Special environmental monitoring around Tokai-mura after the accident of the Fukushima Dai-ichi Nuclear Power Station

H. Fujita[1], M. Nagaoka[1], T. Kono[1], M. Takeyasu[1], M. Kawasaki[2], T. Okura[2], N. Tsujimura[1], S. Sumiya[1], T. Momose[1], S. Furuta[1]

[1]Nuclear Fuel Cycle Engineering Laboratories, Japan Atomic Energy Agency, 4-33 Tokai-mura, Naka-gun, Ibaraki 319-1194, Japan

[2]Nuclear Science Research Institute, Japan Atomic Energy Agency, 2-4 Shirane Shirakata, Tokai-mura, Naka-gun, Ibaraki 319-1195, Japan

The nuclear accident at the Fukushima Dai-ichi Nuclear Power Station of Tokyo Electric Power Co. (TEPCO) was caused by the Tohoku District – off the Pacific Ocean Earthquake. Various kinds of radionuclides were released from the power station right after the accident occurred, and then reached our laboratories. Therefore, environmental monitoring was reinforced by observation of air absorbed dose rate, periodic collection of airborne dust and precipitation. The collected samples were used to determine radioactivity of gamma ray emitters by High pure Germanium semiconductor detector. Moreover, rainwater and humidity were collected to do the usual routine monitoring.

Radionuclides of ^{131}I, ^{134}Cs, ^{137}Cs, etc. were detected in airborne samples collected after 13th of March, continuously, although these nuclides had never been found in any samples for the last ten years around our site. Then, ^{90}Sr was also detected using chemical analysis of an airborne sample. These radionuclides such as ^{90}Sr, ^{131}I, ^{134}Cs and ^{137}Cs were determined around the accident station, too. Therefore, our monitoring detected influence of the accident into our site. In other samples such as seawater, seabed soil, etc., similar radionuclides were detected including the influence of the accident.

KEYWORDS: nuclear accident at Fukushima Dai-ichi Nuclear Power Station of Tokyo Electric Power Co., environmental monitoring, iodine-131 (^{131}I), Cesium-134 (^{134}Cs), Cesium-137 (^{137}Cs)

1. Introduction

On March 11, 2011, reactors at the Fukushima Dai-ichi Nuclear Power Station (NPS) of Tokyo Electric Power Co. (TEPCO) in Japan were shut down following the Tohoku District – off the Pacific Ocean Earthquake. By the earthquake, the external power supply was damaged. The first enormous tsunami hit the Fukushima Daii-ichi NPS at 15:27 on March 11 (41 minutes after the earthquake), followed by the next enormous wave around 15:35 (Government of Japan, 2011). The seawater pump facilities for cooling auxiliary systems were submerged and stopped functioning. In addition to that, all the emergency diesel power generators and the distribution boards installed in the basements of the reactor buildings and turbine buildings were inundated and stopped functioning. Fires, explosions and possible partial core meltdowns happened by no

operation of water pumps. Radioactive fission products such as iodine-131 (^{131}I), Cesium-134 (^{134}Cs), Cesium-137 (^{137}Cs), etc. were then discharged from the Fukushima Dai-ichi NPS into the atmosphere (e.g. MEXT, 2011a). These radionuclides were detected in various foreign countries (Olivier Evrard, et al., 2012, S. MacMullin, et al., 2012)

After the accident, our section reinforced environmental monitoring of air absorbed dose rate, airborne dust and fallout. Our safety control building is in the Nuclear Fuel Cycle Engineering Laboratories (NCL), located about 100 km south of the Fukushima Dai-ichi NPS. The dose rate was continuously observed by on-site and off-site monitoring posts and stations, except for a few monitoring stations inoperative due to a temporal power failure. Measurement of radioactivity concentration in airborne sample was begun from 13th of March (2 days later of the earthquake) because our section also received damage of the earthquake and was also cut off from the commercial power supply system. Moreover, we started to collect fallout using temporal sampler to measure its radioactivity from 15th of March. In this paper, the results of special monitoring are shown with a part of usual monitoring data.

2. Measurement method

2.1. Air absorbed dose rate

Our section controls 8 monitoring posts (to measure only air absorbed dose rate) and 2 monitoring stations (to measure dose rate and radioactivity concentration of gaseous β-ray emitter such as Kripton-85 (^{85}Kr), and to collect airborne dust) on-site the NCL, and 3 monitoring stations off-site the NCL. The instrument used was a 2"Φ×2" energy-compensated NaI(Tl) scintillation detector as a dose rate monitor. The monitoring posts and stations are usually used in routine monitoring around Tokai reprocessing plant located in the NCL.

2.2. Radioactivity concentration in airborne sample

Portable Sampler (PNC-800) collecting airborne and gaseous dust was set in front of Safety control building (which is on-site the laboratories) on 13th of March for the special monitoring. Filter (HE-40T, Toyo-roshi Kaisha Ltd., Japan) was used to collect airborne dust and charcoal with TEDA (tri-ethylene-diamine) (CHC-50, Toyo-roshi Kaisha Ltd., Japan) was used to collect gaseous dust such as iodine. As a general rule their filters were replaced every 12 hours. However, replaced interval of the filters was changed depending on the dose rate in air. The intervals were between 3 hours and 7 days. After the collection, the sample was set on high-pure germanium semiconductor detector (HP-Ge detector) to determine radioactivity concentration of gamma-ray emitters with measurement time between 1000 and 5000 seconds. The radioactivity concentration was corrected using each half-life, to obtain the value at start time of the sampling.

2.3. Radioactivity concentration in fallout

A large water basin (0.5 m^2 in area) was set on the rooftop of the safety control building for the special monitoring. As a general rule the fallout from atmosphere was collected when it rained. After the collection, the sample was transferred into 2 L marinelli type vessel and set on the HP-Ge detector to determine radioactivity concentration of gamma-ray emitters. The radioactivity concentration was corrected using each half-life, to obtain the value at start time of the sampling.

2.4. Radioactivity concentration in usual monitoring items

As a routine environmental monitoring around the Tokai reprocessing plant, our section determines radiation dose and radioactivity concentration in environmental samples such as leafy vegetable, milk, surface soil, seawater, marine product, etc. In the monitoring program, tritium (^{3}H), total beta radioactivity, ^{90}Sr, ^{137}Cs, plutonium-239, 240 ($^{239,240}Pu$) are analyzed in the samples, periodically. A part of radionuclide concentration was influenced by radioactivity discharged from the Fukushima Dai-chi NPS and therefore the obtained data were researched in view point of radioactivity ratio dependency on each radionuclide and of comparison with other radioactivity concentration.

3. Results and discussion

3.1. Air absorbed dose rate

Figure 1 shows air absorbed dose rate obtained by the monitoring stations and posts during a period from 13th to 31st of March. After 1:00 am on 15th, the dose rate increased on all of the stations and posts simultaneously and reached 5 μGy h^{-1} (1 minute average) at 7:13 am on the same day on MP-2 and MP-7. Three maximal dose rates were recorded on 15th, 16th and 21st of March but dose rates on the later 2 days did not exceed the first highest dose rate. The dose rates on each post and station decreased with time after the accident. During the period, no accident was happened at our facilities handling

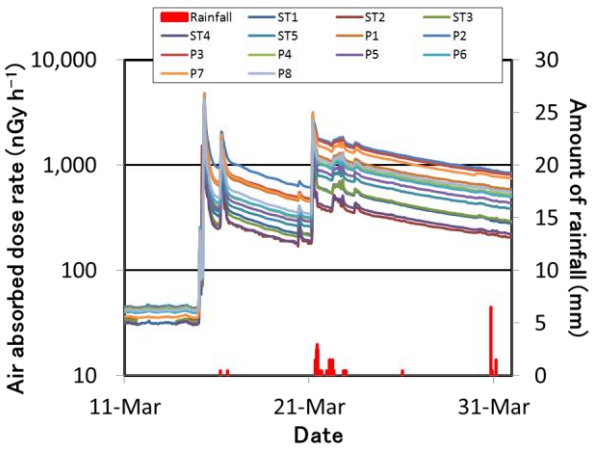

Figure 1

Transition of air absorbed dose rate measured at monitoring stations and posts with amount of rainfall.

radioactivity and at other facilities and therefore the sudden increases of the dose rates were attributed to radioactivity discharged for the NPS.

Levels of the dose rates were dependent on each monitoring station and post. Particularly, all the monitoring posts were situated within 1 km radius in our site but the levels were different. The difference could be caused by various environmental conditions, for example, whether post is covered by leaves of trees or not. The levels were supposed to be controlled by the environment.

3.2. Radioactivity concentration in airborne sample

Figure 2 shows change of radioactivity concentration in airborne sample collected from 13th of March to 2nd of May. We collected radioactivity using dust filter and charcoal cartridge, separately. The observed radionuclides were ^{131}I, ^{134}Cs and ^{137}Cs, with minor contribution of ^{129m}Te, ^{133}I, ^{136}Cs, etc. Surprisingly, radioactive cesium was detected in charcoal cartridge. The reason of the detection could be considered that the chemical form was easy to pass through dust filter (Furuta, et al., 2011).

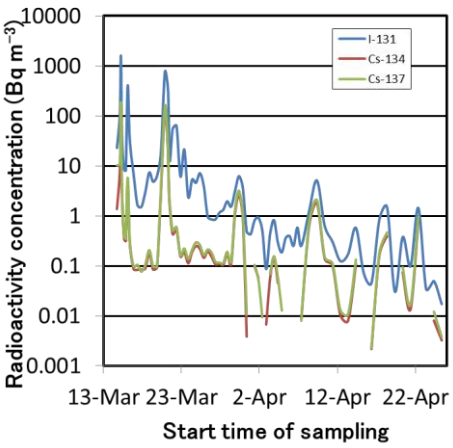

Figure 2

Transition of radioactivity concentration in airborne sample.

Total amount of ^{131}I, ^{134}Cs and ^{137}Cs was plotted in figure 2. Moreover radioactivity ratio of each nuclide to ^{137}Cs was plotted in figure 3. In figure 2, ^{131}I was detected over minimum detection limit for whole sampling duration but radioactivity concentration of the other nuclides was determined depending on sampling date. The change of each radioactivity concentration showed, on the

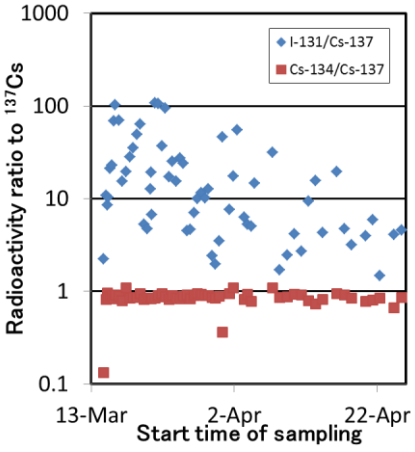

Figure 3

Transition of radioactivity concentration ratio of ^{131}I or ^{134}Cs to ^{137}Cs in airborne sample.

whole, a decreasing tendency. On the other hand, the ratio of ^{131}I to ^{137}Cs fluctuated and the ratio of ^{134}Cs to ^{137}Cs was constant as shown in figure 3. The ratio tendency was supported by the difference of chemical property such as gaseous material or particle.

Takeyasu, et al. (2012) calculated the committed dose due to inhalation using the concentration measured at the NCL from 13 March to 23 May in 2011. The committed dose due to inhalation was estimated on the basis of the air concentration at the NCL, that is, from the radioactivity inhaled by a person who would have continuously stayed outdoors. The committed effective dose for adults and children was estimated to be about 0.6 mSv and 0.9 mSv, respectively. The committed equivalent dose to the thyroid was about 8 mSv and 15 mSv for adults and children, respectively.

3.3. Radioactivity concentration in fallout

Figure 4 shows change of radioactivity in fallout from 15th of March to 2nd of May, with rainfall data. Totally, the radioactivity decreased with time after the accident but did not show any relationship with rainfall. Comparing figure 2 with figure 4, each radioactivity was similar tendency, in other words higher radioactivity in airborne sample was higher radioactivity in fallout and the opposite tendency was observed.

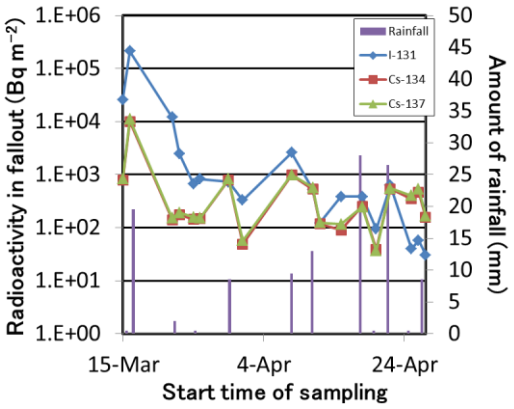

Figure 4

Transition of radioactivity in fallout.

3.4. Radioactivity concentration in usual monitoring items

As routine monitoring, radioactivity concentration in environmental samples was measured using the HP-Ge detector or some detectors after the chemical separation. In all the samples collected from April to December in 2011, ^{137}Cs concentrations were detected with higher level than the previous environmental background level before the accident in the past 10 years monitoring around the NCL. It was concluded that the ^{137}Cs was determined with influence of the accident because ^{134}Cs was also detected on similar level to the ^{137}Cs. On the other hand, ^{90}Sr concentration exceeded the fluctuation level in airborne dust and a part of marine sample such as seaweed but was low level, relatively. Table 1 shows radioactivity concentration of ^{137}Cs and ^{90}Sr determined in the sample, with both of the concentrations exceeded the environmental background levels. In table 1, radioactivity concentration ratio of ^{90}Sr to ^{137}Cs was added. The ratio was between one thousandth and one tenth and therefore ^{90}Sr concentration was very low in environmental sample comparing with ^{137}Cs.

Table 1 Radioactivity concentration of ^{137}Cs and ^{90}Sr in environmental samples with the radioactivity ratio

Sampling items	^{137}Cs	^{90}Sr	$^{90}Sr/^{137}Cs$
Airborne dust	1.0 – 910 (mBq/m^3)	0.010 – 0.36 (mBq/m^3)	$(0.033 - 2.2) \times 10^{-2}$
Seawater	0.93 – 1.1 (Bq/L)	0.024 – 0.027 (Bq/L)	$(2.5 - 2.6) \times 10^{-2}$
Seabed soil	$(1.5 - 4.3) \times 10^2$ (Bq/kg dry)	0.51 – 0.55 (Bq/kg dry)	$(1.3 - 3.4) \times 10^{-3}$
Shellfish	7.1 – 14 (Bq/kg raw)	0.025 – 0.063 (Bq/kg raw)	$(1.8 - 8.8) \times 10^{-3}$
Seaweed	5.4 – 50 (Bq/kg raw)	0.076 – 1.1 (Bq/kg raw)	$(0.48 - 8.4) \times 10^{-2}$
Coastal water	0.14 – 0.19 (Bq/L)	$(4.7 - 6.0) \times 10^{-3}$ (Bq/L)	$(2.7 - 4.2) \times 10^{-2}$

On the other hand, $^{239,240}Pu$ concentration was not influenced by the accident in all the samples. Although Pu caused from the accident was detected near the Fukushima Dai-ichi NPS (MEXT, 2011b), Pu could not reach the NCL.

4. Conclusions

In the special monitoring, it was found that air absorbed dose rate, radioactivity concentration in air borne dust and radioactivity in fallout were influenced by the radionuclides discharged from the Fukushima Dai-ichi NPS but decreased with time after the accident. Therefore, it was suggested that the new radionuclides could not be released from the NPS.

In the routine monitoring, radioactivity concentrations of ^{90}Sr, ^{131}I, ^{134}Cs and ^{137}Cs were raised by the amount of each radionuclide discharged from the NPS, depending on the sample. Particularly ^{134}Cs were only detected by the accident because ^{134}Cs did not exist in the environment. Although both the ^{134}Cs and ^{137}Cs concentrations were a little high level, the ^{90}Sr concentration was low level even if the ^{90}Sr was influenced by the accident. Moreover, the radioactivity concentration ratio of ^{90}Sr to ^{137}Cs was between one thousandth and one tenth.

References

Sadaaki Furuta, Shuichi Sumiya, Hitoshi Watanabe, Masanao Nakano, Kenji Imaizumi, Masanori Takeyasu, Akira Nakada, Hiroki Fujita, Tomoko Mizutani, Masato Morisawa, Yuji Kokubun, Takahiko Kono, Mika Nagaoka, Hiroya Yokoyama, Tomonori Hokama, Tokuju Isozaki, Masashi Nemoto, Yoshinori Hiyama, Toshimitsu Onuma, Chiaki Kato and Tamotsu Kurachi, 2011. Results of the Environmental Radiation Monitoring Following the Accident at the Fukushima Daiichi Nuclear Power Plant – Interim Report (Ambient Radiation Dose Rate, Radioactivity Concentration in the Air and Radioactivity Concentration in the Fallout) -. JAEA-Review 2011-035.

Government of Japan, 2011. Report of the Japanese Government to the IAEA Ministerial Conference on Nuclear Safety - The Accident at TEPCO's Fukushima Nuclear Power Stations -.

Ministry of Education, Culture, Sports, Science and Technology (MEXT), 2011a. Readings of Radioactivity Concentration of Nuclides in the Air and the Soil inside of the 20 km Zone of Fukushima Dai-ichi NPP.

Ministry of Education, Culture, Sports, Science and Technology (MEXT), 2011b. Analysis

Results of Soil Samples Taken inside 20 km Zone of TEPCO Fukushima Dai-ichi NPP.

Olivier Evrard, Pieter Van Beek, David Gateuille, Véronique Pont, Irène Lefèvre, Bruno Lansard, Philippe Bonté, 2012. Evidence of the radioactive fallout in France due to the Fukushima nuclear accident. Journal of Environmental Radioactivity doi:10.1016/j.jenvrad.2012.01.024.

S. MacMullin, G.K. Giovanetti, M.P. Green, R. Henning, R. Holmes, K. Vorren, J.F. Wilkerson, 2012. Measurement of airborne fission products in Chapel Hill, NC, USA from the Fukushima Dai-ichi reactor accident. Journal of Environmental Radioactivity doi:10.1016/j.jenvrad.2012.01.026.

Masanori Takeyasu, Masanao Nakano, Hiroki Fujita, Akira Nakada, Hitoshi Watanabe, Shuichi Sumiya and Sadaaki Furuta, 2012. Results of environmental radiation monitoring at the Nuclear Fuel Cycle Engineering Laboratories, JAEA, following the Fukushima Daiichi Nuclear Power Plant accident. Journal of Nuclear Science and Technology, http://dx.doi.org/10.1080/00223131.2012.660014.