, at sites where only air monitoring posts are installed, the total radioactive iodine concentration for each nuclide is calculated using the following procedure and used for exposure assessment.

- 1) Calculate the ratio of particulate radioiodine to gaseous radioiodine from the measurement results of samples collected using an iodine sampler with an auto sample changer via the germanium semiconductor detector.
- 2) Measure the particulate radioiodine concentration by analyzing the filter paper retrieved from the air monitoring post using a germanium semiconductor detector.
- 3) Calculate the total radioiodine concentration for each nuclide from the ratio of particulate radioiodine to gaseous radioiodine estimated in 1) and concentration of particulate radioiodine measured in 2).

The ratio of particulate radioiodine to gaseous radioiodine in the atmosphere may not be constant over time and space. Therefore, when calculating gaseous radioiodine from the air monitoring post results using the obtained ratio, it is necessary to match as closely as possible the time and location of the air monitoring post with those of the iodine sampler with an auto sample changer in which the sample was collected. Use reading from an air monitoring post at a nearby location and that matches the sampling times of the iodine sampler with an auto sample changer on which the ratio is based.

[Drinking water and food samples]

In areas where the air radiation dose rate exceeds 0.5 $\mu\text{Sv/h}$, radionuclide concentrations in food and drink produced in the area are measured to determine the protective measures to be implemented in accordance with OIL6. OIL6 is the standard for measuring radionuclide concentrations in food and drink within a week and for promptly enforcing intake restrictions on products exceeding the standard value.

Because the intake of food and drink is restricted if products exceed the OIL6 standard value, when these products are measured using a germanium semiconductor detector, the results must also be compared with the OIL6 standard.

[Soil samples]

Soil analysis results are evaluated not only in terms of the radioactivity concentration per weight (Bq/kg) but also in terms of the deposited amount per unit area (Bq/m^2) for the purpose of understanding the extent of ground surface contamination by radioactive materials deposited on the ground. Therefore, when soil samples are collected, the area and depth of sampling should be recorded.

Using radioactivity per unit weight (Bq/kg) obtained from the measured sample weight, the deposited amount per unit area (Bq/m^2) is calculated using the following formula.

Because evaluating uncertainty in the initial monitoring during an emergency is a complex task, the counting error calculated in conjunction with the calculation of the radioactivity concentration is described here as standard deviation.

$$A_S \pm \Delta A_S = (A_W \pm \Delta A_W) \times W \times \frac{1}{S} \times 10^4$$

A_S : Deposited amount per m² (Bq/m²)

ΔA_S : Standard deviation based on the A_S count

A_W : Radioactivity concentration per kilogram of sample (Bq/kg)

ΔA_W : Standard deviation based on the A_W count

W : Weight of the sample collected (kg)

S : Collection area (cm²)

Information

Information A Study on γ -ray Spectrometry Using a Germanium Semiconductor Detector

This section provides reference data for the measurement and analysis of radioactive iodine via γ -ray spectrometry using a germanium semiconductor detector.

A-1 Measurement Time and Lower Detection Limit

The quantifiable levels of I-131 in this method are shown in Tables 3-3 and 3-4, which are partially adopted from The Series of Environmental Radioactivity Measuring Methods No. 24 “Preparation of Samples for Gamma-ray Spectrometry in Emergencies (AP-E)”⁶⁾. I-131 quantifiable levels for some sample types are presented in Figure A-1-1.

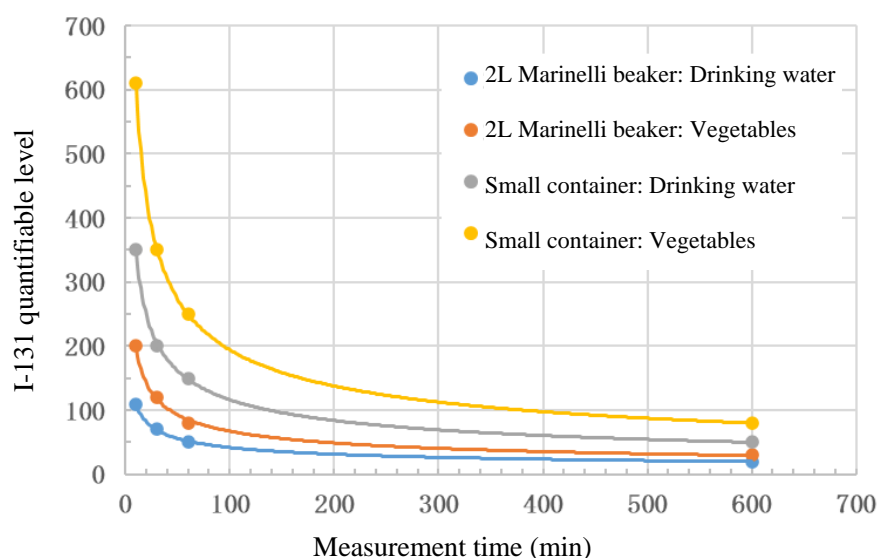


Figure A-1-1 Possible I-131 quantification levels in drinking water and vegetables (from Table 3-3 and Table 3-4)

These values are based on spectra measured using a germanium semiconductor detector with a relative efficiency of ~30% utilizing environmental samples collected after the 1F accident in a Marinelli beaker (2 L) or a small container (100 mL: 50 mm ϕ \times 50 mm). The lower detection limit calculated for various measurement times and for each sample type is used as the quantifiable level.

Examples of spectra used to calculate quantifiable levels are shown in Figures A-1-2 and A-1-3. Quantifiable levels are calculated based on the results of these analyses. The lower detection limit is calculated according to the method described in The Series of Environmental Radioactivity Measuring Methods No. 7 “Gamma-ray Spectrometry using a Germanium Semiconductor Detector (AI-Ge).”⁷⁾

The measured samples for spectral analysis used in calculations contain a large number and amounts of radionuclides, and the baseline of the spectra is high. Therefore, the quantifiable level is also high. However, the radioactivity concentration of the measured samples is not necessarily this high; a study based on simulated samples of I-131 and Cs-137 (Ge relative efficiency: 30%) reported a lower detection limit (Table A-1-1).

Table A-1-1 Results of the study of I-131 quantifiable levels using simulated samples

| Sample name | Test quantity | Container | Quantifiable level |
|-----------------|---------------------|-----------------|--------------------------|
| | | | Measurement time 10 min. |
| Air | 0.25 m ³ | Small container | 6 Bq/m ³ |
| Drinking water | 2 L | 2 L Marinelli | 2 Bq/L |
| Bovine milk | 2 L | 2 L Marinelli | 2 Bq/L |
| Green vegetable | 0.5 kg | 0.7 L Marinelli | 6 Bq/kg |

The present calculations of quantifiable levels for other samples are obtained by adopting the spectra of environmental samples collected after the 1F accident. The results shown here are only a guide. This is because the results will vary depending on the types and concentrations of radionuclides contained in measured samples due to the conditions of contamination at the time of emergency.

For reference, the measurement results of samples with lower radionuclide concentrations than the reference spectrum are shown in Figures A-1-4 and A-1-5.

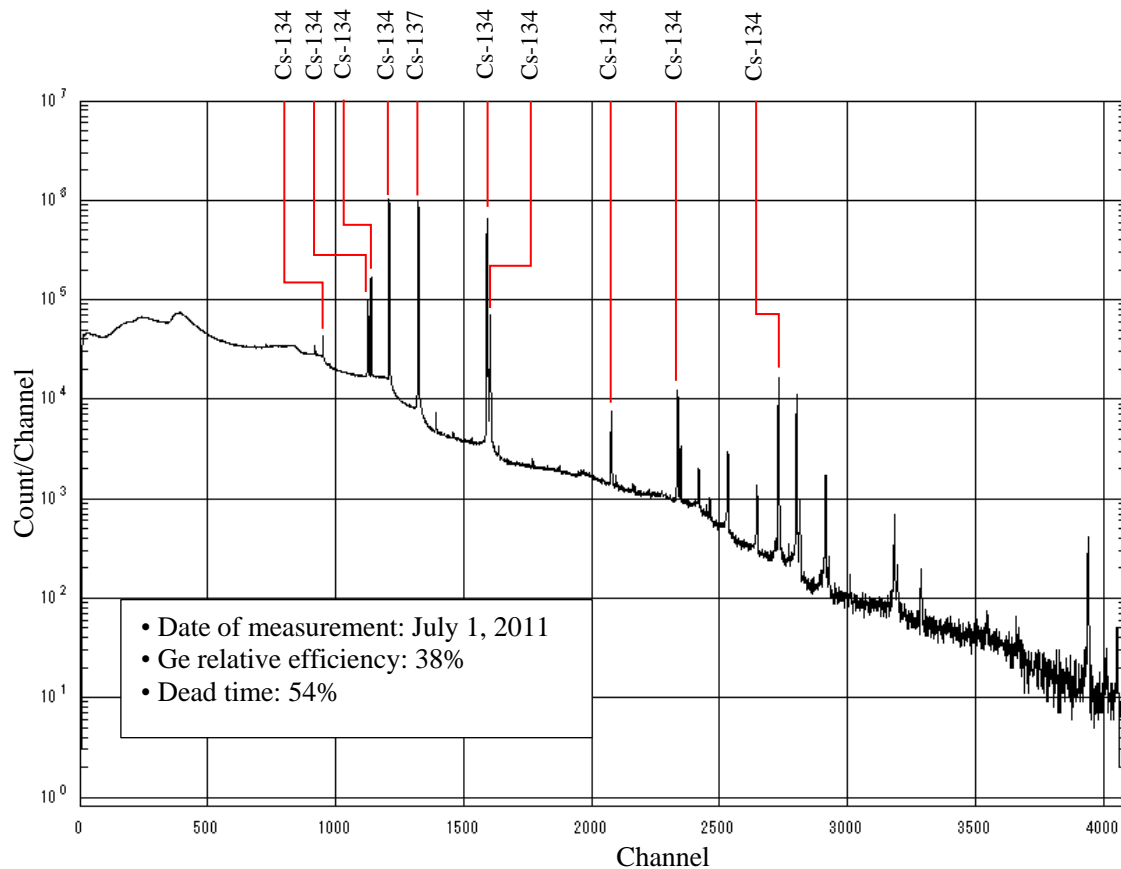


Figure A-1-2 γ -ray spectra used to calculate quantifiable levels for a 2-L Marinelli beaker (fallout)

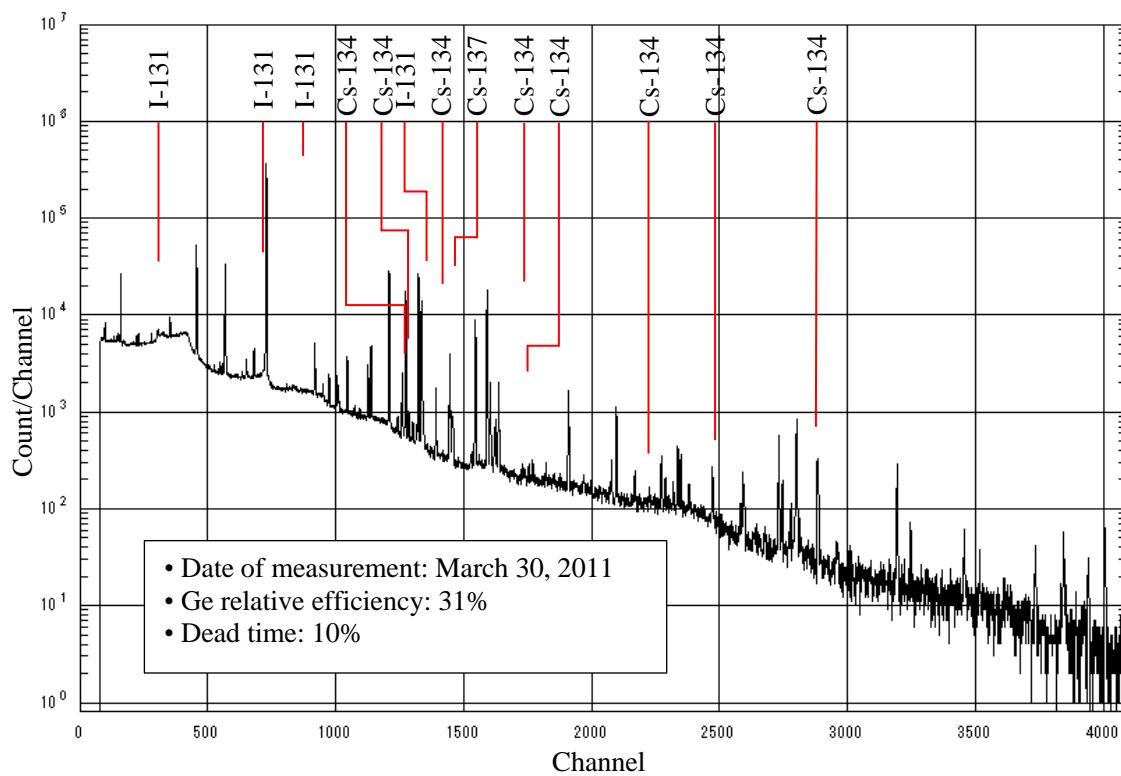


Figure A-1-3 γ -ray spectra used to calculate quantifiable levels for a small container (soil)

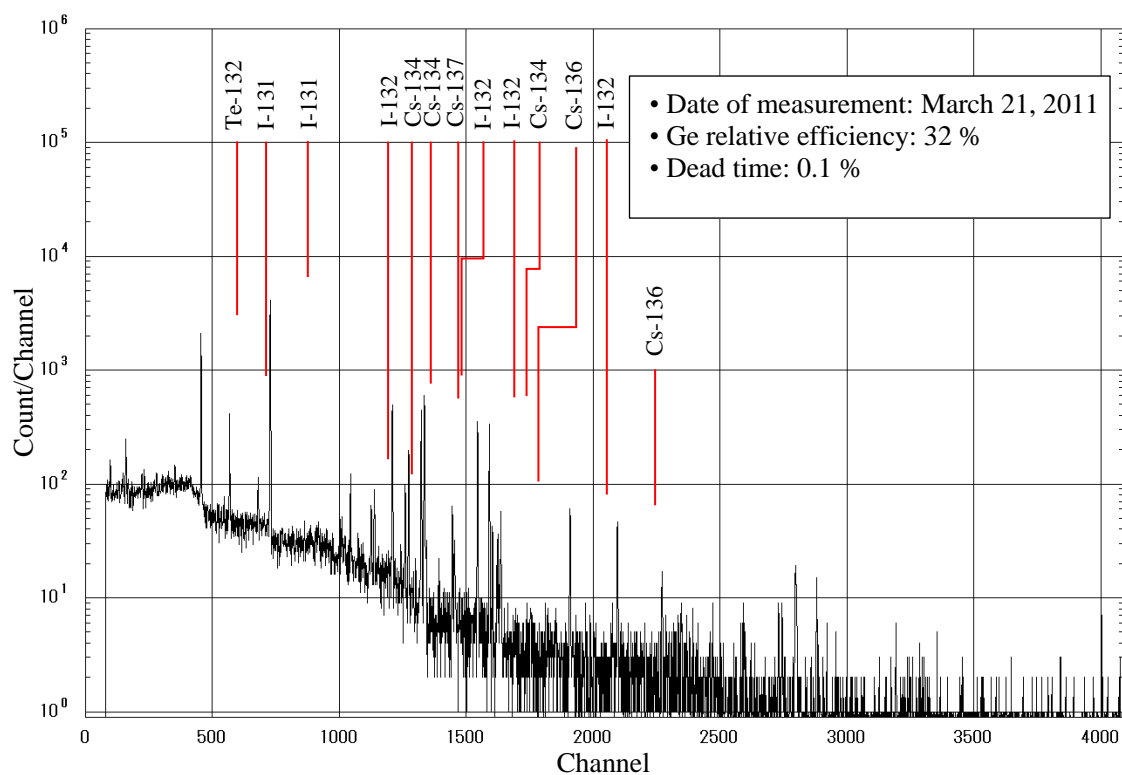


Figure A-1-4 γ -ray spectra of leafy vegetables (small container)
(Sample weight: ~40 g, measurement time: 60 min, and I-131 lower detection limit: 30 Bq/kg)

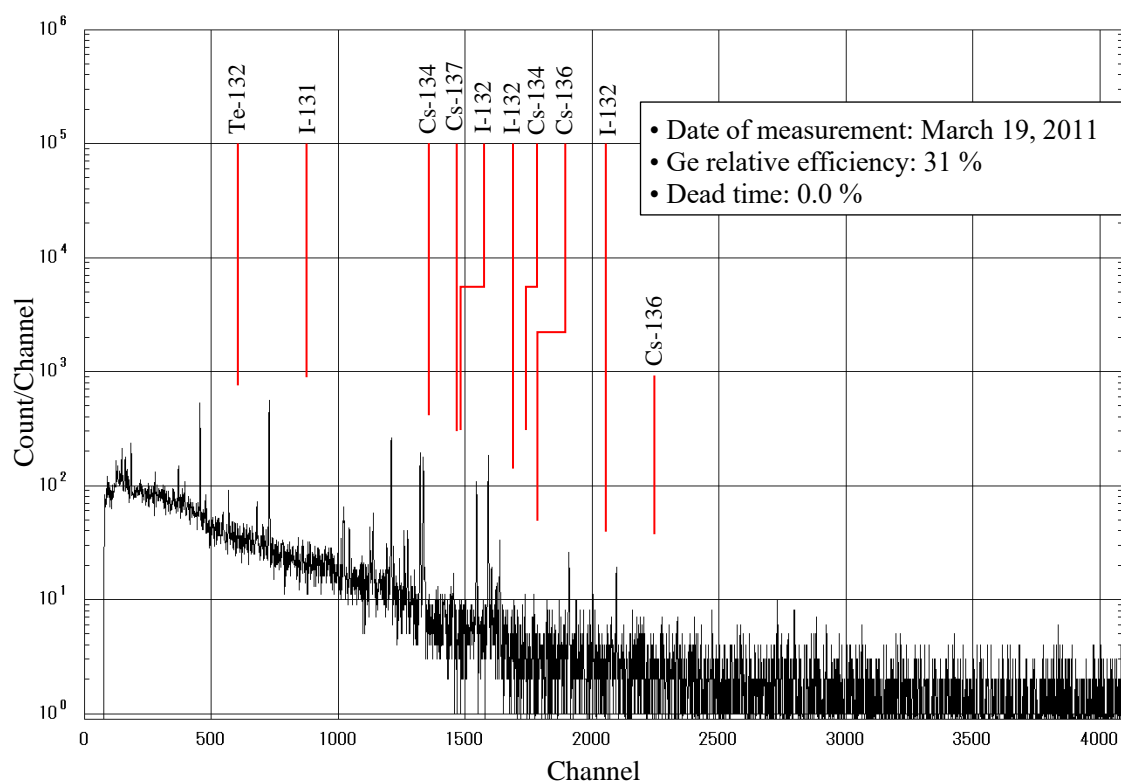


Figure A-1-5 γ -ray spectra of tap water (2-L Marinelli beaker)
(Sample volume: 2 L, measurement time: 330 min, and I-131 lower detection limit: 0.2 Bq/L)

A-2 Suction Position and Counting Efficiency of an Activated Carbon Cartridge

When gaseous iodine is collected using an activated carbon cartridge, iodine is adsorbed from the activated carbon layer on the suction surface side (upstream side); thus, it is distributed in the depth direction. The thickness of the layer where iodine is adsorbed and the corresponding iodine concentration varies with the suction volume and flow velocity. Therefore, the distribution of iodine collected in the activated carbon cartridge is not uniform and varies depending on the collection conditions. In normal times, when quantifying such samples using a germanium semiconductor detector, activated carbon is transferred to a separate container, stirred, and homogenized before measurement. However, in emergency, this method is labor-intensive and carries a high risk of contaminating the surrounding environment. Therefore, methods that can be used for directly measuring activated carbon cartridges and the bias of quantitative results are explored.

Assuming an activated carbon cartridge (inner diameter of 52 mm and thickness of 18 mm) and a small container (inner diameter of 48 mm), counting efficiency is calculated for each sample thickness (corresponding to the adsorption layer thickness) varying from 1 to 18 mm and for the adsorption layer at 18 mm (when iodine is adsorbed from the top of the container). The relative efficiency of a typical germanium semiconductor detector is 27%, which is set as the counting efficiency of I-131 at 364 keV. The results are shown in Figure A-2-1.

These results show that peak efficiencies when the sample thicknesses are varied from 1 to 18 mm are 5.6% to 3.8%, a difference of approximately 1.5 times, when the suction surface faces the detector. Even when the suction surface is facing away from the detector, a difference of approximately 1.5 times is observed, with the efficiency ranging from 2.5% to 3.8%. When the average of the two is taken, the results show a difference of only about 1.1 times, with the efficiency ranging from 4.1% to 3.8%. This is comparable to the results obtained when activated carbon is once removed from the iodine-adsorbed activated carbon cartridge, homogenized, and refilled into the cartridge for measurement¹⁰⁾.

This indicates that by performing measurements with both sides of the activated carbon cartridge and determining the average value, radioiodine can be quantified with a bias of less than 10%.

However, in emergency monitoring, performing the measurement twice (three times if the glass fiber filter paper that collected particulate iodine is included) for one sample is a complex task, and there is also the problem of the machine time for the detector. Therefore, although this method is less accurate, measuring with the suction surface facing the detector is a safe evaluation method.

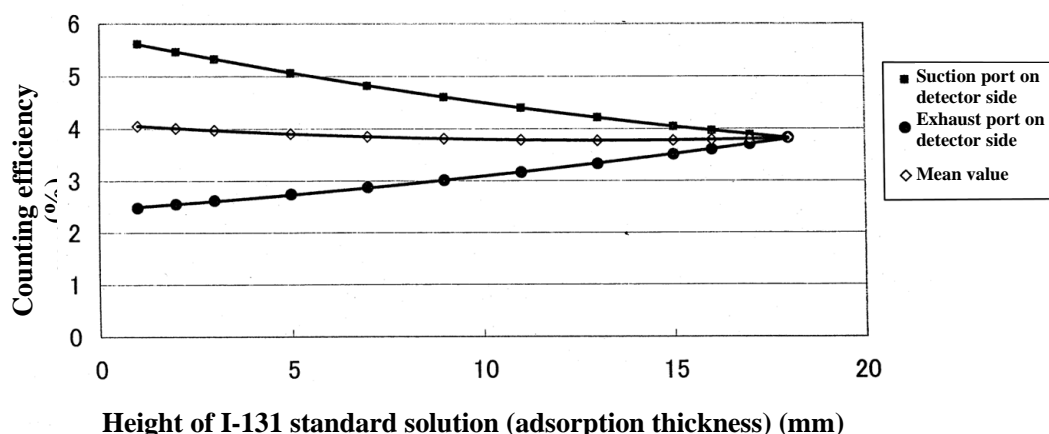


Figure A-2-1 Counting efficiency of I-131 (364 keV)
in the different adsorption layers of an activated carbon cartridge

A-3 Sum Peak of I-132

This section is quoted from Explanation B of The Series of Environmental Radioactivity Measuring Methods No. 29 “Gamma-ray Spectral Analysis Using a Germanium Detector in Emergencies (AI-Ge-E)”⁸⁾.

I-132, which has a half life of 2.3 h, emits multiple γ -rays and is a typical artificial radionuclide for which the sum peak is detected. The main γ -rays associated with the decay of I-132 are listed in Table A-3-1.

Table A-3-1 Main γ -rays associated with the decay of I-132

| γ -ray energy (keV) | Emission rate (%) | γ -ray energy (keV) | Emission rate (%) |
|-------------------------------|----------------------|-------------------------------|----------------------|
| 262.9 | 1.28 | 780.0 | 1.18 |
| 505.8 [(1)] | 4.94 | 809.5 | 2.6 |
| 522.7 [(2)] | 16.0 | 812.0 [(9)] | 5.5 |
| 547.2 | 1.14 | 876.6 | 1.04 |
| 621.2 | 1.58 | 954.6 [(10)] | 17.6 |
| 630.2 [(3)] | 13.3 | 1136.0 [(11)] | 3.01 |
| 650.5 | 2.57 | 1143.3 | 1.35 |
| 667.7 [(4)] | 98.70 | 1172.9 | 1.09 |
| 669.8 [(5)] | 4.6 | 1290.8 | 1.13 |
| 671.4 [(6)] | 3.5 | 1295.1 | 1.88 |
| 727.0 | 2.2 | 1372.1 | 2.47 |
| 727.2 [(7)] | 3.2 | 1398.6 [(12)] | 7.01 |
| 728.4 | 1.6 | 1442.6 | 1.40 |
| 772.6 [(8)] | 75.6 | 1921.1 | 1.23 |

Note 1: The source of the nuclear data is ENSDF (as of October 2017).

Note 2: γ -rays with an emission rate of more than 1% and less than 2000 keV are listed.

Note 3: γ -rays with an emission rate of 3% or higher are assigned numbers (1) to (12) in order of energy.

Note 4: γ -ray energies are shown with one decimal place, and emission rates are shown with two decimal places.

When the emission rate does not have two decimal places due to the energy of γ -rays, it is shown with one decimal place.

The decay scheme of I-132 is shown in Figure A-3-1. Because I-132 emits a large number of γ -rays, the pattern of the energy transitions of the cascade of γ -rays emitted is complicated. Owing to the lower probability of γ -rays with smaller emission rates that cause the sum effect, for the sake of simplicity, γ -rays with emission rates of 3% or higher, i.e., γ -rays (1) through (12), are considered, as shown in Table A-3-1. Table A-3-2 shows the results of the determination of the likelihood of γ -rays appearing as a sum peak in the γ -ray spectrum by combining γ -rays numbered from (1) to (12) in order of decreasing energy.

2.2 hr.

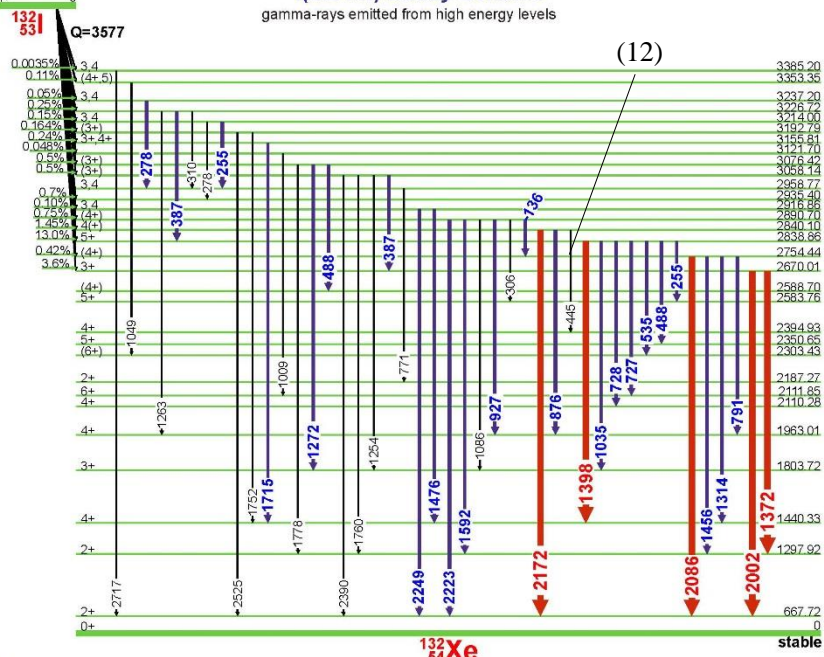
 ^{132}I (2.2 hr.) Decay Scheme
gamma-rays emitted from high energy levels

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2.2 hr.

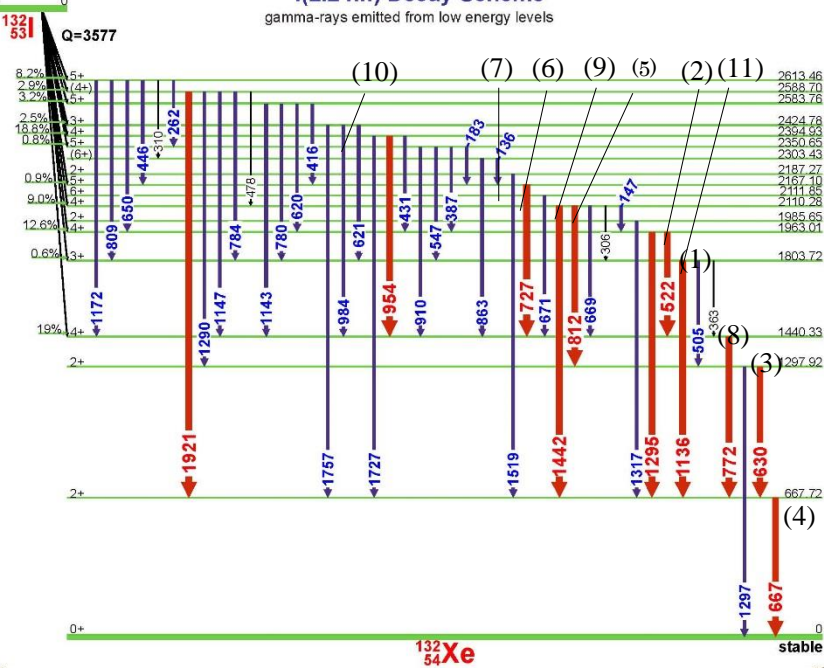
 ^{132}I (2.2 hr.) Decay Scheme
gamma-rays emitted from low energy levels

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Note: Arrows indicating γ -ray energy transitions are color-coded using sensitivity indices obtained from the peak and baseline regions in the γ -ray spectrum.

Identifying peaks in the γ -ray spectrum is easier in the order of red > blue > black (not in the order of emission rate).

Figure A-3-1 Decay scheme of I-132

Source: Online Spectrum Catalogs for Ge and Si(Li), Idaho National Laboratory

(URL: http://www4vip.inl.gov/gammaray/catalogs/ge/catalog_ge.shtml)

Table A-3-2 Table used for determining the appearance of γ -ray
sum peaks associated with the decay of I-132

| | (1) | (2) | (3) | (4) | (5) | (6) | (7) | (8) | (9) | (10) | (11) | (12) |
|------|-----|-----|-----|-----|-----|-----|-----|-----|-----|------|------|------|
| (1) | | | *1 | *4 | | | | | | | | |
| (2) | | | | ○ | | | | *2 | | | | |
| (3) | | | | *2 | | | | | *2 | | | |
| (4) | | | | | ○ | ○ | ○ | ○ | ○ | ○ | ○ | *3 |
| (5) | | | | | | | | *2 | | | | |
| (6) | | | | | | | | *4 | | | | |
| (7) | | | | | | | | ○ | | | | |
| (8) | | | | | | | | | | *4 | | *3 |
| (9) | | | | | | | | | | | | |
| (10) | | | | | | | | | | | | |
| (11) | | | | | | | | | | | | |
| (12) | | | | | | | | | | | | |

*1: Energy transition equivalent to the γ -rays of (11)

*2: Energy transition equivalent to other γ -rays

*3: 2000 keV or higher

*4: Difficult to distinguish from energy transitions induced by other γ -rays.

From Table A-3-2, the energies of sum peaks that appear in the γ -ray spectrum are as follows. Note that I-132 emits γ -rays at 1727.2 keV, and although it is difficult to distinguish this emission peak from the sum peak [(8) + (10)], it is listed below as the sum peak because its γ -ray emission rate is 0.067%.

- 1190.4 keV [(2) + (4)]
- 1337.5 keV [(4) + (5)]
- 1339.1 keV [(4) + (6)]
- 1394.9 keV [(4) + (7)]
- 1440.3 keV [(4) + (8)]
- 1479.7 keV [(4) + (9)]
- 1499.8 keV [(7) + (8)]
- 1622.3 keV [(4) + (10)]
- 1727.2 keV [(8) + (10)]
- 1803.7 keV [(4) + (11)]

Figure A-3-2 shows a γ -ray spectrum that exhibits some of the sum peaks listed above.

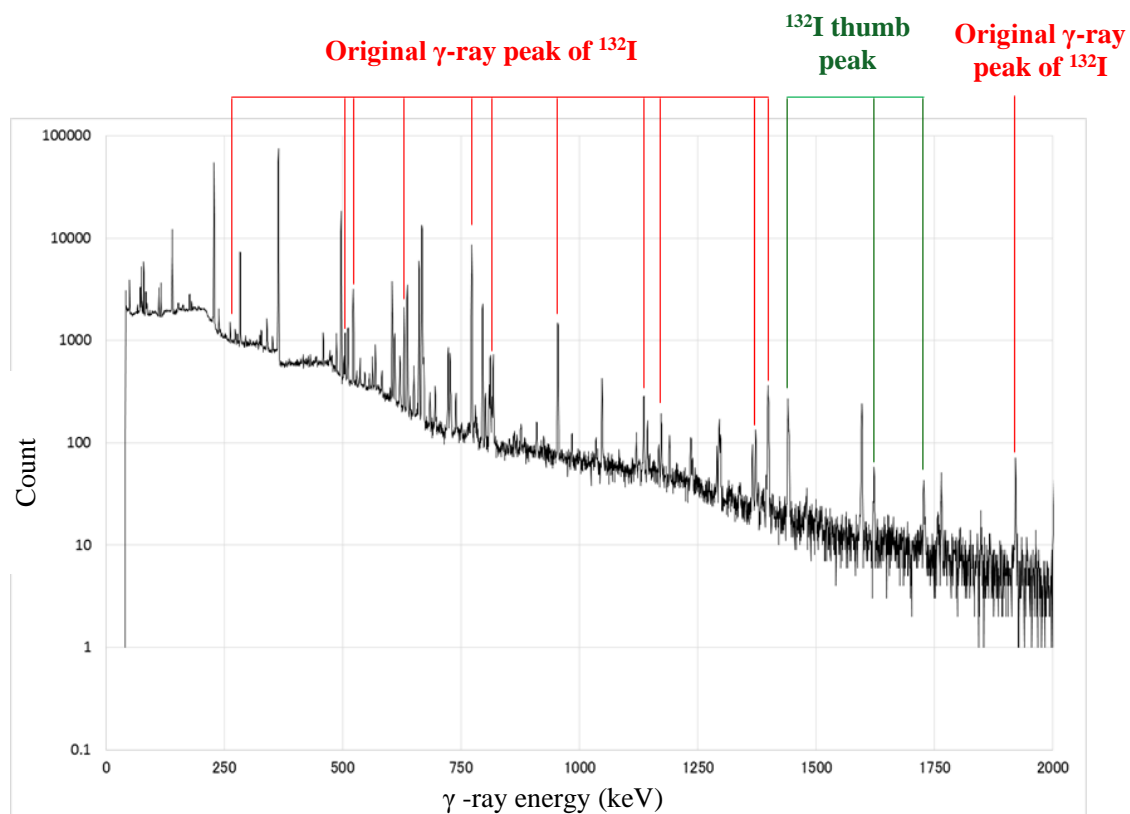


Figure A-3-2 γ-ray spectrum demonstrating the sum peaks of I-132
(Airborne dust collected during the Chornobyli nuclear power plant accident)

Almost all energy transitions after the β -decay of I-132 shown in Figure A-3-1 are via the energy transition of 667.7 keV (emission rate: 98.70%). Therefore, as the sum peaks other than those mentioned above may appear, there are cases where the sum peaks due to the combination of 667.7 keV γ -rays and γ -rays with an emission rate of 3% or less should be considered.

Information B In-situ Measurement

In situ measurements can identify radioactive materials that have been released into the environment and deposited on the ground surface as well as estimate radioactivity concentrations and spatial radiation dose rates at any location, allowing the determination of the areal distribution of radionuclides.

These measurements are highly mobile and quick because the real-time measurement results can be obtained on site. In addition, because measurements are taken over the entire ground surface, average measurement results around the measurement point can be obtained. Therefore, the approach has the advantage of eliminating the problems of sample heterogeneity and representativeness that arise when soil is collected and brought back to the facility for measurement.

To measure radioactive iodine via an in situ measurement, information on the energy of γ -rays emitted from radioiodine is required. γ -rays spectrometry using a germanium semiconductor detector or an NaI(Tl) scintillation detector is effective for this purpose.

Germanium semiconductor detectors and various scintillation detectors used for in situ measurements are described below.

For details regarding in situ measurements using a germanium semiconductor detector, refer to The Series of Environmental Radioactivity Measuring Methods No. 33 “In Situ Measurement Using a Germanium Detector (part1, part2) (DR-SITU)”¹⁶⁾.

[In situ measurements using a germanium semiconductor detector]

The germanium semiconductor detector used for in situ measurements is basically the same as the stationary detector normally employed in facilities; however, because measurements are performed outdoors, it has a compact design for easy transportation and installation.

Energy and peak efficiency calibrations for this detector must be performed in the same way as for the ordinary germanium semiconductor detector. Additionally, as γ -rays incident from the entire ground surface are measured, angular dependence must be considered for peak efficiency calibration. In addition, because measurements are performed outdoors, care should be taken to protect the measuring equipment and to prevent its contamination.



Figure B-1 Example of a liquid nitrogen-cooled germanium semiconductor detector for in situ measurements

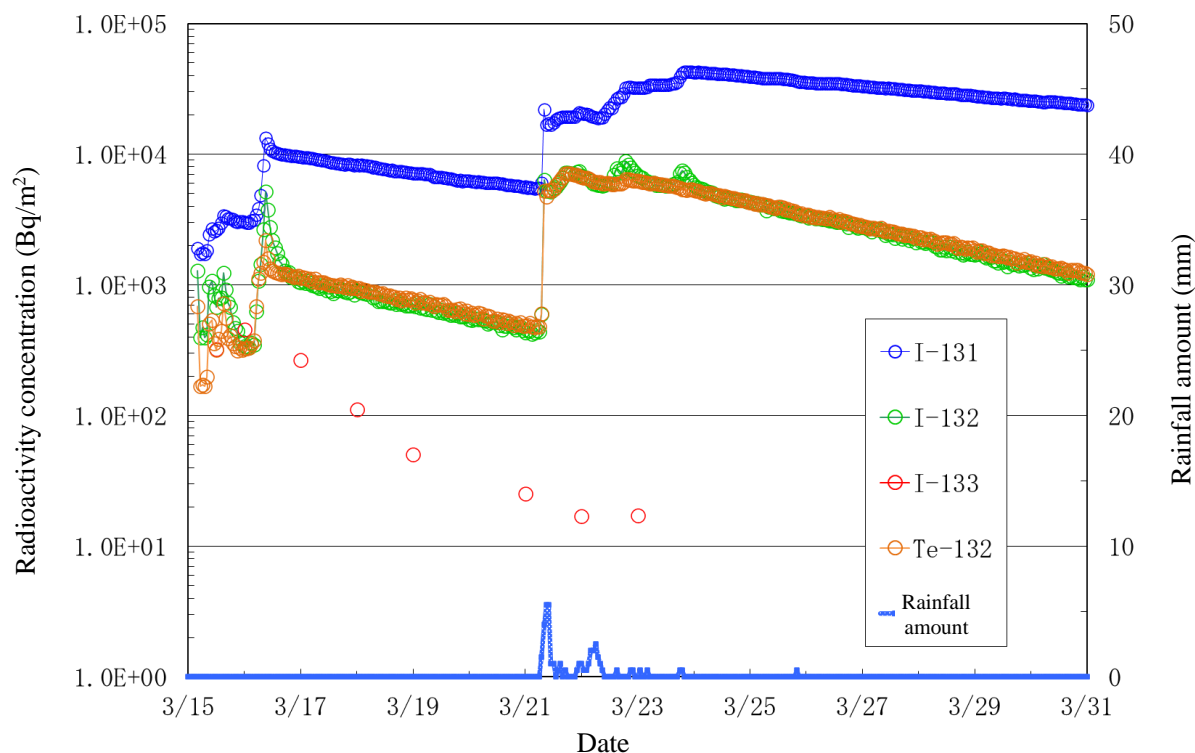
Using the HASL* method developed by Beck et al. (hereinafter referred to as the HASL method), the radioactivity concentration (radioactivity per unit area: Bq/cm²) and ambient radiation dose rate for radioactive materials deposited on the ground surface can be calculated.

*HASL: Health and Safety Laboratory

(now renamed to National Urban Security Technology Laboratory (NUSTL))

The HASL method calculates the radioactivity concentration and ambient radiation dose rate under several assumptions, such as the surrounding topography, vertical distribution of radioactive materials in soil, and detector installation height (1 m). Although the degree of soil penetration is important in analysis, in the case of radioiodine, it is assumed that it decays before penetrating due to its short half life; therefore, only the distribution on the ground surface is considered.

Figure B-2 shows an example of the evaluation of the deposited amounts of radioiodine, etc. measured in Chiba City, Chiba Prefecture, in March 2011 using the HASL method. Radioiodine and other elements are detected from March 15, and on March 21, radioiodine that had existed in air is deposited on the ground surface due to rainfall, resulting in an increase in the radioactivity concentration. Because this method assumes that all radioiodine is deposited on the ground surface, accurately evaluating the contribution from radioiodine (gaseous and particulate) present in air is difficult.



* Data are one-hour values, and periods with no plot correspond to the case where radioiodine was not detected.

* Germanium semiconductor detector: installed about 1 m above the ground and has a relative efficiency of about 25%.

Figure B-2 Example of the evaluation of deposited amounts of radioiodine, etc.
in Chiba City, Chiba Prefecture

[Example of a transportable spectrometer]

An NaI(Tl) scintillation detector is often used to obtain γ -ray spectra, but detectors employing scintillators made of other materials are also commercially available.

As for materials, inorganic scintillators made of halogen compounds are often used and have advantages, such as higher luminous efficiency and absorption coefficient and a larger proportion of photoelectric effect than organic scintillators. Furthermore, some materials may be deliquescent and difficult to handle, such as being vulnerable to shocks. In recent years, however, detectors made of materials that are not deliquescent and have high density have been put to practical use. Table B-1 lists the primary scintillator characteristics.

Although the energy resolution of detectors made from other materials is not as good as that of germanium semiconductor detectors, the former is more transportable because they do not require cooling.

Table B-1 Main scintillator characteristics

| Materials | Density (g/cm ³) | Relative luminous intensity* |
|------------------------|------------------------------|------------------------------|
| NaI(Tl) | 3.67 | 100 |
| CsI(Tl) | 4.51 | 45-50 |
| SrI ₂ (Eu) | 4.59 | 210 |
| LaBr ₃ (Cs) | 5.29 | 130 |

*Relative values with NaI(Tl) as 100.

The scintillation detectors listed in Table B-1 exhibit the following characteristics:

- NaI(Tl)

Because it has good energy resolution and is easily available, it is often used as a detector to measure γ -rays. Because it is sensitive to temperature changes and has a deliquescent nature, it is used as sealed in a metal container. The energy resolution is ~8% for the 662-keV peak of Cs-137.

- CsI(Tl)

Crystals have a cubic structure with no cleavage planes and relatively low hardness, making them resistant to cracking and impact. Therefore, it can be used in places where vibration and environmental changes are considerable. The energy resolution is comparable to that of NaI(Tl).

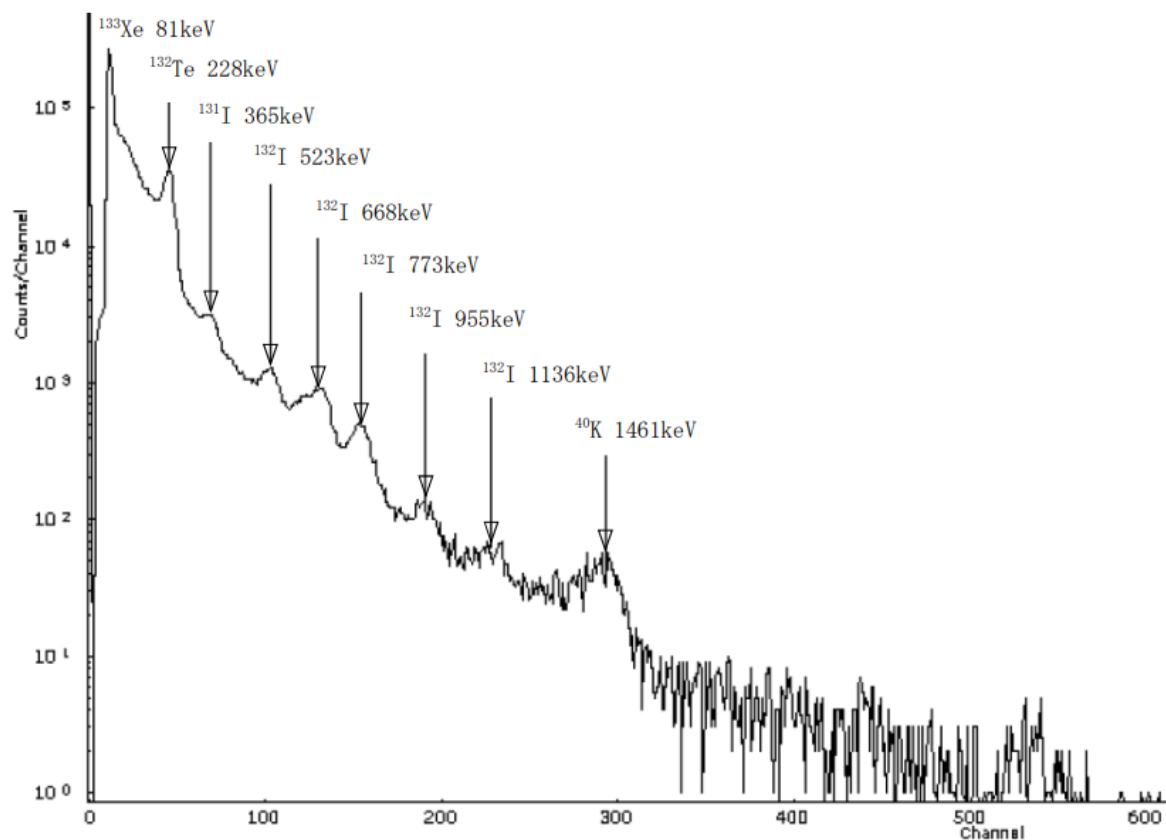
- SrI₂(Eu)

A scintillator with a high luminous output and an energy resolution of less than 4% as well as low background due to the absence of self-radioactivity (self-dose) inherent to the detector.

- LaBr₃(Cs)

The energy resolution is about twice as that of NaI(Tl), and temperature stability is high. The short decay time makes it suitable for measurements in high-counting-rate fields.

Figure B-3 shows the spectrum measured during the 1F accident, although data were obtained using a NaI(Tl) scintillation detector installed at a fixed monitoring post, rather than a transportable one. Peaks ascribed to radionuclides such as I-131 and I-132 can be identified.



Source: The Series of Environmental Radioactivity Measuring Methods No.17 “Method for Measurement of Environmental Gamma-rays with a Continuous Monitor (DR-CNT)”¹⁷⁾

Figure B-3 NaI spectra (600-s measurement) measured on March 13, 2011, in the vicinity of the TEPCO Fukushima Daiichi Nuclear Power Plant

Information C Measurement of Radioiodine via Continuous Monitoring

An iodine monitoring post is a device that combines an iodine sampler with an automatic sample changer and a detector. The purpose of this device is to continuously perform tasks from sampling air to measuring radioactive iodine. Detectors used include NaI(Tl) scintillation detectors and germanium semiconductor detectors.

In this measurement method, as a general rule, the glass fiber filter and activated carbon cartridges with the sample are retrieved from the iodine sampler with an automatic sample changer and accurately measured using a germanium semiconductor detector at an analytical laboratory or other facility. However, the iodine monitoring post measures the radioiodine concentration at the same time as it is collected, allowing the rapid determination of the radioiodine concentration. It shall be noted that the NaI(Tl) scintillation detector does not give quantitative results, while the germanium semiconductor detector is more expensive to install and requires the cooling of the germanium crystal.

[Example of an iodine monitoring post]



Photo C-1 Example of an iodine monitoring post

Detector: $\varnothing 2'' \times 2''$ NaI(Tl) scintillation detector

Measurement target: I-131, etc.

Absorbent: activated carbon cartridge, etc.

Detection limit: 3.7×10^{-7} Bq/cm³ (I-131 equivalent)

Information E Estimation of the I-131 Radioactivity Concentration using Iodine Isotope Ratios

I-131 has a physical half life of ~8.0 days. Therefore, as more time is elapsed after the release of radioactive materials from a nuclear facility, the more difficult it becomes to assess its released and deposited amount. For this reason, a study¹⁸⁾ exploring a method that can be used to estimate the I-131 radioactivity concentration using I-129, an isotope of iodine with a long physical half life of 1.57×10^7 years, as an indicator has been conducted after the 1F accident. It is introduced here.

The background of the study is as follows. Soil samples collected from approximately 2,200 sites (5 samples per site) in June 2011 after the 1F accident were analyzed. Radiocesium was detected in most of them. However, most I-131 had already decayed, and it was detected only in 421 samples. Therefore, the map used to determine the distribution of radioactive materials for radioiodine was coarser than that for radiocesium (Figure E-1). In an attempt to refine the I-131 distribution map, a method was developed to estimate the I-131 concentrations from the I-129 concentrations at sites where I-131 was not detected.

E.1 Analytical Methods for I-129

Soil samples collected on June 2011 (generally within 80 km of the 1F) were analyzed for I-129.

Iodine was separated from soil by heating the mixture to realize iodine volatilization and then collecting it in a trap solution. The I-127 concentration was measured from a portion of the trap solution via ICP-MS spectrometry. The remaining trap solution was chemically treated to prepare silver iodide (AgI) and utilized to measure the I-129/I-127 atomic ratio using accelerator mass spectrometry (AMS)^{15),19)}.

E.2 Calculation of the I-131 concentration

The I-131/I-129 concentration ratio required to calculate the deposition amount of I-131 from the I-129 analysis results was “9,368,” which was obtained by analyzing I-129 in samples in which I-131 was detected in 2011 when the 1F accident occurred. The results showed a good correlation between the I-129 and I-131 concentrations (correlation coefficient of 0.84)¹⁸⁾. The depth distribution of I-129 indicated that most of it was retained in the surface layer (5 cm). Therefore, the I-131 deposition amount could be estimated by analyzing I-129 in the surface soil. Additionally, it has been reported that this method results in an error of up to 35%.

The method used for calculating the amount of I-131 fallout (Bq/m²) from the results of I-129 analysis is outlined below.

- 1) Convert the I-129 concentration (by weight) in soil to radioactivity concentration (mBq/kg)
- 2) Calculate the amount of I-129 deposited (Bq/m²) from the radioactivity concentration of I-129 (mBq/kg)
The radioactivity concentration was converted to the deposition amount considering the weight of the topsoil collected at a depth of 5 cm (contained in a small container) and area of the small container and converting it into the I-129 radioactivity concentration per unit area (1 m²).
- 3) The I-131 deposited amount (Bq/m²) was calculated by multiplying the deposition amount of I-129 determined above by the aforementioned I-131/I-129 concentration ratio (decay correction was as of June 14, 2011).

E.3 Estimation of the average deposition of I-131 at soil sampling sites

When measuring with a germanium semiconductor detector, as a rule, five samples were measured at each measurement point and the average value was taken. However, because the analysis of I-129 is time-consuming and laborious, only one sample was selected for analysis at each measurement point. Therefore, to correct for the variation within measurement sites, the ratio of the Cs-137 deposition amount of the collected samples to the average Cs-137 deposition amount for five samples from each site was calculated to estimate the average I-131 deposition amount. In other words, the average I-131 deposition amount was calculated by multiplying the I-131 deposition amount in the sample by the following factor (F).

$$F = \frac{\text{Average value of the Cs-137 deposition amount of five samples from each site}}{\text{Cs-137 deposition amount of the sample}}$$

The I-131 distribution map was improved using the I-131 deposition amount obtained via the above method. Figure E-1 shows the distribution map of 421 samples in which I-131 was directly detected. Figure E-2 displays a distribution map of 1,162 samples, including the I-131 deposition of 741 samples estimated based on I-129.

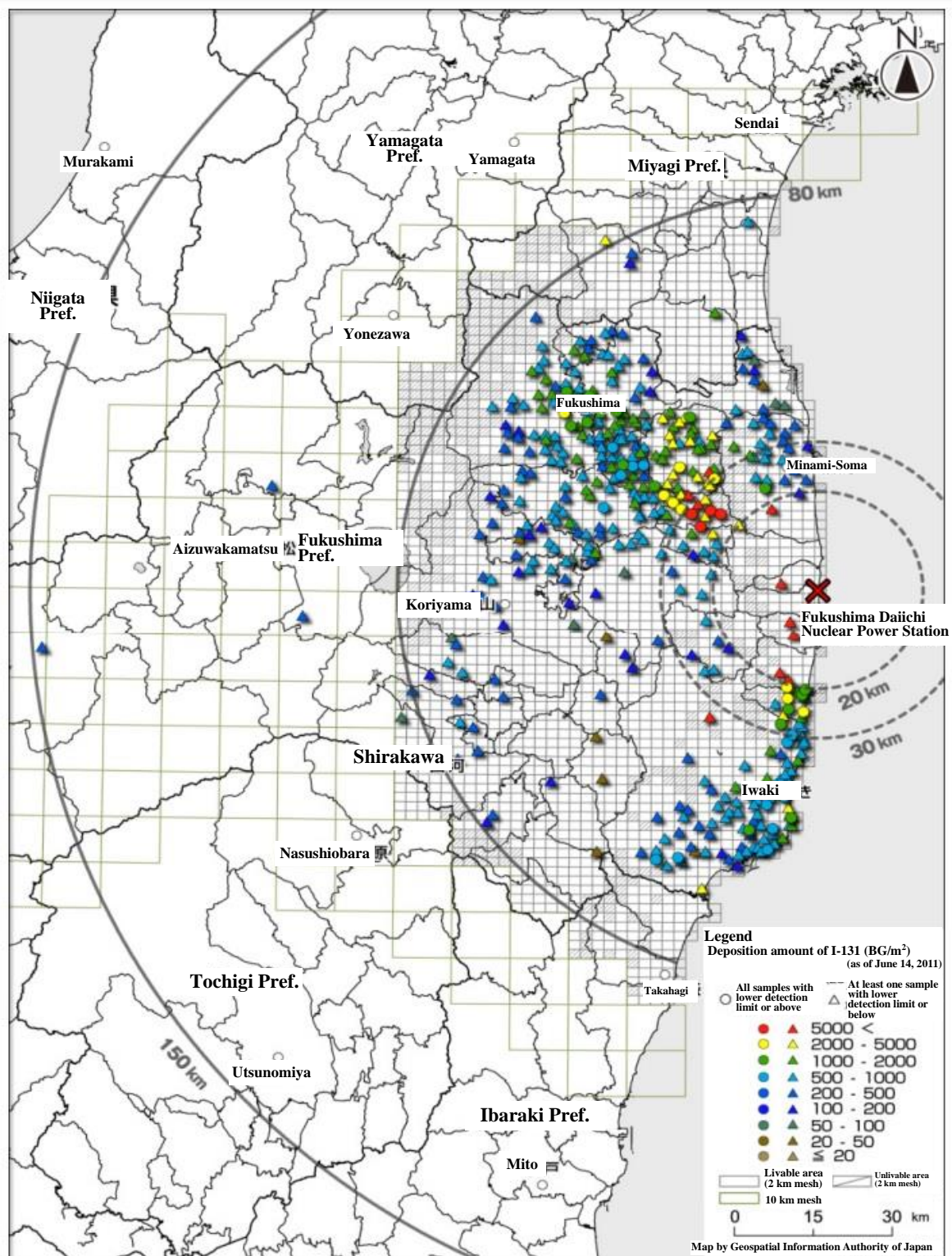


Figure E-1 Map of the I-131 deposition amount measured in the June 2011 survey (Bq/m^2)¹⁸⁾
(Decay correction: June 14, 2011)

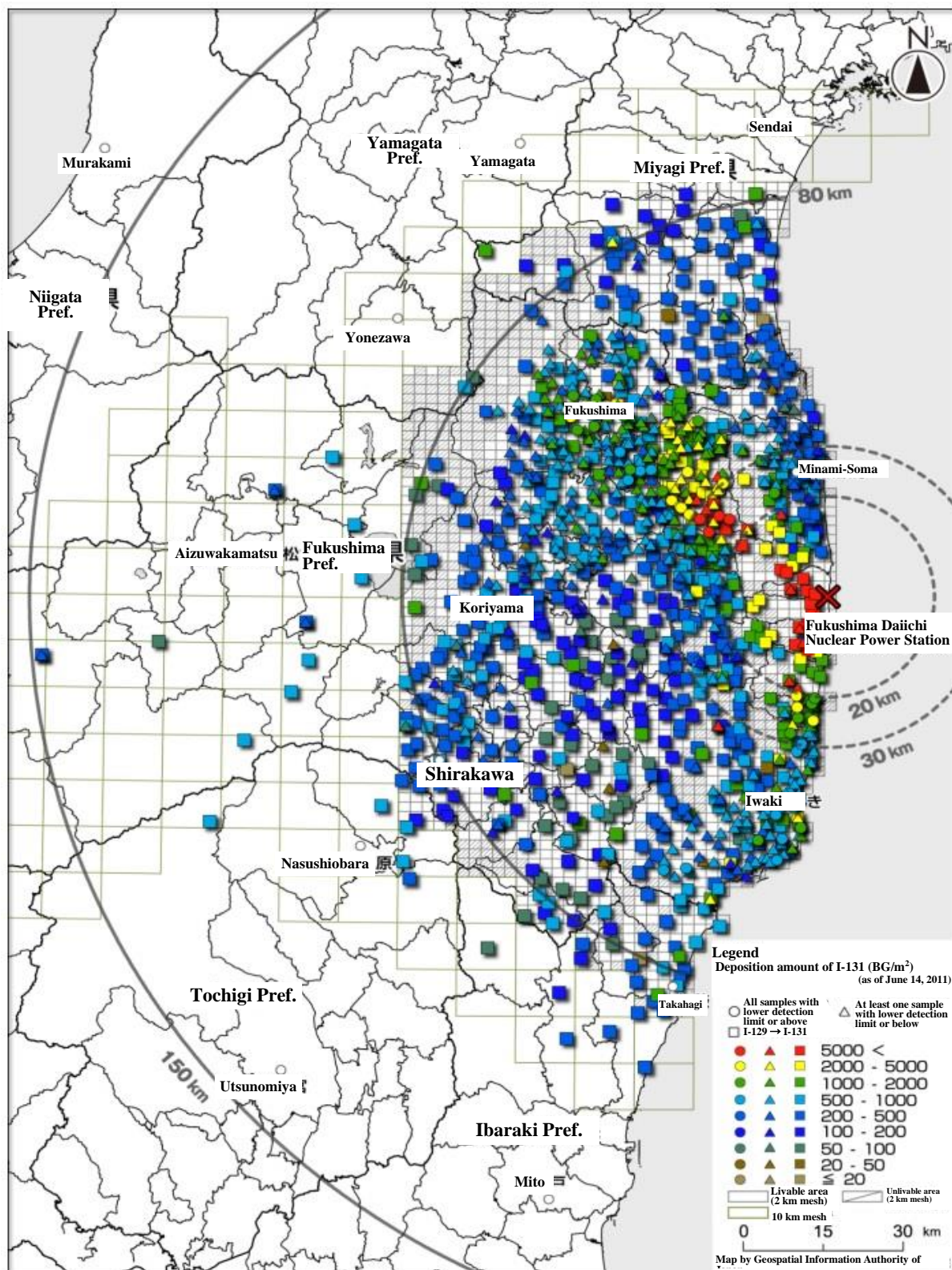


Figure E-2 Map of the I-131 deposition amount reconstructed from overlying the I-129 map on Figure E-1
(Bq/m²)¹⁸⁾
(Decay correction: June 14, 2011)

Appendix

Appendix A Terminology

Definitions of terms used in this measurement method are given below.

| Term | Description |
|--|---|
| Emergency environmental radiation monitoring | Environmental radiation monitoring to be conducted at the time of facility-site emergency and full-scale emergency situations based on the Guideline for Emergency Preparedness and Response. |
| Information gathering situation | A stage where it is necessary to confirm whether or not radioactive materials have been released. |
| Alert situation | A stage in which an abnormal event has occurred or is likely to occur at a nuclear facility, and it is necessary to start preparing protective measures such as collecting information, preparing for emergency monitoring, and evacuating people on the facility premises in an emergency. |
| Facility-site emergency situation | A stage in which an event that may have a radiological impact on the public has occurred at a nuclear facility, and it is necessary to begin preparations for major protective measures, such as evacuation, in preparation for an emergency in the vicinity of the nuclear facility. |
| Full-scale emergency situation | A stage in which an event has occurred at a nuclear facility that is highly likely to have a radiological impact on the public, and prompt protective measures must be implemented to avoid (minimize) severe deterministic effects and reduce the risk of stochastic effects. |
| OIL | Abbreviation for operational intervention level. |
| Iodine sampler with an auto sample changer | A device used for the continuous sampling of particulate and gaseous radioactive iodine by replacing glass fiber filters and activated carbon cartridges at regular intervals. |
| Germanium semiconductor detector | A device employed in γ -ray spectrometry by exploiting the properties of germanium as a semiconductor. |
| Transportable sampler | Transportable dust sampler. In the measurement method described in this text, two main types of samplers are used: low-volume and high-volume air samplers. |
| Low-volume air sampler | A device that collects airborne dust for several days to a week at a suction rate of 20–100 L/min. |
| High-volume air sampler | A device that collects airborne dust by sucking in a large amount of air in a short period of time at a suction rate of 200–2000 L/min. |
| Air monitoring post | A device used for the continuous measurement of radioactivity concentrations in airborne particles, employed in emergencies. Although the measurement principle is the same as that of dust monitoring posts, in this measurement method, the air monitoring post refers to an additional device installed for emergency use, which is designed to measure the concentration of radioactive materials in air quickly and at short intervals. |
| Glass fiber filter paper | In this measurement method, it is used as an absorbent for particulate iodine. |
| Activated carbon cartridge | In this measurement method, it is used as an absorbent for gaseous iodine. |
| Radioiodine | Collective name for the radioactive isotopes of iodine. |
| Particulate iodine | Radioiodine in the solid form, which can be collected using a glass fiber filter paper. |
| Gaseous iodine | Radioiodine in the gaseous form, which can be collected using an activated carbon cartridge. |

| Term | Description |
|--|--|
| Half life | <p>The time required for radioactivity to decrease to half of what it was at a certain reference time.</p> <p>Half lives are specific to each nuclide and can range from much shorter than one second to more than several billion years.</p> <p>Let the half life be T, elapsed time be t, and radioactivity at the reference time ($t = 0$) be A_0, then radioactivity A at time t is expressed as follows:</p> $A = A_0 e^{-\lambda t} \quad \text{or} \quad A = A_0 \left(\frac{1}{2}\right)^{t/T}$ <p>Note that $\ln(2) \cong 0.693$ and that the relationship between the half life and decay constant λ is $T = \ln(2)/\lambda$.</p> |
| Internal exposure | Taking in radioactive materials into the body through inhalation, ingestion, etc. and receiving radiation from a radiation source within the body. |
| γ -ray (Gamma-ray spectrometry) | <p>A technique employed for obtaining γ -ray spectra using germanium semiconductor detectors, etc., and a method for determining radioactivity by analyzing the resulting spectra.</p> <p>Also known as γ -ray spectroscopy.</p> |
| Spectrum | <p>The energy distribution of radiation or pulse wave height distribution, generally refers to the latter.</p> <p>In the case of α- and β-rays, spectra have a nearly similar energy distribution. In the case of a γ-ray spectrum, it is essentially the energy distribution of fast secondary electrons absorbed in the effective volume of the detector. However, the actual γ-ray spectrum exhibits a broad pulse wave height distribution due to various “fluctuations,” such as statistical fluctuations and noise, during conversion from the secondary electron energy to the output pulse wave height.</p> |
| Peak efficiency | <p>Counting efficiency calculated from the net counting rate of a peak in α-ray spectrum or γ-ray spectrum measurement.</p> <p>In its most basic definition, peak efficiency is expressed as</p> $\varepsilon = \frac{n}{I}$ <p>where n is the peak net count rate and I is the measured intensity of radiation emitted from the source.</p> <p>This is the most important quantitative factor in γ-ray spectral analysis. In addition to being dependent on the radiation energy and geometry of measured samples, it may also include complex correction factors, such as self-absorption and summing effects.</p> |
| Counting efficiency | <p>A constant used to determine radioactivity or radiation intensity from a measurement value (counting rate).</p> <p>Let radioactivity be A (Bq), the emission rate of radiation to be measured be I_γ, and the net counting rate be N (s^{-1}), then counting efficiency ε is expressed as follows:</p> $\varepsilon = \frac{N}{A \cdot I_\gamma}$ <p>This relationship includes geometric efficiency, but may not include it when referring to detector efficiency only (also called detection efficiency).</p> |

| Term | Description |
|-----------------------|--|
| Sum peak | <p>A peak that occurs when multiple γ-rays enter the detector almost simultaneously and are detected, and a signal corresponding to the sum of the energies of these γ-rays is output.</p> <p>The sum peaks are prominent for nuclides that emit multiple γ-rays in a single decay event (e.g., Cs-134 and I-132).</p> <p>In the measurement of samples containing high concentrations of radionuclides, peaks also appear due to random summation caused by the accidental detection of γ-rays (e.g., Cs-134 + Cs-137 and Cs-137 + Cs-137) emitted as a result of different nuclear decay events occurring at almost the same time.</p> |
| Lower detection limit | <p>The minimum amount (value) that can be detected for the nuclide being analyzed when the measurement sample and measurement conditions (measurement device, measurement time, etc.) are determined.</p> |
| Quantifiable level | <p>The level at which quantification is considered possible. It is set based on the accumulation of empirical values for the lower detection limit when a series of analysis conditions such as the sample type, sample volume, and measurement time are the same.</p> <p>In this measurement method, it is determined by calculating the lower detection limit for each type of environmental sample and measurement time using the actual spectra of the environmental samples collected and packed in Marinelli beakers or small containers after the 1F accident.</p> |

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