V. FUKUSHIMA SURVEY & RELATED RESEARCH

Preface to Special Section

Our Mission as Nuclear Physicists

On March 15th, 2011 at 10:00 am, high radiation level shown on the monitor prevented a radiation worker to leave the controlled area of the Nishina RI Beam Factory (RIBF), followed by many others who encountered the same problem. We soon realized that a high level of radioactivity did not originate from anything indoors but already existed outdoors (the radioactivity inspection is usually performed only at an exit). Ironically, the fallout from the Fukushima Dai-ichi Nuclear Power Plant made the controlled area of our accelerator facility the least contaminated place in the Wako campus of RIKEN.

For older generation, this situation wasn't new. Many years ago, atmospheric nuclear-bomb tests that took place overseas caused fallout to drift over to Japan that led to the similar radioactive situation observed in the Japanese accelerator facilities. This time, however, the fallout was due to the nuclear power plant disaster.

We gathered in the radiation proof control room of RIBF, and watched the monitoring posts to assess the situation. Then, we decided to upload the logged data to RIKEN's public website, and began taking several other measurements to determine the radioactive nuclear species of the fallout. While desperately hoping that the situation would not worsen, we convened special meetings every week to exchange latest knowledge and information.

The Nishina Center also functioned as the base to send radiation surveyors to Fukushima's affected area to measure the dosage of bodies, clothes, furniture, cars and any items that the refugees had with them. At the beginning, it was tough since no public transportation was available. Even in such dire situation however, it was very remarkable that many nuclear physicists from all over Japan joined to conduct this survey. Later as a follow-up, we joined the large-area soil survey initiated by the Research Center of Nuclear Physics of Osaka University and the Center of Nuclear Study of the University of Tokyo.

This special section of the Accelerator Progress Report records the works we were involved in 2011 in the aftermath of the Fukushima nuclear disaster. The Nishina Center is the largest Japanese research institute for nuclear physics, and our RIBF was built to study the nuclear synthesis at the beginning of the universe. Nuclear fuel is a gift from the nuclear synthesis, and atomic energy is the most important social application born from nuclear physics. As nuclear physicists, many of us feel that we could not escape from original sin for creating the world that led to the Fukushima nuclear accident. We have therefore done whatever we could as radiation workers with expertise in nuclear physics to deal with the aftermath of the disaster.

Yet what we were able to do and achieve were very limited in the face of nuclear crisis of such magnitude, we hope from the bottom of our hearts a swift recovery from the tragedy and that this kind of tragedy will never happen again.

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Diffusion of radioactive materials from Fukushima Daiichi Nuclear Power Plant detected by gamma-ray measurements on expressways†

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The accident at the Fukushima Daiichi Nuclear Power Plant caused by the enormous earthquake and subsequent tsunami on March 11, 2011, scattered a large amount of radionuclides in the Tohoku and Kanto areas. On the request of the Ministry of Education, Culture, Sports, Science and Technology, we visited Fukushima city on March 14, and we were asked by the Fukushima prefectural government to measure the dose rates along expressways in Fukushima prefecture.

By using an NaI(Tl) scintillation survey meter and a $LaBr_3$ γ-ray spectrometer, whose energy resolution is significantly superior to that of NaI(Tl), we measured γ-rays along the roads, shown as thick lines in Fig. 1, on March 15−17 and April 8.

As an example, the measured data on the Tohoku expressway are shown in Fig. 2. As indicated by the open squares, when we started measurements in Fukushima city at 1:33 p.m. on March 15, the dose rate was less than 0.1 μSv/h, which was the dose rate of the natural background. A radioactive plume that contained ^{133}Xe, ^{132}Te, ^{132}I, ^{131}I, ^{134}Cs, and ^{136}Cs was first observed at the Koriyama-Higashi interchange at 3:25 p.m. We measured dose rates in Iwaki city, and found them to be approximately 2 to 3 μSv/h. However, when we returned to Fukushima city at 8 p.m., we found the city to already be polluted by as much as approximately 10 μSv/h.

On March 16, the dose distributions in south Fukushima and the Aizu area were measured. The dose rate at Shirakawa interchange, which is the southern end of Fukushima, was 4.6 μSv/h at 2 p.m.; however, it was lower than the 1 μSv/h dose rate measured in southern Aizu. In Aizuwakamatsu city, the dose rate was approximately 1−2 μSv/h.

The dose distribution till the Ibaraki prefecture was measured on March 17. The dose rate in Fukushima prefecture was similar to that measured on March 15 and 16. The dose rate remained high outside Fukushima prefecture and was higher than 1 μSv/h up to the Yaita-Kita parking area in Tochigi prefecture.

The dose rate distribution was measured again on April 8 from Ibaraki prefecture to Miyagi prefecture, and it was found that the relative distribution had not changed. The attenuation of the absolute value was explained as the decay of radionuclides that were detected by the γ-ray spectroscopy. This implied that for the measured area, most of the radioactive material was deposited with the rain on March 15, that it did not move from the initial location, and that there was effectively no additional deposition of radioactive material after March 15.

Although the dose rate distributions of ^{132}Te, ^{132}I, ^{134}Cs, ^{136}Cs, and ^{137}Cs were similar, the distribution of ^{131}I was different from those of the others. The dominant nuclides in terms of the dose rate were ^{132}Te and ^{132}I for the March 15 to 17 measurements and ^{134}Cs and ^{137}Cs for the April 8 measurements.

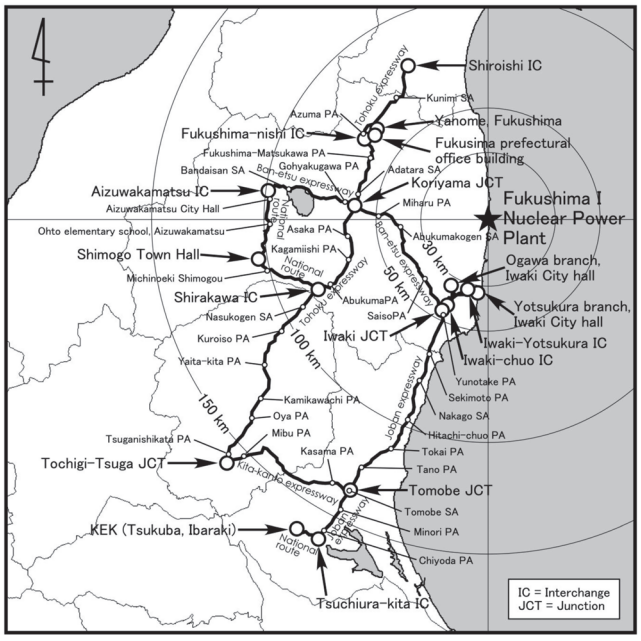

Fig. 1. Route map of the gamma-ray measurements.

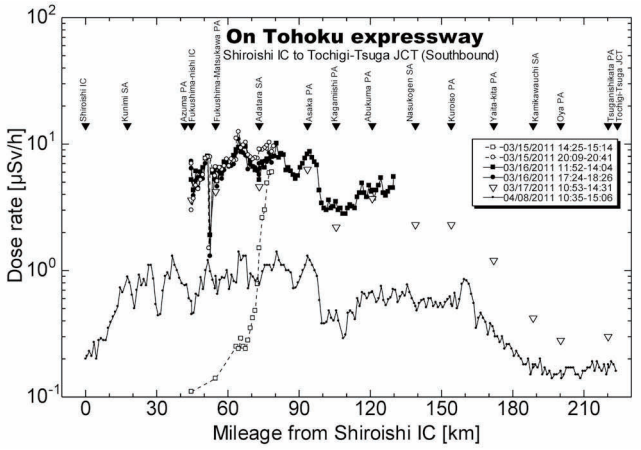

Fig. 2. Comparison of distributions of the dose rate measured along Tohoku expressway, from the complete measurement series.

† Condensed from the article in Transactions of the Atomic Energy Society of Japan **10**, 152 (2011), in Japanese.

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Radiation screening of evacuated people in Fukushima

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Immediately after the accidents of the Fukushima Daiichi nuclear power plant in March 2011, the experimental nuclear physics society organized a collaborative effort to aid evacuated people in Fukushima. The joint action was initiated by RNC together with RCNP, Osaka University, and was supported by the crisis management team of RIKEN and the MEXT. In the early stages, RNC served as a logistics base for the detachments from west Japan. Since public transportation to Fukushima was disrupted and gas supply was limited, we could only access Fukushima using automobiles with special permission from the police. The effort commenced on 21st March and was conducted continuously till May end and intermittently till early August. In total, RNC contributed 103 man-day to the effort, which was about 28% of the total contribution from the nuclear physics society team.

The team worked together with many other teams from different organizations such as universities and local autonomous bodies under the medical team of Fukushima prefectural government, which was supported by the Radiation Emergency Medical Assistance Team (REMAT) from National Institute for Radiological Science. All teams would assemble at the temporary Fukushima prefectural office in Fukushima city and were assigned the next day's tasks. The screening center for the evacuated people were scattered throughout the Fukushima prefecture, and the participants from RNC visited the centers in Koriyama-city, Kawamata-town, Yamakiya-chiku in Kawamata, Nakoso in Iwaki-city, and Tamura-city for the early emergency screening. Since late May, we have also contributed to the screening of residents within 20 km of the power plant who temporarily visited their home town. The base stations for the temporary visits were Bajikoen in Minamisouma-city, Hirono-town, and Hurumichi in Tamura-city.

In the end of April, the nuclear physics society team submitted a written proposal to the Fukushima prefectural government, outlining their suggestions on how best to conduct the screening activity.

Fig. 1. Screening of local citizens at Kawamata public health center on 24th March.

Many in the RNC staff were also very supportive of our efforts, in particular, the members of the administration office of RNC led by Mr. S. Asakawa. Some participants from RNC reported their experiences during the support work in Ref. 1, and in the following.

T. Fujinawa: I participated in the screening over three separated periods: late March to early April, May, and late July to early August. In late March, I had to drive a car from Wako to Fukushima to transport colleagues from Osaka and Kyushu. The highway was damaged by the earthquake, which made it difficult to drive a car. Access to Fukushima improved drastically once the Tohoku Shinkansen restarted operation on 29th April.

I participated in the emergency screening of evacuees at Big-palette in Koriyama but none of them exceeded the official reference level of 13 kcpm; however, I advised the residents to wash their hands and shoes, and gargle frequently, because many of them showed more than our standard level. In the early stages, the food supply at the hotel was limited and the temporary office would run short of copy paper. We brought many boxes of paper, batteries for survey meters, disposable body warmers, as well as our own food. The situation there changed with every visit I made. In August, only a few volunteers from our team remained, but much work had yet be done; I was even asked to stay longer. All of us hoped that the lives of those in the stricken area would soon return to normalcy.

H. Otsu: On March 28, I visited Kawamata public health center, where our team was conducting an emergency survey as well as thyroid gland screening for children. The radiation dose rate outside the building was 3 μSv/h, and ^{131}I had been found to be the main component. The radiation dose rate inside the building at 9:30, the beginning of the screening was 0.1 μSv/h and gradually increased as people entered and exited. About 500 people underwent surface screening and 230 children underwent thyroid gland screening. Almost the same number of people visited on March 29. Kawamata is located between Fukushima Dai'ichi nuclear power plant and Fukushima city. Hence, a variety of people, not only residents from towns near the power plant, such as Namie, but also prefecture officials, self-defense personnel, police, and radiation inspectors, passed through this screening center.

Surface survey were performed on the upper body, lower body, stomach, back, arms, legs; and soles of

shoes using a GM survey meter. If the count rate was more than 100 kcpm, we asked the self-defense personnel to wash the entire body, while if the count rate was more than 13 kcpm, we cleaned the contaminated part with a wet tissue or flowing water.

Even with high radiation doses outside, the surface of the cloths were not so contaminated. The count rates were 200∼500 cpm, almost equivalent to the room background in that area. High contamination was sometimes found on the soles of shoes. In one instance, the backs of a lady's shoes showed a count rate exceeding the reference level. We accompanied her while she washed them under flowing water, but this was in vain, because even after washing, the counting rate was not significantly reduced. We advised her to let us dispose of them. The self-defense office gave her a pair of sandals, which the Crocs company provided. Subsequently, we noticed that the adhesion of contaminants to polyvinyl acetate was much lower than that for other materials such as polyvinyl chloride.

In an exception, high count rate of 4 kcpm was detected on the upper body of a fire fighter in Namie town. He continued to work and provide water and treat the bodily wastes of people who had been evacuated from towns near Namie. He had to handle water hoses with his hands, and his upper jacket was contaminated. We advised him to take off the jacket outside the entrance of his house and not wear it indoors to reduce the contamination in residential spaces.

As mentioned above, one of the key points we focused on was the soles of shoes. We had the choice of recommending that they be disposed of. However, a girl of the same age as my daughter told me before being surveyed that her shoes were a birthday present from her grandmother. I held my breath while surveying her from top to her shoes. The count rate of her shoe soles was about 1 kcpm, much lower than the first reference level. I could just advise her mother to keep them outside the entrance and to wash hands every time when she came back to home.

During that time, I read a local newspaper every morning in order to gauge residents' mood as well. The anger of the people of Fukushima was evident there. However, although many would express their anger, there were also many who kept calm and tried to keep themselves informed about the radiation situation as much as possible. I admire their behavior.

M. Wada: An important task in the first weeks after the explosion was to investigate the radioactive iodine concentration in the thyroid gland, in particular for children. Together with other teams, we surveyed several hundred people and found that no one exceeded the official limit. The survey was performed in a relatively low background environment in the center with a NaI(Tl) scintillation survey meter. However, the background level was much higher than ordinary and the statistical treatment in the survey meter was not sufficient to avoid criticism afterwards. We could have taken more precautions during the measurements. For example, using a simple metal tube to cover the sides of the detector could have reduced the background rate significantly and the use of a simple scaler, which can count the number of pulses, could have afforded the results with much higher statistical accuracy.

In the later stages, we encountered few cases where the contamination of the body surface exceeded the reference levels, which coincided with increased demands to survey food, drinking water, soil in rice fields. However, screening of such materials was not an official mission and there were no suitable detectors for such purposes at the screening center. Occasionally, we received some samples and performed measurements at RIKEN with HPGe spectrometers. We found that the drinking water in the wells, including a well in Nagadoro-chiku of Iitate-mura which was known as a highly contaminated area, were clean from radioactive iodine or cesium at all. The soil in the rice filed was highly contaminated, but the water in the rice fields was also as clean as drinkable level. However, we did not have an official channel through which we could convey such information. We could only inform prefectural officers on the next visit. When the surfaces of vegetables were highly contaminated, we could detect using a GM survey meter, however, the sensitivity was much lower than the reference level of 500 Bq/kg. If we could have taught the residents a simple ashing method[2], they could have prepared the concentrated samples at home, which could have been easily tested at the screening center. Unfortunately, we were not aware of this simple and sensitive method at that time.

Overall, we did contribute some extent; however, we served more as radiation workers. As researchers, we could have worked towards finding better solutions to the problems and contributed in a more organized way.

Fig. 2. Screening of thyroid of local children at Yamakiya community center on 24th March.

References

1) H. Hasebe, J. Particle Accelerator Society of Japan, **8**, No. 3, (2011) 132-136.
2) M. Wada et al., Acc. Prog. Rep. 45 (2012).

Gamma-ray dose resulting from the accident at Fukushima Daiichi Nuclear Power Plant, as measured by the radiation monitoring posts in Wako campus

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A large heavy-ion accelerator complex, RIBF, and the Radioisotope Center are used in the RIKEN Wako campus, and the dose rates at the site boundary are continuously measured by six radiation monitoring posts, as shown in Fig. 1.

In March 2011, a magnitude-9.0 earthquake and a subsequent severe tsunami occurred, and huge amounts of radionuclides were released from the Fukushima Daiichi Nuclear Power Plant. Plumes containing radionuclides from the power plant passed over the Wako campus several times in March, and some amounts of radionuclides were deposited.

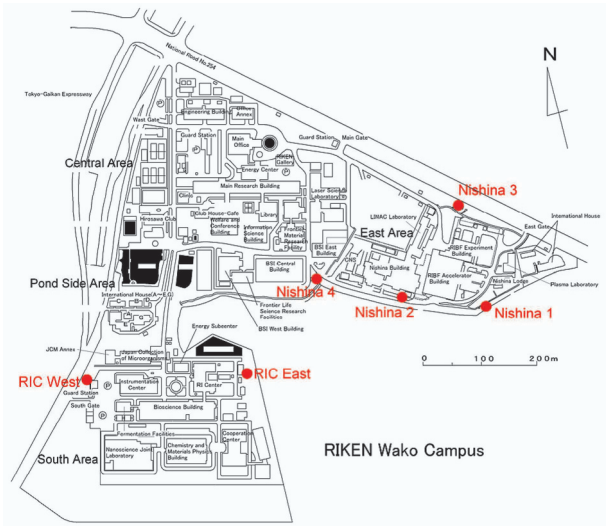

Fig. 1. Locations of radiation monitoring posts.

As shown in Fig. 2, a strong spike of dose rate was observed at 10 a.m. on March 15. This spike was mostly due to the γ-rays from ^{133}Xe gas[1], and the dose rate quickly decreased again because the radionuclide deposit on the ground was small. However, a second big plume came on March 21, and the rain on that day caused the deposition of ^{132}Te, ^{131}I, ^{134}Cs, and ^{137}Cs.[1] This deposit has been keeping the environmental dose rate high, as shown in Fig. 3.

In Fig. 2, small spikes occurring simultaneously with the rainfall are seen. These spikes are not due to the accident but due to the deposit of natural descendant radionuclides of uranium and thorium. Spikes are also seen in Fig. 3. The longer-term fluctuations in Fig. 3 are thought to be due to changes in the conditions around the monitors, that is, heavy rainfall, pileup of fallen leaves, etc.

The dose rate due to the accident was estimated to be 0.1 μSv/h on March 30. The contributions due to individual radionuclides were estimated on the basis of the radionuclide concentration in the soil: ^{132}Te, 22%; ^{131}I, 25%; ^{134}Cs, 38%; and ^{137}Cs, 15%. From the half-lives of these nuclides, the accumulated dose during a year from March 15, 2011, was estimated to be about 0.5 mSv. The internal dose due to the inhalation of the airborne radionuclides, which was measured by Haba *et al.*[2], was also calculated, and the total annual dose due to the accident was estimated to be 0.55 mSv.

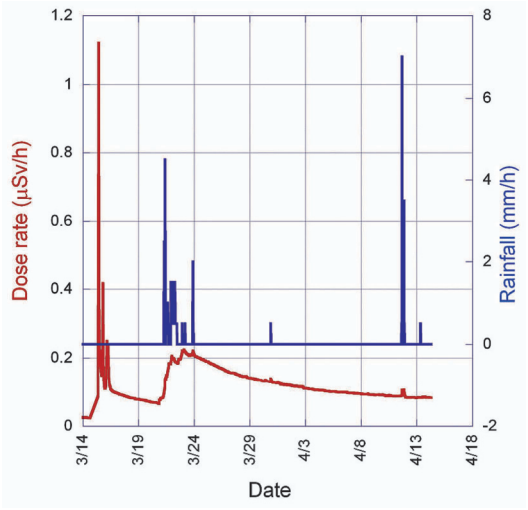

Fig. 2. Hourly averaged dose rate measured by RIC West post, along with the rainfall amount.

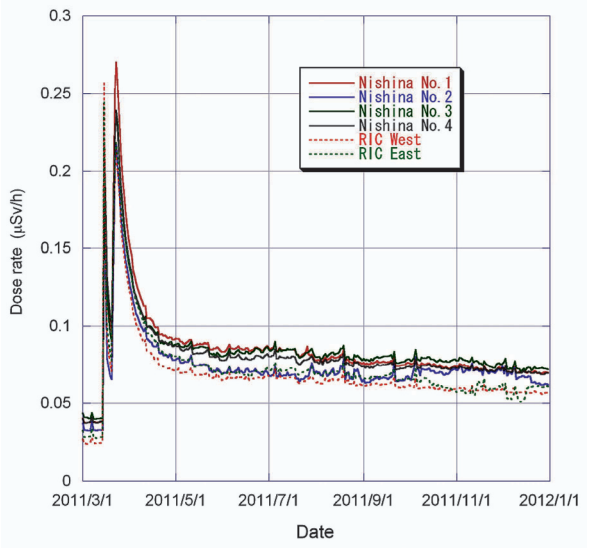

Fig. 3. Daily averaged dose rates measured at six posts.

References

1) H. Otsu *et al.*, elsewhere in this issue.
2) H. Haba *et al.*, elsewhere in this issue.

Measurement of radioactivity concentrations of airborne radionuclides in Wako after the Fukushima Dai-ichi nuclear power plant accident

H. Haba, J. Kanaya, H. Mukai, T. Kambara, and M. Kase

On Mar. 11, 2011, an earthquake of magnitude 9.0 occurred near the east coast of Honshu, Japan, and was followed by a large tsunami. The disasters caused damage to the Fukushima Dai-ichi nuclear power plant (FDNPP), resulting in the release of radionuclides into the environment. On Mar. 15, at a monitoring post of the RIKEN Wako Institute, located about 220 km to the southwest of FDNPP, we observed a rapid increase of dose rate from the usual 0.03 μSv h^{-1} at 4:00 (JST) to 1.2 μSv h^{-1} at 10:00. Hence, we initiated an urgent measurement of the radioactivity concentrations of airborne radionuclides.

Air dust was collected using a commercially available air dust sampler (M&F Enterprise SP-30) installed at the RIKEN Wako Institute (35°46'32" N, 139°37'04" E). The sampling flow rate was 30 L min^{-1}. A cellulose glass-fiber filter (ADVANTEC HE-40T) was used. No activated carbon filter was used; hence, our measurements were not sensitive to gaseous radioiodine. Dust was collected for 30 min for the first two samples in the period Mar. 15 11:15–11:45 and Mar. 16 13:15–13:45. After the third sample (Mar. 16 18:32–Mar. 17 9:00), dust was continuously collected for about one year, except for short interruptions due to the filter change. The filter samples were subjected to γ-ray spectrometry using a Ge detector. The detector efficiency was calibrated to an accuracy of 2%–10% using a multiple γ-ray standard source with the same size as the sample. A calibrated ^{134g}Cs source was also used to correct for the coincidence summing for ^{134g}Cs.

In this work, radionuclides of ^{140}La, ^{140}Ba, ^{137}Cs, ^{136g}Cs, ^{134g}Cs, ^{133g}I, ^{132g}I, ^{132}Te, ^{131}I, ^{131m}Te, ^{131g}Te, ^{129m}Te, ^{129g}Te, ^{110m}Ag, ^{99m}Tc, ^{99}Mo, and ^{95g}Nb were identified. Figure 1 shows a typical γ-ray spectrum of the sample collected in the period Mar. 29 9:40–Mar. 30 11:15. The peaks of volatile ^{131}I, ^{137}Cs, and ^{134g}Cs are very intense since they have a high release probability. In Fig. 2, the time

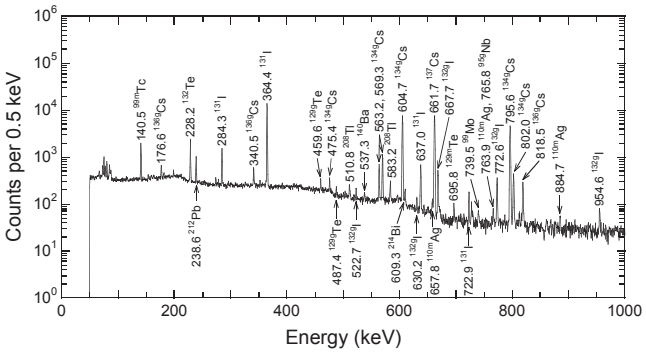

Fig. 1. The γ-ray spectrum for the air dust sample collected in the period Mar. 29 9:40–Mar. 30 11:15 (JST). The filter sample was measured for 35866 s, 1353 s after the end of the sampling.

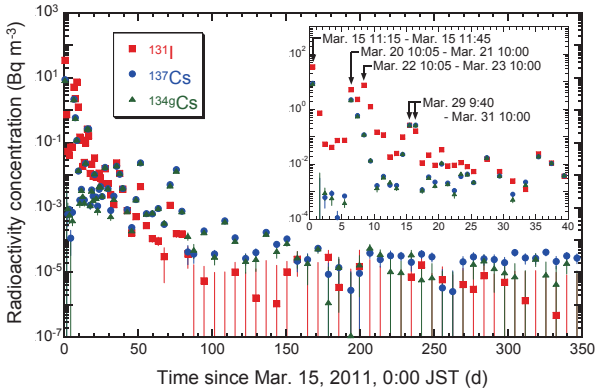

Fig. 2. Time variation of the activity concentrations of ^{131}I, ^{137}Cs, and ^{134g}Cs, as observed at the RIKEN Wako Institute. Inset shows the time variation for 0–40 days.

variations of the activity concentrations of ^{131}I, ^{137}Cs, and ^{134g}Cs are shown at the end of sampling as reference time. In the first sample, the maximum activity concentrations of ^{131}I, ^{137}Cs, and ^{134g}Cs were 35 ± 1, 8.8 ± 0.2, and 8.5 ± 0.2 Bq m^{-3}, respectively. Although the activity concentrations decrease with time, one can see two prominent peaks corresponding to Mar. 20 10:05–Mar. 21 10:00 and Mar. 29 9:40–Mar. 31 10:00 as indicated by arrows in Fig. 2. It is interesting to note that a distinct peak corresponding to Mar. 22 10:05–Mar. 23 10:00 is observed only for ^{131}I, indicating that the behavior of radioiodine is different from the behaviors of ^{137}Cs and ^{134g}Cs. These time variations are very similar to those measured at Japan Chemical Analysis Center, Chiba, located about 220 km to the south-southeast of FDNPP.[1] Amano et al.[1] measured the ratios of gaseous iodine to total iodine to be 0.52–0.71, using the cellulose glass-fiber filter in combination with an activated charcoal cartridge. Thus, the total activity concentrations of ^{131}I measured at the RIKEN Wako Institute are estimated to be 2.1–3.4 times those presented in Fig. 2. The activity concentrations of ^{137}Cs and ^{134g}Cs are almost equal for all samples. The ^{134g}Cs/^{137}Cs ratio is calculated to be 1.0 ± 0.1 for Mar. 15, and this value is consistent with other observations related to the FDNPP accident.[1]

We also determined the activity concentrations for other radionuclides such as ^{140}Ba, ^{136g}Cs, ^{133g}I, ^{132}Te, ^{129m}Te, ^{110m}Ag, ^{99}Mo, and ^{95g}Nb.[2] Our data, together with the measurements from other monitoring stations, will help provide further information on the characteristics of the fuel, release mechanism, and the transport of the radionuclides.

References

1) H. Amano et al.: J. Environ. Radioact. (in press).
2) H. Haba et al.: submitted to Geochemical Journal.

Environmental radioactivity at Wako from Fukushima Daiichi Nuclear Power Plant

H. Otsu, N. Aoi, [*2] H. Baba, R.J.Chen, H. En'yo, T. Fujinawa, S. Go, [*1] H. Hasebe, T. Hashimoto, [*1,*2] H. Watanabe, T. Ichihara, E. Ideguchi, [*1] T. Isobe, J. Lee, K. Ozeki, K. Kisamori, [*1] S. Kubono, [*1] M. Kurokawa, G. Lorusso, K. Miki, [*2] S. Michimasa, [*1] H. Miya, [*1] T. Motobayashi, S. Ota, [*1] P. Doornenbal, M. Sako, H. Sakurai, S. Sakaguchi, D. Steppenbeck, M. Takaki, [*1] M. Takechi, [*3] S. Takeuchi, A. Taketani, Y. Togano, [*3] H. Tokieda, [*1] Y. Wakabayashi, [*1] K. Yoshinaga, T. Kawahara, [*1] H. Imao, Y. Ito, and N. Sakamoto (fGamma collaboration)[*]

After the severe accident at the Fukushima Daiichi Nuclear Power Plant, we started monitoring the environmental radioactivity at Wako campus in order to detect a possible increase in the radioactive elements dispersed from the plant.

We set a germanium detector with volume of 146 cm^3 outside the RIBF building, next to the entrance facing the open space with a lawn. Gamma rays could be measured directly in the environment for 23 days, from March 15 to April 8. Initially, the data were collected by a multi-channel analyzer at 15-min intervals. On March 19, we installed an automatic data acquisition system that continuously processed the signals and generated histograms from the data every 15 minutes. Data accumulation continued with 24h a day except during the intervals when liquid nitrogen was supplied for cooling the germanium crystal and preamplifier.

Energy spectra obtained on March 24 and April 1 are shown in Fig. 1. Many of the observed peaks correspond to gamma-ray radiations, which would originate from the Fukushima Daiichi Nuclear Power Plant. Among them, ^{132}Te, ^{131}I, ^{132}I, ^{134}Cs, and ^{137}Cs isotopes are observed as prominent peaks. Gain shift and offset deviation in the detector or electronics caused by fluctuations in environmental parameters such as temperature or humidity were corrected by referring to each gamma ray peak.

The time dependence of the radiation is shown in Fig. 2. Before March 21 (6th day on the horizontal axis in the figure), the main contributions were from ^{131}I, ^{132}Te, and its daughter ^{132}I. March 21 and 22 were rainy. During these two days, the radiation from these four isotopes increased very significantly. However, after the 22nd, we did not observe such increases associated with the rain.

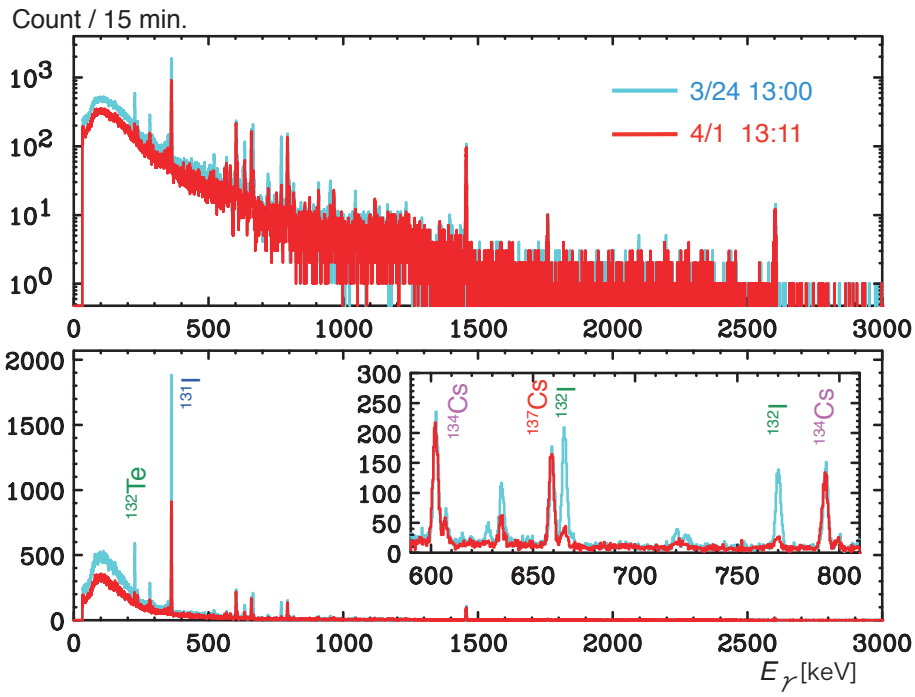

Fig. 1. Gamma-ray energy spectra obtained using the Ge detector on March 24 (cyan) and April 1 (red). Top frame shows energy spectra with logarithmic vertical axis, whereas bottom frame shows the same spectra with linear axis. Inset of the bottom frame shows spectra from 600 to 800 keV region.

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*3 Present affiliation: GSI

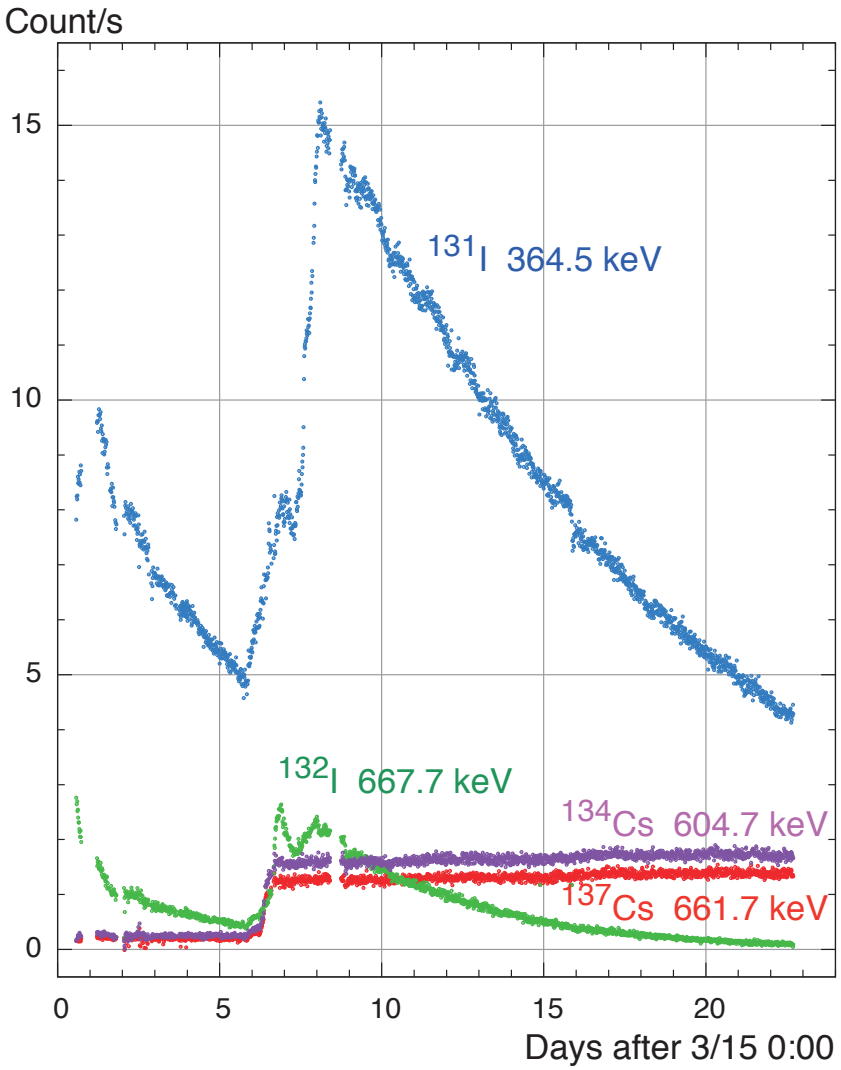

Fig. 2.
Time dependence of observed radiation.
Horizontal axis corresponds to days after March 15 at 0:00 A.M. and vertical axis indicates the counts per second.
March 21 and 22 were rainy. A drastic difference of trends occurred before and after these days. Before March 21 (6th day on the horizontal axis in the figure) the main contributions were from ^{131}I, ^{132}Te, and its daughter ^{132}I. During rain, the radiation from the isotopes indicated increased very significantly.
However, we did not observe such increases associated with rain after the 22nd. After the rains, iodine isotopes decreased with their decay half lives or on the same order. Cesium isotopes did not decrease in this period. In fact, they seemed to increase slightly.

These tendencies indicate that most of the radioactive elements in the air were fell down to the ground by the rain around the 21st. Subsequently the cesium 134 and 137 isotopes exhibited a small but continuous increase during the measurement because of the further deposition of these isotopes.

In order to evaluate the amount of radioactive elements accumulated on the ground, we performed a Monte Carlo simulation with the GEANT4[1] toolkit by taking into account the area (ΔS) corresponding to the scoping angular acceptance, detection efficiency ($\varepsilon(E_\gamma)$), and absorption effects ($f(E_\gamma \boldsymbol{r})$) by air and other materials. We obtained the coupled parameters $f\varepsilon\Delta S$ as:

$$f\epsilon\Delta S = \epsilon(E_\gamma)\int_S f(E_\gamma,\vec{r})\frac{dS}{\cos(\theta)}.$$

The obtained values were 6.2×10^{-4} and 5.0×10^{-4} [m^2] at 364.5 keV and 660 keV, respectively. By taking the branching ratios into account, we evaluated the surface density of radioisotopes on the grass in the Wako Campus as 26 kBq/m^2 for ^{131}I and 2.8~3.0 kBq/m^2 for each of the other three isotopes on March 25. In these analyses, we assumed for simplicity that all radioactive elements remained on the ground surface and did not penetrate or sank into the soil. Further analyses are needed for quantitative estimation, especially for the consideration of depth distribution inside the soil. Systematic errors are estimated to be about 20%, mainly due to ambiguity in the simplified model employed for the simulation.

In summary, we measured the environmental radioactivity in the Wako area in the period following the Fukushima Daiichi Nuclear Power Plant accident and evaluated the results.

References

1) http://geant4.cern.ch/

Radioactive Xe arriving in and passing through Wako area

H. Otsu, fGamma collaboration *1

Immediately after the severe accident that occurred at the Fukushima Daiichi Nuclear Power Plant, we started the spatial radiation dose measurement in Wako campus[1]. In this article, we report on the evidence of gaseous xenon isotopes arriving in and passing over the Wako area. The major components were ^{133}Xe, ^{133m}Xe, and ^{135}Xe, as identified by our Ge detector measurements. Experimental details can be found in Ref. 1 of this volume.

From March 15 to 16, abrupt changes in the environmental dose were observed at three instances[2] by area monitors at the RIKEN Nishina center as burst-like phenomena. The equipment was not ready for the first and second abrupt changes. In the early morning on the 16th, the third abrupt change occurred, although the scale of the burst was smaller than those of the previous two events. At that occasion, we successfully observed gamma-ray peaks originating from the burst.

Figure 1 shows gamma-ray energy spectra recorded between 5:09 and 8:50. Representative spectra are shown in red, with a maximum at 6:15, and in cyan, as passing over the period at 8:39. The spectrum at 13:50 on the 15th is also shown by solid green line for comparison. Magnitudes of the three peaks at 81, 233, and 250 keV had already increased at 5:09. They rapidly reduced at 8:50 while those of other peaks stayed almost in constant during these 4 hours. From gamma-ray energies of 81, 233, and 250 keV,

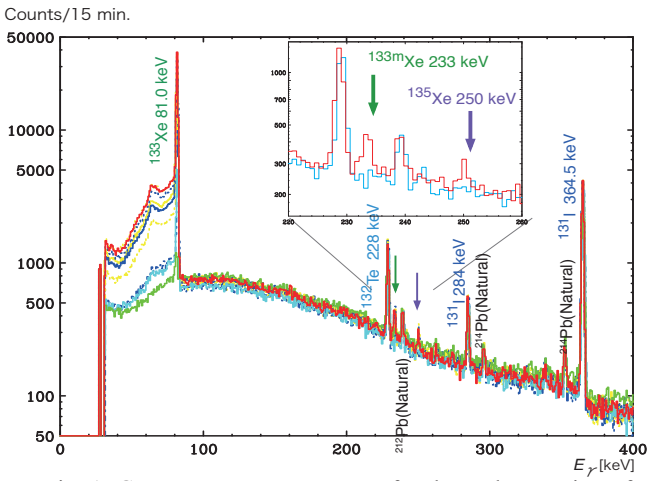

Fig. 1. Gamma-ray energy spectra for the early morning of the 16th of March with green indicating a reference, which corresponds to the spectrum at 13:50 on March 15. Horizontal axis indicates gamma-ray energy. Vertical axis shows detection counts within 15 minutes. In the inset, spectra are enlarged for the range between 220 and 260 keV. Here, data are limited for 6:15 (Red), 8:39 (Cyan), and reference (Green). Peaks related to ^{133}Xe, ^{133m}Xe, and ^{135}Xe were observed during the burst.

*1 Gamma collaboration members are listed in the previous report in this volume.

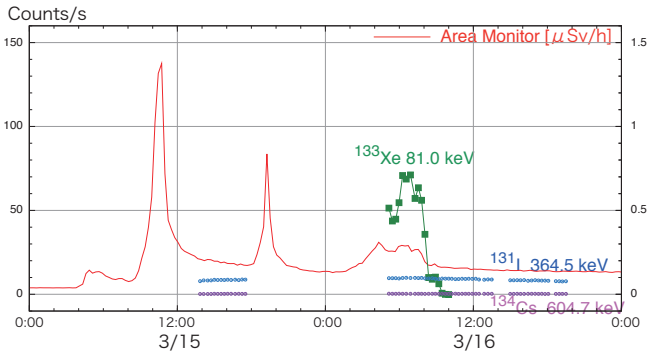

Fig. 2. Time dependence of Xe 81.0 keV peaks (left vertical axis) with dose read out by area monitor in μSv/h unit (right vertical axis). The 81 keV peak is not very prominent during daytime on the 15th.

we identified ^{133}Xe, ^{133m}Xe, and ^{135}Xe, respectively. Other possibilities for the origin of the 81 keV peak, for example, 80 keV from ^{131}I, are rejected because no lines corresponding to their branches were observed. The bump-like structure below the 81 keV peak is interpreted as corresponding to the Compton scattering component.

Figure 2 shows the time dependence of the 81 keV peak in counts per second and the radiation dose measured by the area monitor in μSv/h unit for comparison. The ^{133}Xe yield varied on the morning of the 16th in a manner similar to the area monitor dose, whereas the peak yields for ^{131}I and ^{134}Cs remained almost constant.

These comparisons indicate that rapid increases in the dose were caused by gaseous Xe activities. Another observation is the more rapid decrease of the yield than any of the lifetime of the Xe isotopes: 5.25 d for ^{133}Xe, 2.19 d for ^{133m}Xe, and 9.14 h for ^{135}Xe. This suggests that gaseous Xe passed through the area as a cloud without causing a large amount of activity accumulation on the ground.

The efficiency of the germanium detector with the same geometry (56.8 mmφ × 61.7 mm) was calibrated using several checking sources with a facially distribution. Detection efficiency at 81 keV was calculated with the extrapolation as 0.11. From these properties og the detector, we estimate the gamma-radiation flux density as 2.5×10^5 [$m^{-2}s^{-1}$] at the observed burst maximum (70 Counts/s).

In summary, we observed a rapid increase and decrease in radiation on March 16 at the Wako campus. We conclude from our gamma-ray measurements that they originate from radioactive Xe gases. The Xe isotopes were considered to have passed over the Wako area.

References

1) H. Otsu, fGamma collaboration, this volume.
2) Y. Uwamino, this volume, private communication.

Measurement of ^{137}Cs and ^{90}Sr in a Surface Soil Sample Obtained from Namie, Fukushima

Y. Kobayashi, Y. Saito[*1], M. Iwasaki, and R. Hayano[*2]

An enormous quantity of the radionuclides has been discharged owing to the hydrogen explosion of the nuclear reactor in the Fukushima Daiichi Nuclear Power Plant in March 2011. We performed γ-ray spectroscopy for detecting ^{137}Cs and the liquid scintillation technique for detecting ^{90}Sr with the rapid chemical separation of the soil obtained from Namie, Fukushima Pref., to evaluate the distribution and deposition of the radioactive fallout from the accident[1]. A soil sample with 2-cm thickness was collected from the surface and from an area of 200 cm^2 in Namie on April 29, 2011. The sampling point, at the latitude 37º 35' 34.77" North and the longitude 140º 45' 12.85" East, is located approximately 30 km northwest from the site of the explosion. The spatial dose rate was 25~30 μSv/h at the sampling time. The total wet weight of the sample was 19.8 g.

The complete sample was placed in a plastic bottle for the γ-ray spectrometry that was performed using a Ge detector. The γ-ray spectrum of the soil sample obtained from Namie is shown in Fig. 1. Most γ-peaks could be assigned to long-lived radioactive ^{137}Cs ($T_{1/2}$ = 30.1 y) and ^{134}Cs ($T_{1/2}$ = 2.06 y). The concentration of ^{137}Cs was estimated from the peak area of a typical 661.9-keV γ-ray. The activity of ^{137}Cs was estimated to be 63,400 Bq/kg (wet soil) by a calculation performed using the counting rate of 20.4 cps, the counting efficiency of the Ge detector, and the branching ratio of the 661.9 keV γ-ray.

^{90}Sr can achieve a secular equilibrium with the high-energy β-emitter ^{90}Y in a few weeks. ^{90}Sr concentration was determined by means of the low-background liquid scintillation technique with rapid chemical separation through oxalate coprecipitation[2]. The soil was heat-treated at 450 ºC for 24 h and dissolved in aqua regia. Then, the Y and Sr carrier solution was added to it. ^{90}Y was extracted from bis(2-ethyl-hexyl) phosphoric acid (HDEHP) and toluene solution after the radioactive equilibrium of ^{90}Sr/^{90}Y was reached. Yttrium was separated as a hydroxide, and then, oxalate salt was precipitated with the condition for which the pH was ~1.5. The chemical yield was estimated to be 70%. The sample for the β-ray measurement was prepared by adding a liquid scintillator (Perkin Elmer Ultima Gold AB). The measurement was performed in a Packard low-background detector. Figure 2 shows the β-spectra of a soil sample prepared by rapid chemical separation, a blank test, and a hydroxide as reference including ^{90}Sr/^{90}Y with 160 Bq activity. The end point (E_{max} = 2.2 MeV for ^{90}Y) could not be detected in the spectrum of the soil sample, and the line shape in its case differed visibly from that of the reference precipitation. It was concluded that the activity of ^{90}Sr in the soil sample was below the detection limit. The detection limit of the present method was estimated to be 35 Bq/kg from the counting efficiency and counting rate of the reference sample. The ^{90}Sr/^{137}Cs ratio caluculated in this study was consistent with the analytical report published by MEXT on May 31, 2011.

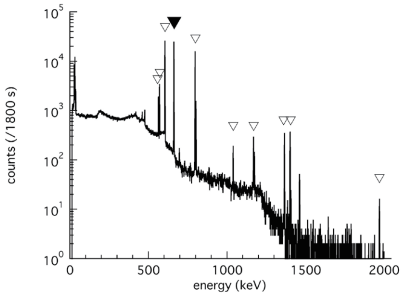

Fig. 1. γ-ray spectrum of the soil sample obtained from Namie, Fukushima. ▼ and ▽ indicate the 661.9-keV γ peak of ^{137}Cs and the γ-rays of ^{134}Cs, respectively. The measuring time was 1800 s.

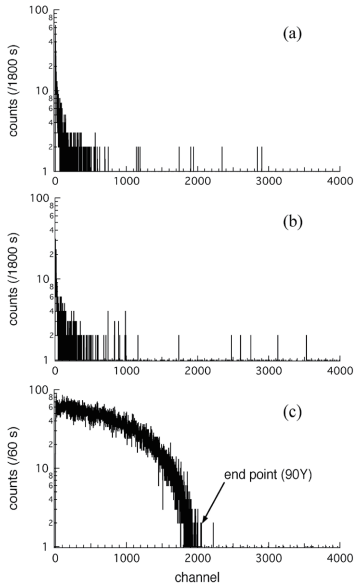

Fig. 2. β-ray spectra of (a) a oxalate salt extracted after chemical separation, (b) the blank test, and (c) the reference which was a precipitation of $Y(OH)_3$ obtained using a ^{90}Sr/^{90}Y tracer with 160 Bq activity.

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*2 Department of Physics, University of Tokyo

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1) *http://www.radiochem.org/kinkyu/index.html*
2) *http://www.kankyo-hosyano.go.jp/series/lib/No2.pdf*
3) *http://radioactivity.mext.go.jp/ja*

Measurement of β-ray emitting isotopes from the Fukushima Daiichi nuclear power plant accident

F. Sakuma, M. Tokuda[*1], Y. Sada[*2] and T. Hashimoto[*3]

[Fukushima reactor accident, β-ray isotopes]

The Fukushima Daiichi nuclear power plant accident, caused by the earthquake that occurred on March 11, 2011, resulted in the release of large amounts of radioactive materials into the atmosphere. Emissions from γ-ray emitting isotopes such as ^{137}Cs and ^{131}I were observed and reported immediately after the accident. However, the isotopes that emitted only β-rays, such as ^{90}Sr, were not detected owing to the difficulty in the measurement of β-rays. As it is known that the ^{90}Sr/^{137}Cs activity ratio was observed to be up to 1.5 at the Chernobyl accident[1], we attempted to measure the β-ray emitters using plastic scintillators and photomultiplier tubes (PMTs) in March and April 2011. The experimental setup is shown in Fig. 1. We wiped the roof of a car ($\sim$1.5 m^2) with a cleaning tissue (Nippon Paper Crecia, Kimwipe S-200) on March 23 at around 17:00: the car was parked in the RIKEN Wako campus from before the earthquake to the time of wiping. In order to measure the spectra of β-rays with a few MeV of energy, a thin (t1 mm, PMT1) and a thick (t50 mm, PMT2) plastic scintillator were used. A sample was set on the thin scintillator, and an event trigger was generated by a coincidence signal of the PMT1 and the PMT2. The ADC spectrum of the β-rays was obtained using the thick scintillator. In addition, two large plastic scintillators (PMT3 and PMT4) that covered the complete system were used as cosmic-ray veto counters.

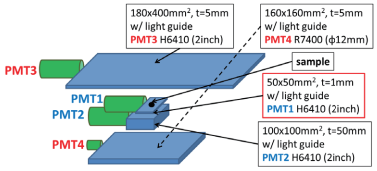

Fig. 1. Schematic drawing of the setup.

The ADC spectra obtained from the sample are shown in Fig. 2; these spectra were obtained with the same setup but on a different days: April 1, 21:30 to April 2, 4:36, and April 20, 16:22 to April 21, 16:26. Each ADC spectrum was fitted with the background cosmic-ray spectrum and referenced ^{90}Sr spectrum in the ADC range between 500 ch and 800 ch. The cosmic-ray and ^{90}Sr spectra were obtained with an empty sample and ^{90}Sr source of $\sim$0.9 MBq, respectively. The endpoint energy of ^{90}Sr is 2.280 MeV, and its half-life is 28.79 y, i.e., $^{90}_{38}\mathrm{Sr} \rightarrow (^{\beta^-0.5459\mathrm{MeV}}_{28.79\mathrm{y}}) \rightarrow {}^{90}_{39}\mathrm{Y} \rightarrow (^{\beta^-2.280\mathrm{MeV}}_{64.053\mathrm{h}}) \rightarrow {}^{90}_{40}\mathrm{Zr}$. In the figures, the spectrum shape can be seen to obviously change, i.e., the count rate in the range over 500 ch decreased with time. As the half-life of ^{90}Sr is very long, this decreasing radioactivity indicates that we could not have observed the β-rays emitted by ^{90}Sr. In this case, what isotope had been observed that emitted β-rays similar to ^{90}Sr? Figure 3 shows the count rate in the ADC range between 500 ch and 800 ch after subtracting the background, in which each data point is obtained on a different day and the start time corresponds to April 1, 21:30. The fitting result of a half-life is 3.20 $\pm$ 0.23 d, which is consistent with the half life of ^{132}Te, i.e., $^{132}\mathrm{Te} \rightarrow (^{\beta^-2.137\mathrm{MeV}}_{3.20\mathrm{d}}) \rightarrow {}^{132}\mathrm{I}$. In fact, a radiation monitor group in KEK reported the observation of ^{132}Te in March 2011[2].

In conclusion, we could not observe β-rays emitted by ^{90}Sr, although we observed a β-ray emitting isotope, ^{132}Te, which was released from the Fukushima reactor after the accident in March 2011.

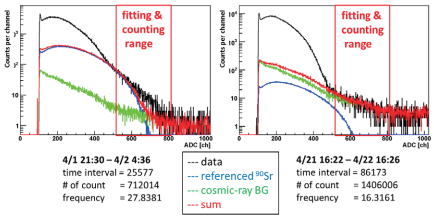

Fig. 2. ADC spectra obtained from the sample. (see text)

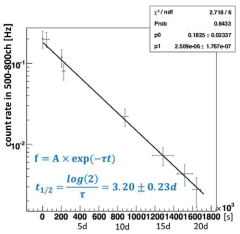

Fig. 3. Count rate in the ADC range between 500 ch and 800 ch after subtracting the background.

References

1) Environmental Consequences of the Chernobyl Accident and their Remediation (IAEA, 2006)
2) http://www.kek.jp/quake/radmonitor/

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Pilot survey of feral cattle in the vicinity of Fukushima Daiichi Nuclear power plant

M. Wada, H. Haba, S. Sato,[*1] S. Roh,[*1] and M. Toyomizu[*1]

A pilot survey of radioactive contamination of feral cattle in Minamisoma city was performed in late August 2011. After the accident of Fukushima Daiichi nuclear power plant, many cattle left their enclosures and survived by consuming plants and water in a ditch that may have been radioactively contaminated. Therefore, it is worth surveying the radioactive contamination in the muscles, organs, blood, urine, and feces of cattle before slaughter disposition.

Increasing internal radiation dose in human body because of contaminated food became a sensitive problem especially after contaminated beef was found in the market. The beaf cattle consumed contaminated straw immediately before being butchered, and the meat contained more than 2 kBq/kg of radioactive cesium. The transfer rate of cesium from the contaminated food to the body and the metabolism rate have been previously studied[1,2]. If the specific radioactivities in cattle muscle could be evaluated from that in the blood or urine when the cattle are alive, it might aid in assessing the use of those cattle.

In the first sampling, we evaluated 5 cows captured at Long. 140.929E, Lat. 37.572N, about 19 km NNW from the nuclear power plant. Samples of blood, urine, feces, and amniotic fluid were taken in 20 cc vial bottles, and samples of muscles and other organs were taken in 100 cc bottles at the location. The 5 cows that were evaluated inhabited a trench with abundant availability of water. We observed that the urine color of these cows was clearer than that of cows in a stock farm. One cow was pregnant and we sampled both the fetus and amnionic fluid.

The radioactivity levels of samples were measured using a well shielded HPGe detector with a relative efficiency of 20%. The absolute efficiency for each sample bottle was calibrated using a mixed standard source in a water matrix. The sum correction factors for the γ-rays from ^{134}Cs were evaluated with a sample itself but placed at 10 cm apart from the detector.

The specific activities of the sum of ^{134,137}Cs were plotted for individual cattle (Fig. 1). Cs activity did not show a uniform distribution even within the muscles, and the internal organs showed much lower radioactivity level than that in the muscles. The specific activities were also plotted as a function of the blood activity (Fig. 2). Positive correlations were found, for example, ratios of the fillet and tongue to the blood were approximately 33 and 12, respectively. Since evaluation of the total amount of urine per day was difficult for such feral cattle, we could not use urine samples for quantitative evaluation of whole body activities. In a liver sample, some amounts of ^{110m}Ag 148(5)Bq/kg) and ^{129m}Te 20(1) Bq/kg) were found. These radionuclides were also detected in the soil of the area inhabited by the cattle; further, we speculated that the cattle consumed contaminated soil when drinking the water in the ditch, which resulted in the heavy metals to concentrate in the liver.

Although we planned to continue the survey, because of an organization issue, only the first pilot survey was conducted, and the number of samples was limited because of the allowed sampling time at the location. However, we consider our limited data worth reporting, because our data may be used in combination with other surveys.

References

1) J.E. Johnson, et al.: J. of Nutrition, **94**, 282 (1968).
2) G. Voigt et al.: Science of Total Environment, **85**, 329 (1989). S. Ambe et al.: Chem. Lett. **2001**, 149.

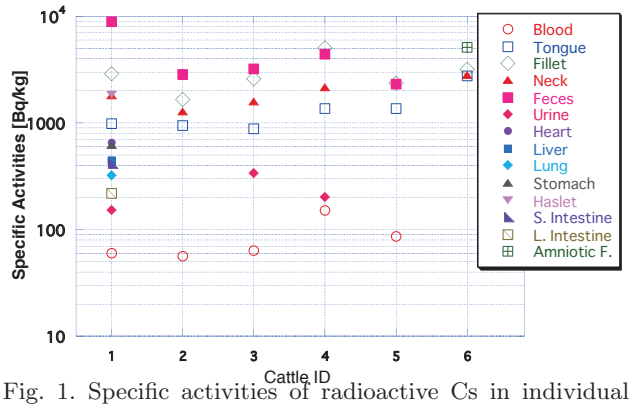

Fig. 1. Specific activities of radioactive Cs in individual cattle samples. #6 is the fetus of #5.

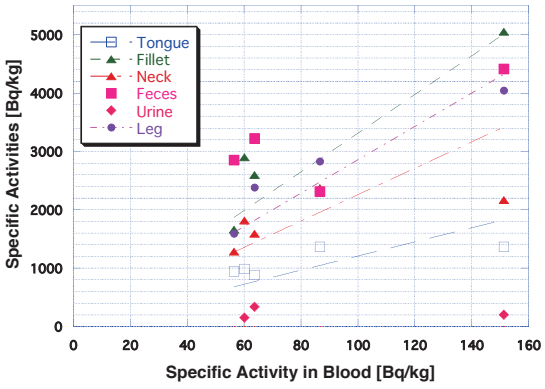

Fig. 2. Specific activities of radioactive Cs as functions of the specific activity of blood.

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Gamma ray spectroscopic analysis of underwater environmental samples from ocean near Fukushima Daiichi Nuclear Power Plant

S. Moriya and H. Otsu

Radioactive materials have been released into the nearby ocean from Fukushima Daiichi nuclear power plant after severe accidents in March, 2011. Here, we discuss a series of analyses that we performed to access the incorporation of such radioactivity into members of the coastal eco-system.

Oceanic biomasses and environmental samples were collected at the Fishery port of Hisanohama and the surrounding coastal regions (figure 1). Sample collection was done through self contained underwater breathing apparatus (SCUBA) diving. The SCUBA sampling was done in collaboration with the Japanese Association of Underwater Science. We collected 10 litter of sea water and filtrated it using a 0.22 μm Omuni pore filter (Millipore) until the filter was clogged, after which we obtained microbial biomass. The filtrated water was then evaporated to obtain sea salts. We also collected biofilm materials from the surfaces of rocks and macro benthos (Ascidiacea, Sponge, Sea urchin, Starfish and Shellfish) from seabed.

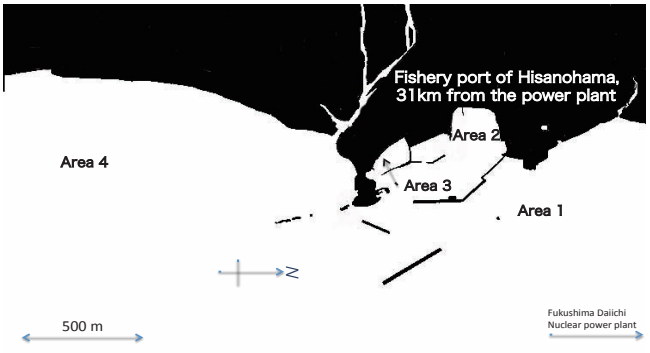

Fig.1 : Sampling spots in Hisanohama.

Biofilm on the rock was collected using a wire brush with distilled water. The samples of biofilm and macro benthos were dried at 120 ℃ in a drying oven, then milled using a food mixer. The obtained samples were analyzed with gamma ray spectroscopy.

Results of the gamma ray spectroscopic analysis using a Germanium detector are shown in table 1. According to these results, the environmental water and primary producers (oceanic microbes and biofilms) contain ^{134}Cs and ^{137}Cs. However, the macro benthos on the sea floor contain not only radioactive Cs but ^{110m}Ag in their body.

To estimate the source of ^{110m}Ag in such the organisms, we performed additional sampling to obtain bottom mud using a short piston core sampler operated by the SCUBA divers. The core sampler is assembled with an end cap and a PVC tube of 40 mm diameter and approximately 25 cm length. The piston of the core sampler is made from craft resin with the same PVC tube as that in the template (40 mm diameter and approx. 5 cm length) and is inserted into

Table 1. Detected radioactive elements from each sample. Corrections are needed for ^{134}Cs and ^{110m}Ag for sum effects. Error(RMS) indicates statistical only.

		Radiation [Bq/kg(Dry)], or [Bq/m^3] for Microbes, evaluated on 2012/03/11					
Area	Material	^{134}Cs	Error	^{137}Cs	Error	^{110m}Ag	Error
1	Sea salt	4.4	0.2	6.1	0.3	–	
2011/12/10	Microbes	15	3	15	3	–	
Lat. 37.151N	Biofilm	874	36	1431	53	–	
Long. 41.005E	Sponge	221	4	381	7	39	2
	Ascidiacea	181	2	293	3	128	2
	Star fish	7.2	0.4	10.1	0.6	29	1
	Sea Urchin	110	17	205	29	125	29
2	Sea salt	4.0	0.5	6.1	0.6	–	
2011/11/20	Microbes	109	9	176	9	–	
Lat. 37.150N	Biofilm	611	45	926	47	–	
Long. 41.001E	Sponge	245	4	422	5	4.6	1.1
	Ascidiacea	134	2	215	3	82	2
	Sea Urchin	101	1	156	2	81	6
3	Sea salt	2.7	0.2	4.7	0.2	–	
2011/12/3	Microbes	31	2	51	3	–	
Lat. 37.147N	Biofilm	1416	67	2343	85	–	
Long. 41.002E	Ascidiacea	295	5	465	2	64	1
	Shell fish	107	1	160	1	45	1
4	Sea salt	2.7	0.1	4.6	0.2	–	
2011/11/26	Microbes	6.9	1.3	9.1	3.8	–	
Lat. 37.136N	Biofilm	284	20	427	25	–	
Long. 41.003E	Sponge	260	5	431	6	131	12
	Shell fish	95	1	146	1	34	1
	Sea Urchin	30	1	47	1	10.8	0.5
2	Mud(Surface)	819	11	1341	9	13	1
2011/12/17	Mud(Bottom)	500	9	819	8	–	

the PVC tube during the operation. Core samples from the depth of 15-20 cm have been analyzed by sectioning depth of 5 cm each by using the Ge detector. The results (Table 1) show that the materials on surface of the seabed contain ^{110m}Ag and both of $^{134, 137}$Cs but bottom mud from deeper position contain only radioactive Cs.

These results suggest that the once-precipitated radioactive Ag is possibly re-suspended into the oceanic water by the swell of the ocean and is fed by macro benthos. The mechanism for ^{110m}Ag precipitation is still unclear. Further analyses of the seawater contents by fine fractionation and its relationship with land-originated compounds are needed to understand the entry of ^{110m}Ag into the coastal eco-system.

Note that the gamma ray analyses are ongoing especially on the correction of the sum peaks from ^{134}Cs and ^{110m}Ag because of the relative geometry between a sample and the Ge detector. These are planned to be corrected using the GEANT4 code by considering the sample height information.

In summary, we determined the radioactivity absorbed by oceanic plants and animals not only for Cs isotopes but also for Ag isotopes released from the power plant accident. These results will be utilized to understand the future eco-system situation around these areas.

Analysis of Gamma-ray Emitters in Soils for Obtaining Maps of Deposited Radioactive Substances around Fukushima Daiichi Nuclear Power Plant

S. Shimoura*1

In order to prepare maps of radioactive substances deposited on the ground surface, soil samples from the 5-cm surface layer were collected for about 2,200 locations within approximately 100 km from the Fukushima Daiichi Nuclear Power Plant (NPP) from June 6 to 14 and from June 27 to July 8, 2011. About 10,000 samples in total, corresponding to five or three different points for each location, were analyzed by the Japan Chemical Analysis Center (JCAC) (40%) and by a collaboration (60%) among universities and institutes, including Center for Nuclear Study (CNS), RIKEN Nishina Center (RNC), Research Center for Nuclear Physics (RCNP), and other organizations. CNS acted as a hub of the collaboration, where tasks such as receiving and checking about 6000 samples from Fukushima; sending them to 22 analysis groups depending on their capacities; and collecting, checking, and summarizing the results were performed. About 120 samples, collected from within 20 km of NPP, were analyzed by CNS and RNC.

The activities of Cs-134, Cs-137, and I-131 were analyzed by measuring γ-rays emitted by all the samples using the germanium detectors of each analysis group. Activities of other substances such as Te-129m, Ag-110m were also analyzed for about 2000 samples. Each analysis group calibrated the absolute magnitudes of the activities by using the IAEA-444 standard samples[1] filled in the same U8 type containers ($5\ \mathrm{cm}^{\phi} \times 5\ \mathrm{cm}^{H}$) as the soil samples that were circulated among the collaboration. Since Cs-134 and Cs-137 are involved in the IAEA-444 standard, their absolute activities were determined by the relative yields from the IAEA-444 sample and each soil sample.

Measurement time for a sample was basically one hour, whereas long runs with around 10 hours were also performed for some samples to measure the radiation from I-131. However, large portion of the samples have no significant value for I-131 because of decay out through more than 10 half lives and of Compton backgrounds from the radiation of Cs-134 and Cs-137.

The same $\sim$300 samples were analyzed by multiple groups, one of which is either CNS or JCAC, for the purpose of a cross checking. Results from these different groups were consistent within 30% for each sample and within 15% for an average of five samples for each location. The deviation is considered to be due to the inhomogeneity of the activities in soil samples, since it was found that for some samples, counting rates depended on the direction of the sample relative to the detector.

The maps based on the resultant activities (Cs-137, Cs-134, I-131, Te-129m, and Ag-110m) are found from the Web pages of MEXT[2], JAEA[3], and RCNP.[4]

As shown in ref.[2], the ratios of the activities of Cs-134 to Cs-137 were independent of the locations about 0.9–1.0 . On the other hand, activity ratios of I-131 to Cs-137 of the soils in the southern coastal areas are about 4 times larger than those in the northern areas. The present results are expected to be used as a basis for assessing the radiation dose in the concerned areas, further investigations in the environmental research works, and other such purposes.

The measurements at RNC and CNS were performed by H. En'yo, Y. Watanabe, S. Yokkaichi, H. Watanabe, M. Nishimura, T. Kishida, H. Otsu, H. Ohnishi, T. Tada, K. Ozeki, K. Morita, D. Kaji, H. Haba, J. Kanaya, E. Ikezawa, T. Maeyama, M. Nabase, K. Ikegami, A. Yoshida, T. Hori, Y. Yamaguchi, S. Sonoda, Y. Ito, Y. Uwamino, H. Mukai, S. Fujita, M. Fujinawa, H. Sakata, T. Kambara, K. Yamada, T. Otsuka, E. Ideguchi, S. Ota, D. Nakajima, M.-H. Tanaka, R. Yoshino, Y. Sasamoto, S. Noji, S. Go, M. Takaki, K. Kisamori, R. Yokoyama, T. Fujii, Y. Kubota, R. Saito, and S. Shimoura.

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3) http://ramap.jaea.go.jp/map/
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Appendix:

The participants of the soil sampling from RNC and CNS are as follows: S. Fujita, K. Ikegami, Y. Ito, T. Kageyama, S. Kubono, K. Morimoto, H. Mukai, S. Nishimura, K. Ogawa, T. Sonoda, and M. Wada.

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Simple screening method for radioactively contaminated food using ashing method with Geiger-Müller counter

M. Wada and H. Haba

After the accident of the Fukushima Daiichi nuclear power plant in the middle of March, 2011, radioactive contamination of food has become a serious concern, especially for those who live in the area where the soil is highly contaminated. It is worth developing a simple screening method, which can be used at home.

In general, β-rays can be detected with a much higher efficiency than γ-rays, and the shielding of β-rays is considerably easier than that of γ-rays. However, β-rays can be shielded by the sample itself. Only surface contamination can be detected by a thin-window Gaiger-Müller (GM) counter. If the radioactivity could be concentrated into a thin sample, the total β radiation detection method would be a significantly sensitive method for contamination surveys.

Most foods consist of water, carbohydrates, proteins, lipids and ash. The concerned radionuclides, such as Cs or Sr, belong to the ash that can be extracted from the sample by an ashing process. The content of ash in food is on the order of 1%, which implies 100 times concentration can be achieved.

First, we tested this method at home using a kitchen grill for fish in order to ash the food samples without contamination. This required approximately 20 min under a medium flame, and the temperature of the samples was less than 500 °C. The weight of ash was less than 1 g. The ashes were flattened and covered with a 10 μm kitchen film (polyvinylidene chloride). The β-rays from the ash were detected using a 5 cmϕ aperture GM survey meter (TCS146B). The amount of potassium (K) in foods was noted from an ingredients table and the specific activity of them could be calculated from the natural abundance of ^{40}K (0.0117%) and the half-life (1.28×10^9 y). The calculated counting rate ΔI_{cal} from the table, with a branching ratio of 90% and a detection efficiency of 25%, was compared with the measured counting rate ΔI_{exp} after subtracting the background (Tab. 1). They agreed well within the limits of a statistical error.

Next, samples that were radioactively contaminated were tested at laboratory (Tab. 2). The specific activities of ^{134,137}Cs and ^{40}K were determined with a HPGe detector before ashing. The expected β-ray count rate I_{cal} was evaluated with the cover area of this setup (75%); the efficiencies of β-rays (14.1%, 12.5% and 22.5% for ^{137}Cs, ^{134}Cs and ^{40}K, respectively), which were evaluated by accounting for the self absorption with a typical thickness (30 mg/cm^2) and for the mean β-energies and branching ratios. When the amount of ash was less than 1 g, the experimentally measured β-ray counting rates ΔI_{exp} agreed with the calculated values.

Table 1. Specific activities of ^{40}K in foods and the counting rates (I_{cal} and I_{exp}) obtained with a GM counter.

spices	ash	K	*S.A.*	m	ΔI_{cal}	ΔI_{exp}
	%	%	Bq/kg	g	cpm	cpm
Spinach	1.7	0.69	219	59	174	169(19)
Soy bean	1.4	0.49	155	33	70	72(14)
W. mellon	0.2	0.12	38	100	51	67(11)
Onion	0.4	0.15	48	114	73	70(9)
Milk	0.7	0.11	35	76	36	34(19)

Table 2. Measurements of contaminated samples.

sample	Cs	K40	mass	ash	ΔI_{cal}	ΔI_{exp}
	Bq/kg	Bq/kg	g	g	cpm	cpm
Berry1	135	76.7	21.1	0.53	33	38(4)
Berry2	135	76.7	22.6	2.1	36	22(3)
Urine1	1727	532	23.9	4.0	378	217(5)
Urine2	549	377	21.0	0.9	149	158(5)
Milk1	145	48.8	20.2	0.2	28	24(3)
Milk2	268	47	20.0	<0.1	42	44(3)

In the Milk1 measurement, the background count $N_{\rm B}$ was 350 in 10 min and the count from the sample N was 592 in 10 min. The detection limit in this case was $N_{\rm m} = 84$ counts (8.4 cpm) with a decision criteria of 3. It roughly corresponded to a total activity of 1.2 Bq in the sample. The maximum amount of the sample was limited by the amount of ash. Considering the self shielding of β-rays, the amount of ash should be less than 1 g. If a 50 g sample is used, the detection limit, in terms of the total specific activity including ^{40}K, reaches 24 Bq/kg.

The contribution from ^{40}K is an important concern. For most foods, the approximate amount of K can be determined from an ingredients table, or, if an exact value is needed, a commercial K ion meter, such as C131K, can be used. We tested many samples with a widely varying K concentration and the values determined showed good agreement with the activity measurements recorded with a HPGe detector.

Although the total β radiation measurement method has unavoidable drawbacks in the identification of radionuclides and in the efficiency evaluation, it is still a useful method for screening with a reference level of 100 Bq/kg. It should also be noted that this method is sensitive to radionuclides that do not exhibit γ-ray radiation, such as ^{90}Sr, even though Sr is not considered a major radionuclide in the presently discussed accident.

NaI scintillation detectors for quantitative measurement of radioactive cesium in milk, blood, urine, and soil

M. Wada

After the accident of the Fukushima Daiichi nuclear power plant in mid-March in 2011, radioactive contamination of soil, food, and human's body became a serious concern in Japan. An HPGe semiconductor detector is the best device to measure the specific activity of weakly contaminated samples, however, it is worth evaluating a possibility of using NaI(Tl) scintillation spectrometers for this purpose, as they are available within a reasonable price range and are relatively easy to operate. Several months after the accident, most short-lived nuclei disappeared, and major radionuclides that remained in the environment are ^{134,137}Cs. This implies that we can use low-resolution spectrometers without identification problems.

Statistically, the detection limit, in units of the count number, according to the Cooper's method[1] is

$$N_{\rm m} = (A_{\rm m}^2 + A_{\rm m}\sqrt{A^2+8B})/2 \approx A_{\rm m}\sqrt{2B}, \quad (1)$$

where $A_{\rm m}$ is the decision criteria, and typically, $A_{\rm m} = 3$ is used. B denotes background counts. The minimum detectable specific activity is then,

$$D_{\rm m} = N_{\rm m}/(\epsilon f T m) \approx A_{\rm m}\sqrt{2B}/(\epsilon f T m), \quad (2)$$

where ϵ is the efficiency, f is the branching ratio, T is the measurement time, and m is the weight of the sample. B and ϵ, T, and m are strongly correlated, and hence, the actual dependence of $D_{\rm m}$ is

$$D_{\rm m} \propto A_{\rm m}\sqrt{w}/(f\epsilon^{\alpha}m^{\beta}\sqrt{T}), \quad (3)$$

where α and β are approximately 0.5, but are variable due to the background and the sample condition, and w is the peak width. If we compare $D_{\rm m}$ for a typical HPGe and a 3 in. NaI detector, $w_{\rm Ge}/w_{\rm NaI} \approx 1/50$ and $\epsilon_{\rm Ge}/\epsilon_{\rm NaI} \approx 1/5$. In such a naive evaluation, NaI shows only $\sqrt{10}$ times less sensitivity as long as disturbing γ-ray peaks are not present in the region of interest.

A typical spectrum of ^{134,137}Cs consists of three peaks (Fig. 1): the one on the right at 800 keV is a mixture of peaks at 795 keV ($f = 85.44\%$) and 801 keV (8.73%) for ^{134}Cs; the one in the center is a 662 keV γ-ray from ^{137}Cs; the one on the left at 600 keV is a mixture of peaks at 605 keV (97.56%), 569 keV(15.43%), and 563 keV (8.38%) for ^{134}Cs. The specific activities (D) and the statistical errors (σ_D) can be deduced from the net counts of these peaks (N_i) and the standard deviation (σ_{Ni}) using the mixed branching ratios ($f_{600} = 121.73\%, f_{662} = 85.1\%, f_{800} = 94.17\%$), and the detection efficiencies ϵ_i, including the sum corrections. For example,

$$D_{134} = N_{800}/(f_{800}\epsilon_{800}Tm), \sigma_{D134} = \sigma_{800}/(f_{800}\epsilon_{800}Tm). \quad (4)$$

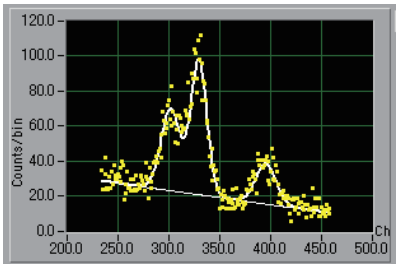

Fig. 1. γ-rays from ^{134,137}Cs of 2.8(1), 3.4(1) Bq measured with 3 in. well-type NaI spectrometer.

The net counts and their standard deviations can be obtained by the least-square fitting of the spectrum with three Gaussian peaks and a linear background (Fig. 1). Alternatively, the classical Covell's method, which determines the background line from the gross counts in specified regions on both sides of the peak, can be used to evaluate the net counts. Although this method cannot separate overlapped peaks, stable values of the sum counts N_L and the standard deviation σ_{NL} can be obtained. The contribution from ^{134}Cs can be subtracted with N_{800} using the ratio, $\alpha = f_{600}\epsilon_{600}/(f_{800}\epsilon_{800})$. The specific activities of ^{137}Cs and the statistical error are

$$D_{137} = (N_L - \alpha N_{800})/(f_{662}\epsilon_{662}Tm), \quad (5)$$

$$\sigma_{D137} = \sqrt{\sigma_{NL}^2 + \alpha^2\sigma_{800}^2}/(f_{662}\epsilon_{662}Tm). \quad (6)$$

A 3 in. well-type NaI scintillation spectrometer with an auto sample changer (Perkin Elmer) has been used to measure milk, blood, and urine from cattle, and soil samples. Although the volume of the sample is limited to 20 cm^3, the 4π solid angle provides a 15% photo-peak efficiency for 662 keV γ-rays. The detection limit for each radioisotope reached 0.15 Bq (7.5 Bq/kg) with a low potassium abundance sample for a one-hour measurement. The limit depends on the level of the continuum background, which is mainly caused by the Compton scattering of 1461 keV γ-rays from ^{40}K. In less contaminated samples, particularly for soil samples, peaks from environmental radioactivities such as ^{214}Bi and ^{214}Pb can cause misidentification problems. The annihilation peak at 511 keV also causes a problem in the background determination.

Well-shielded NaI spectrometers are indeed a useful tool for conducting radioactive contamination surveys.

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Development of Pulse Height Analyzer for Education about Radiation

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The first prototype radiation detector for educational purposes was developed with the peripheral interface controller (PIC). After the Fukushima atomic reactor accidents, people inadequate knowledge abou radiations and radio activities bocame worried about their health and reacted improperly. We as physicists, can contribute to addressing their concern by helping them understand radiation and training them to take suitable actions. We are a group consisting of teachers, a curator, and physicists, and we have been discussing on how to develop a good education system for this purpose and have identified important items "experience of radiation measurement", "understand of radiation detection principle," and "assembling of radiation detector." The feature requirements for a radiation detector kit are

(1) Easy to assemble without a soldering iron.
(2) Inexpensive, with easily procurable parts.
(3) Ablity to detect environmental radiation.
(4) Tunable detection threshold.

The complete design of the first trial detector is described in Kawamo's article[1]. The detector consits of two main items, a sensor component and a pulse height analyzer (PHA). Radiations are injected into the sensor component and converted readable voltage signals. The sensor component consists of a sensor, a preamplifier, a shaping amplifier and a peak hold circuit.[1] Then, the PHA converts the voltage signal into digital values and displays their energy spectrum. The PHA consists of a comparator, an ADC, a pulse generator, and a display. The function diagram is shown in Fig. 1. When the signal from the peak hold circuit of the sensor component is fed into the comparator and ADC, only the signals exceeding the threshold are converted into a digital pulse height value by the ADC. Then, the digital values are transmitted to the PC for displaying the energy spectrum. The PHA was made from PIC18F4550[2] by Microchip; it is an 8-bit CPU with an ADC, comparators, digital inputs, digital outputs (DO), and a USB interface within a single chip. The chip has flash ROM for programing, which can be written into by a dedicated writer or a USB interface. The program for the PIC is written in C language[3]. The board with the PIC and other necessary parts costs 1100 yen. The typical AD conversion time required after signal rises in the peak hold circuit is 10μs. After AD conversion, the digitized pulse height values are stored in the PIC memory and transferred periodically to the PC through the USB interface. This chip can measure up to 100 KCPS radiation with adequate PIC memory. However, this value is extreamly high radiation to handle in a classroom. The data acquisition program on the PC was developed by using Microsoft Visual C#.[4] The program is event driven. When the PIC transmits the pulse height data, the PC receives them and updates the histogram display. Transmission requires about the 5 ms inclusive of the USB interrupt process. Fig. 2 shows the obtained histogram of ^{137}Cs the checking source.

The prototype of the PHA for the radiation detector was developed for educational purposes. It satisfied the functional requiements. We will produce 200 radiation detector assembly kits for supplying to classrooms.

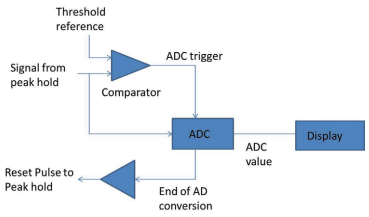

Fig. 1. Functional block diagram of pulse height analyzer.

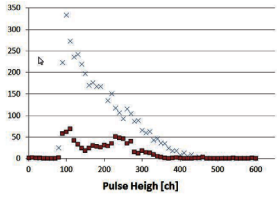

Fig. 2. Histogram of the pulse peight for ^{137}Cs. Squares and crosses represent backgrounds and signals respectively.

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Alternative method to evaluate internal dose using simple γ-ray spectrometer

M. Wada

Internal dose has been a critical topic of concern after the accident at the Fukushima Daiichi nuclear power plant. A significant amount of radionuclides released from the power plant has been spread over a wide area in Japan. After the such incident the human body may take radioactive substances directly from the plume in the initial days and indirectly from contaminated foods afterward. In order to evaluate the internal dose, the concentration of radioactive substances in the whole body should be measured. A so-called Whole Body Counter (WBC) has been exclusively used for such a purpose. However, popularly used WBCs have not been designed for low-level contamination and cannot be aimed for use in a high background environment. Furthermore, the number of WBCs for public use is limited compared to the demand. Hence, an alternative way to evaluate the concentration of radioactive substance in the human body is worth considering.

The main radioactive substances of concern are ^{137}Cs and ^{134}Cs. From a survey conducted on animals, it is known that radioactive Cs is mainly distributed in the muscles and not in the internal organs or bones[1]. It would be more effective to measure the "muscle-rich" parts of a human body with a well-shielded γ-ray detector. A good candidate for such a part is the leg. A "U"-form shielding can be placed on top of the leg and a detector with a cylindrical shielding block can be placed at the bottom. Although the palm would not be muscle-rich, we evaluated the measurement of specific activity in the palm using standard γ-ray spectrometers used for food screening, without any modification to the shielding.

A phantom sample was made using a globe filled with 173 g contaminated rice with specific activities of 62.2(1.9) Bq/kg and 50.5(2.0) Bq/kg for ^{137}Cs and ^{134}Cs, respectively. The phantom was placed on top of the standard HPGe spectrometer to mimic a palm is grasping the head of the detector. At first, the detection efficiency, including the sum correction, was calibrated with a long measurement time. Then, the same phantom and an actual hand were measured with a realistic measurement time of 10 min. The net counts and their statistical error σ_N, the deduced activities, and the detection limits D_m in case of the decision criteria of 3 were listed in Table 1. The detection limits reached ≈2 Bq, which correspond to specific activities of ≈11 Bq/kg.

Table 1. γ-ray measurement of phantom palm (173 g) (top) and human hand (bottom) for 10 min measurements using a HPGe spectrometer.

γ-ray keV	efficiency %	count (σ_N)	Activity Bq	D_m Bq
662	1.40(5)	73.7(9.0)	10.3(1.3)	1.87
604	1.28(2)	68.3(9.0)	9.1(1.2)	2.18
795	1.05(2)	51.0(7.1)	9.45(1.3)	1.55
1460	0.63(9)	8(2.8)	16.5(7.0)	26
662	1.40(5)	3.8(2.8)	0.53(0.39)	1.68
604	1.28(2)	1.5(3.5)	0.20(0.47)	2.05
795	1.05(2)	1.8(2.5)	0.33(0.47)	2.28
1460	0.63(9)	19(4)	43.9(9.9)	19.0

A similar test was also performed with a 3" $\phi\times$3" NaI(Tl) scintillation spectrometer used for food screening (OKEN FNF-401). The efficiency, including the sum correction, for the mixed peaks of 795 keV and 801 keV γ-rays (branching ratio is 0.942) from ^{134}Cs was 3.1%, and the detection limit was 6.0 Bq for the same 172 g sample with a 10 min measurement. The result corresponds to a specific activity of 36 Bq/kg.

These two test measurements showed promising results. With a slight modification of the shielding block, γ-ray spectrometers for food samples can be used to evaluate the concentration of radioactive substances in a human palm.

Another problem in the WBC involves determining the detection efficiencies. Usually, phantom human bodies are used for the efficiency calibrations. However, the geometry of the human body is quite different for each individual, and the different distributions of radioactive substances within the body can cause large ambiguities in the efficiency evaluation. We propose here a simple way to evaluate the concentration of radioactive substances in the whole body as a ratio to that of potassium. Although the actual distributions of potassium and cesium are not equivalent[2], we can approximate that the two alkali elements are equally distributed to some extent. Under such an approximation, the ratio of the specific activities can be

$$\frac{A_{137\mathrm{Cs}}}{A_{40\mathrm{K}}} = \frac{N_{661}}{N_{1460}}\frac{\epsilon_{1460}}{\epsilon_{661}}\frac{b_{1460}}{b_{661}} \approx 0.056\frac{N_{661}}{N_{1460}}, \qquad (1)$$

where N_x are the net counts, ϵ_x are the efficiencies, and b_x are the branching ratios. It should be noted that 1) N_{1460} is usually too small for a short-time measurement with a small HPGe spectrometer; 2) a sum peak of ^{134}Cs at 1401 keV can be overlapped with the ^{40}K peak in NaI measurements; and 3) the internal absorptions of different γ-rays can be slightly different.

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