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Review

# Rapid Test Method of Radio-iodine Radioactivity in Foods/foodstuffs

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Rapid test of radioactivity contamination in foods/foodstuffs is required to ensure the radiological protection of the public in emergency situation. Just after the Fukushima accident rapid screening of radio-iodine radioactivity in foods/foodstuffs was carried out through over the country. In the present paper, the rapid test method based on the Ministry of Health, Labour and Welfare manual for radioactivity measurements in emergency, which were specified as the normative references for radioactivity monitoring of foods/foodstuffs by testing laboratories, is shown with some considerations for the expected future improvement.

*Key words:* screening, radioactivity measurement, nuclear accident, NaI(Tl) detector

## 1. Introduction

The magnitude 9 huge earthquakes happened in the east part of Japan in March 2011. Successively, a big tsunami hit the Pacific coast causing serious damage to buildings, houses and facilities in the north east district of Japan. At Fukushima Dai-ichi Nuclear Power Station (NPS) was also damaged due to the earthquake and seawater flooding caused by the sub-sequent tsunami. In consequence, a large amount of radioactive materials were released to the environment due to hydrogen explosions[1]. To meet such an emergent situation, Ministry of Health, Labour and Welfare (MHLW) in Japan implemented the regulations including the indices for food and beverage intake restriction as provisional regulation values of radioactive materials in foods in accordance with the food sanitation act on 17th March, 2011 (see Table 1)[2]. At the same time MHLW familiarize local authorities and institutes concerned with the procedure-manual for radioactivity measurements in emergency[3]. According to the MHLW survey data (as of 26th March, 2011) acquired after the notification of the provisional regulation values radioactivity in excess of the provisional regulation values was detected in 23 cases out of 128 samples of raw milk (detected value: radioactive iodine 310-5,300 Bq/kg, radioactive cesium 420 Bq/kg), 76 cases out of 356 samples of vegetables (detected value: radioactive iodine 2,080-54,100 Bq/kg, radioactive cesium 510-82,000 Bq/kg)[4]. Figure 1 shows the spectrum obtained with a tap water sample collected at Japan Radioisotope Association (JRIA) located in Tokyo on 17th March. As radio-iodine monitoring should be given top priority for foods and beverages. The primary health concern after a nuclear accident is the internal exposure of thyroid with radio-iodine[5]. Since rapid tests for radio-iodine should have priority to time-consuming accurate measurements in this situation, the methods employed in the guidance for radioactivity measurements were designed not only for ordinal gamma-ray spectrometry using a HP-Ge detector but also for rapid test of radio-iodine radioactivity in foods/foodstuffs by simple gamma-ray detection with

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**Table 1.** Provisional regulation values of radioactive materials in food in accordance with the food sanitation act (excerpted version)

| Nuclide | | Index value (Bq/kg) |
|---|---|---|
| Radioactive iodine[a] | Drinking water | 300 |
| | Milk, dairy products[b] | |
| | Vegetables (Except root vegetables and tubers) | 2000 |
| | Fishery products | |
| Radioactive caesium | Drinking water | 200 |
| | Milk, dairy products[2)] | |
| | Vegetables (Except root vegetables and tubers) | 500 |
| | Fishery products | |

[a] Representative radio-nuclides among mixed radio-nuclides: $^{131}$I
[b] Provide guidance so that materials exceeding 100 Bq/kg are not used in milk supplied for use in powdered baby formula or for direct drinking.

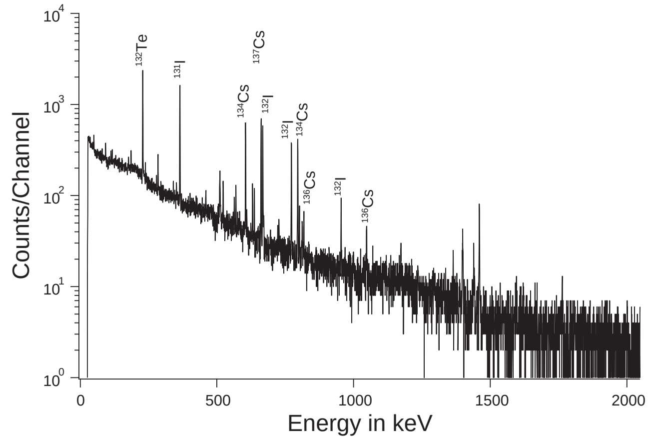


**Fig. 1.** The spectrum obtained with a tap water sample collected at Japan Radioisotope Association located in Tokyo on 17th, March. This spectrum was measured using a HP-Ge detector.

conventional survey meters.

In this paper, rapid tests of radio-iodine radioactivity in foods/foodstuffs carried out in Japan just after the accident based on the manual for radioactivity measurements in emergency specified by MHLW is reviewed.

## 2. Rapid test method for radio-iodine

Radioactivity measurements of foods/foodstuffs are important to make sure whether the sample-activity exceeds the legal limit or not after the accidental situation. When the most accurate determination of gamma emitting radionuclides in food samples is required, gamma-ray spectrometry using a HP-Ge detector should be recommended with an appropriate preparation of the test samples. Though a huge number of samples shall be tested rapidly to avoid sanitation hazards of foods/foodstuffs and beverages in an emergency situation, food inspection laboratories had more excessive work than they can manage in cases of the normal situation. In addition Ge detectors were in limited supply and furthermore they could not use under field conditions. In response to such a situation, the rapid test method for radio-iodine using a portable scintillation survey meter (dose rate meter) was suggested in the MHLW manual for radioactivity measurements in emergency. As a consequence many types of portable survey meter for screening the activity of radio-iodine in foods/foodstuffs had been widely in use.

The conventional portable survey maters have no spectrum-analysis-function, and only indicate the integral count rate of all pulses above the low-energy threshold. In the measurement using such equipment without any nuclide-discrimination-function, $^{131}$I could not be determined separately from other nuclides, and all radioactive nuclides should be treated as $^{131}$I in spite of the possible existence of other radio-iodine and radio-cesium as seen in Figure 1. Even if other radioactive short-lived nuclides (e.g. $^{132}$I) are contained, obtainable apparent activity is overestimated. However if this rapid test method is carried out to screen samples, overestimation towards the safe side should be acceptable on the screening purposes. The role of screening is not accurate activity measurement but reducing the number of samples to be measured with a Ge detector. Thus time-saving techniques shall be preferentially employed for this purpose.

Since these survey meters are intended mostly for ambient dose monitoring, they have no indication for $^{131}$I activity or $^{131}$I activity-conversion-factor in terms of "Bq/kg/(count per second)" or "Bq/kg/(Sv per hour)". Prior to the measurements, the equipment shall be therefore calibrated using a reference solution. However it might be not easy to perform the instrument-calibration by not sufficiently trained staff in such an emergency situation. In response to such restrictions, Japan Radioisotope Association (JRIA) carried out urgently the type testing of $^{131}$I activity conversion factor for some popular types of meters using reference solution, and provided these results on our web site.

According to the manual, NaI(Tl)-scintillation-type ambient dose rate maters are recommended as typical equipment fit for the present purpose. Sample measurements and type testing using a 2L Marinelli beaker and other types of containers were carried out as shown in Figure 2. The test was carried out at the JRIA laboratory in Tokyo. Background ambient dose rate was around 0.1 μSv/h in the room where testing was done. Almost all these instruments are indicated by Sv/h or $s^{-1}$ and can be switched to either indication. In the use of the meter

**Table 2.** The results of type testing of $^{131}$I activity conversion factor for 6 types of meters using reference solution. Detection limit was evaluated from standard deviation of reading of three times measurement. Counting time of one measurement is 60s for counter/timer system. As for rate meter system, typical time constant is 30s. Characteristic limits (DL and DT) were determined by use of ISO11929[6)]

| Manufacturer | Model | detector size | Conversion factor Bq/kg/cps | Detection Limit Bq/kg |
|---|---|---|---|---|
| Fuji Electric | NHC610B1 | 0.5“ϕ ×0.5” | 1.96E+02 | 3.23E+02 |
| Hitach-Aloka Medical | TCS172B | 1“ϕ ×1” | 4.57E+01 | 1.67E+02 |
| Health Physics Instruments | 5000S | 1“ϕ ×1” | 4.48E+01 | 9.83E+01 |
| LUDLUM | Model3 44-2 | 1“ϕ ×1” | 4.50E+01 | 1.44E+02 |
| LIR Radiation | identiFINDER | 1.4“ϕ ×2” | 2.65E+01 | 6.60E+01 |
| Berkeley Nucleonics Corporation | 940-2G | 2“ϕ ×2” | 9.14E+00 | 3.60E+01 |

**Fig. 2.** Assembly for calibration using a reference solution filled in a typical 2L Marinelli beaker.

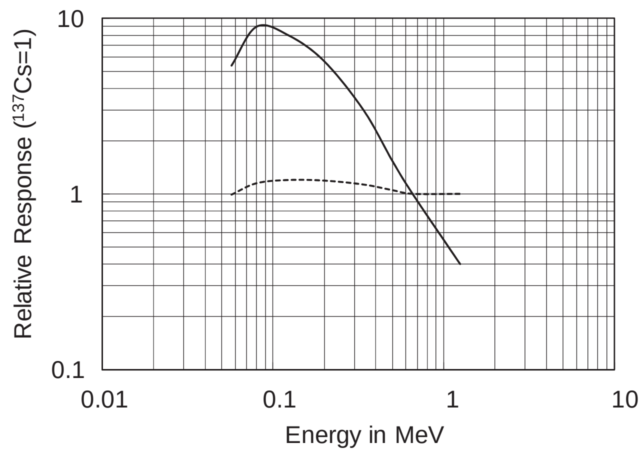


**Fig. 3.** An example of relative response of dose equivalent meters that consist of a NaI(Tl) scintillation detector. The dotted line indicates corrected relative response for $H^*(10)$. The solid line indicates uncorrected relative response of a NaI(Tl) detector for integral count rate.

as an ambient dose (rate) meter with a scale of Sv/h, an special algorism is usually involved in the meter to flatten the response with respect to the ambient dose rate $H^*(10)$ against photon energy. In the other mode with the scale of $s^{-1}$, the indication reflects the direct count rate of pulses from the scintillation detector without any correction. As can be seen in the solid line of Figure 3, the relative response with respect to the simple count rate is enhanced very much in the region below 0.6 MeV, and amounts to nearly 10 times as compared with the response at 662 keV photons. Higher response for $^{131}$I ($E_\gamma$: 0.365 MeV) is therefore expected in the count rate mode. Thus the guideline recommends use of the direct response mode with the indication of $s^{-1}$ rather than the ambient dose mode with the indication of Sv/h, and hence we employed the direct count rate mode on our testing.

Table 2 shows the result of the testing together with the detection limit (DL), with a 0.05 probability of the error of both the first and second kind confidence level, of each instrument. Here, $\tilde{y}_0 = 2y^*$ is employed as a simple approximation, where $\tilde{y}_0$ indicates the DL, $y^*$ decision threshold (DT). Although DL level was dependent on the detector size, setting of time constant (for rate meter), setting of counting time (for counter/timer), background rate and other random or systematic reasons, as seen the result in Table 2, DL level remains below 330 Bq/kg at the worst under the test condition shown in Table 2, and it became clear that 2000 Bq/kg radio-iodine can be detected with enough margin by a portable meter without any shield in this test condition. This simple method can be applied to the screening even for drinking water, milk and other dairy products, if the portable meter is equipped with a NaI(Tl) scintillation detector having the size of 1“ϕ ×1” or larger. Figure 4 shows the relation between the detection limit or the decision threshold and ambient dose rate of background for a type of TCS172B (Hitachi-Aloka Medical, Ltd.). As seen in this figure it is still possible to screen below 500 Bq/kg under the ambient dose rate of 1.0 μSv/h.

Figure 5 shows the results of monitoring of vegetable produced at Fukushima[7)]. As seen in this graph, 50% of samples have activity below 100 Bq/kg in March and 80% in April. It implies that these samples could be screened by this method efficiently in a short time.

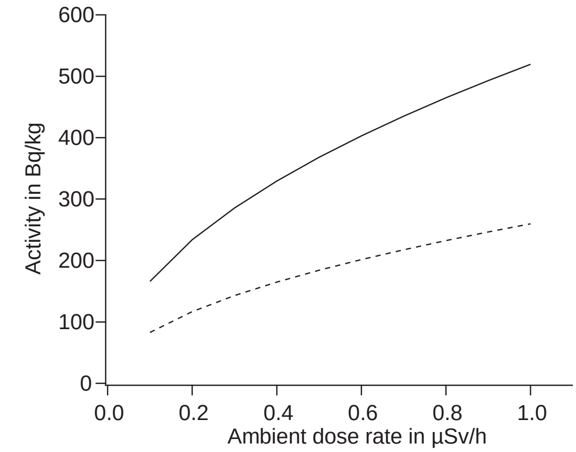


**Fig. 4.** Determined detection limit (solid line) and decision threshold (dotted line) for a type of TCS172B(Hitachi-Aloka Medical, Ltd.) meter ($\tau$=30s) under various ambient dose rate conditions. The size of NaI(Tl) detector is 2"$\phi$ × 2".

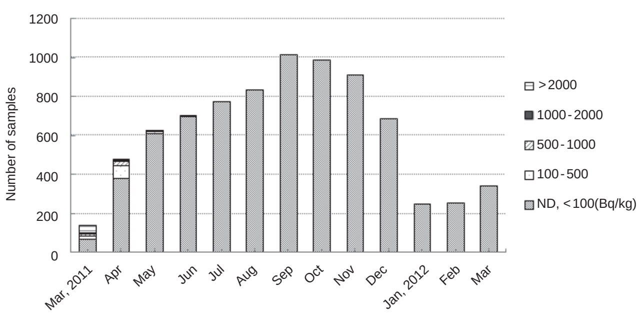


**Fig. 5.** The results of monitoring of vegetable produced at Fukushima in 2011fy.

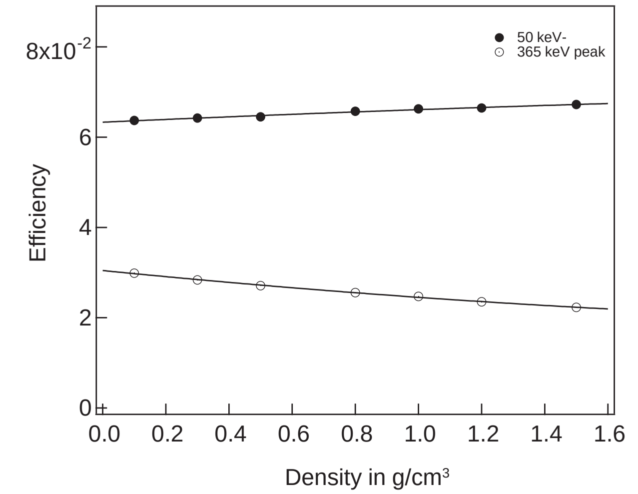


**Fig. 6.** Calculated peak efficiency (closed circle) and total efficiency (open circle) for $^{131}$I of a 2"$\phi$ × 2" NaI(Tl) scintillation detector for various densities of samples filled in a 1L Marinelli beaker. EGS5 was employed for the calculation[8].

## 3. Contribution of radio-cesium on the screening of radio-iodine

We should consider contribution of radio-cesium contained in the samples. As suggested from the observed spectrum shown in Figure 1, a considerable amount of $^{134}$Cs and $^{137}$Cs might be contained in this sample. As seen in Table 1, the provisional regulation limit for radio-cesium (500 Bq/kg) is much lower than the limit for radio-iodine (2000 Bq/kg) for vegetables and fishery products, and so radio-cesium exceeding the guideline value might be contained even in the samples that have passed the screening test for radio-iodine. To confirm the applicability of this method to radio-cesium screening, we also carried out type testing of a type 172B (Hitachi-Aloka Medical, Ltd.) meter. DL level for $^{137}$Cs calculated from the determined conversion factor (71Bq/kg/cps) and standard deviation of background count rates amounted to 750 Bq/kg in case of no radiation shield. If we consider $^{134}$Cs+$^{137}$Cs as the total radio-cesium, the DL value becomes somewhat smaller, since $^{134}$Cs emits multiple gamma-rays per one decay. Nevertheless it is very difficult to make screening of radio-cesium under this screening condition in any way. In order to resolve this difficulty, we also carried out the measurement after adding a radiation shield (5cm thickness lead). In this case DL could be reduced almost by quarter (180 Bq/kg). As a consequence if appropriate radiation shield could be employed, it is likely to become possible to make screening of 500 Bq/kg radio-cesium.

## 4. Effect of sample density

Figure 6 shows peak and total efficiencies for various densities of samples calculated using EGS5 Monte-Carlo simulation code. As the density of the sample becomes higher, self-shielding of gamma-rays within a large volume source increases, the peak efficiency becoming lower, for which we could make a correction along mathematical basis. Even when the correction is not applied to the result, we could avoid underestimation against the safe side for sample sources having the density below 1 g/cm$^3$, since most of reference sources for the efficiency calibration consist of material having around 1 g/cm$^3$ density (e.g. aqueous solution, agar, epoxy resin). Many of food samples have density below 1 g/cm$^3$, and overestimation might be ensured.

On the other hand total efficiency for integral count rate increases gradually as the sample density becomes higher. This situation might result underestimation for a food sample having much lower density below 1 g/cm$^3$. It should be noted that the positive bias of the results due to overestimation towards the safe side should be tolerable, but the negative bias due to underestimation against the safe side shall be avoided in any way in the screening purpose. In order to avoid underestimation due to the large difference between the densities of a sample and reference sources, correction of detection efficiency or safe side estimation should be recommended on the screening test of samples whose specific density is different from that of the reference source. However, effects in efficiency variation for integral count rate could be neglected as compared to that for the total peak, if the difference between the densities is small.

## 5. Conclusion

Rapid test method of radio-iodine isotopes applied to foods/foodstuffs inspection in Japan introduced just after the Fukushima Dai-ichi NPS accident was summarized. As a consequence rapid test method described in the "Radioactivity measurement manual for foods in emergency situation" specified by MHLW functioned effectively after the Fukushima accident as a screening tool. There are some problems, however, that should be considered based on our experience. A triage decision “Acceptable”, “Doubtful” or “Not Acceptable” could be made by use of the screening. In only the case that the samples are judged as “Doubtful”, additional more accurate measurement using a HP-Ge detector should be required to make sure these samples are acceptable or not. The number of samples to be measured with a HP-Ge detector can be reduced considerably by use of this rapid test. However since the result of this screening test is only for contamination with radio-iodine, even “Acceptable” samples might require additional test due to contamination with radio-cesium isotopes to avoid sanitation hazards of foods/foodstuffs and beverages in this situation. In spite the fact that this screening system intends to make sure samples are really “acceptable” without further measurements with a HP-Ge detector, additional evaluation or other test might be required for radio-cesium since DL for radio-cesium is more severe than for iodine. Thus further improvement of this method in efficiency should be studied with careful consideration of the contribution from radio-cesium isotopes. In addition effect of sample density also should be considered when the simple integral counting technique is adopted for samples whose specific density is much different from that of the reference source. MHLW also implemented the screening method for radio-cesium isotopes in foods/foodstuffs in Oct-2011[9]. This method might be useful for further improvements of the test method and the same or similar method also could be applicable to the screening of radio-iodine isotopes as one of improved methods.

Reliable, comparable and purpose-oriented data are essential for any public health decision based on radioactivity measurements internationally agreed standards of test methods are an important tool for the production of such measurement results[10]. Though there are some standards for environmental radioactivity measurement in normal situation, few standards for rapid radioactivity screening can be found. In an emergency situation, testing laboratories shall be requested to judge rapidly whether they are acceptable or not rather than the accurate measurements with a time-consuming technique. Just after an accident, maximum permitted levels ranged up to orders of magnitude higher than the level in normal situations. The international standards for the rapid screening test that could be applied in nuclear emergency situations and that could be implemented in emergency preparedness plans should be established based on the experiences after the Fukushima accident.

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