Radiological Conditions in the Environment around the Fukushima Daiichi Nuclear Power Plant Site

Kimiaki SAITO[1*], Hideaki YAMAMOTO[2], Satoshi MIKAMI[2], Masaki ANDOH[2], Norihiro MATSUDA[3], Sakae KINASE[3], Shuichi TSUDA[3], Tadayoshi YOSHIDA[3], Shinichiro MATSUMOTO[4], Tetsuro SATO[5], Akiyuki SEKI[3] and Hiroshi TAKEMIYA[3]

[1]*Japan Atomic Energy Agency*
2-2-2 Uchisaiwai-cho, Chiyoda-Ku, Tokyo 100-8577, Japan
[2]*Japan Atomic Energy Agency*
7-1 Taira Omachi, Iwaki, Fukushima 970-8026, Japan
[3]*Japan Atomic Energy Agency*
2-4 Shirane, Tokai-mura, Naka, Ibaraki 319-1106, Japan
[4]*International Research Institute for Nuclear Decommissioning*
2-23-1 Nishi-shimbashi, Minato-ku, Tokyo 105-0003, Japan
[5]*Hitachi Solutions East Japan Ltd.*
2-16-10 Honcho, Aoba-ku, Sendai, Miyagi 980-0014, Japan
**email: saito.kimiaki@jaea.go.jp*

Abstract

Large-scale environmental monitoring that has been repeatedly performed has revealed the characteristics of radiological conditions around the Fukushima NPP site. Several radionuclides other than radiocesium were detected three months after the accident, but radiocesium turned out to be far more important than the other radionuclides from the viewpoint of long-term exposures. Radiocesium deposition was found to be heterogeneous at different scales. On the one hand, air dose rates in environments related to human living have decreased much faster than the physical decay of radiocesium; on the other hand, the reduction has been close to physical decay in pure forests. Movement of deposited radiocesium has generally been very slow except in urban areas, while radiocesium deposited on the ground has gradually penetrated into the ground, resulting in decreased air dose rates because of the increased radiation shielding effect.

Key words: distribution, air dose rate, large-scale monitoring, radionuclide deposition density, temporal change

1. Introduction

Enormous amounts of radionuclides released into the atmosphere by the Fukushima Nuclear Power Plant (NPP) accident were deposited on wide regions of East Japan. Several different kinds of large-scale environmental monitoring have been repeatedly implemented since the accident, and the accumulated monitoring data have revealed the characteristics of environmental radiological conditions around the Fukushima NPP. This paper reviews the regional and time-dependent characteristics of the contamination based on an analysis of the observed data. This review mostly cites the results from national so-called radioactive substance distribution mapping projects (mapping projects) (NRA, 2016a; Saito & Onda, 2015) commissioned by the National Government, because those have continuously obtained comprehensive monitoring data. Radiological conditions in the environment provide information fundamental to a discussion of exposures to the public as well as biota. Further, they provide keys to elucidate contamination pathways. Therefore, proper recognition of radiological conditions is essential in several respects.

2. Large-scale Environmental Monitoring

2.1 Air Dose Rates

Three kinds of large-scale measurements for air dose rates have been continuously performed in mapping projects (Saito & Onda, 2015). Further, large-scale aerial monitoring has been repeatedly carried out by others (Sanada *et al*., 2014).These measurements can be characterized briefly as follows:

1) Measurements in Undisturbed Fields

At about 6,500 fixed locations in the zone within

80 km of the Fukushima NPP site, air dose rates have been measured using standard survey meters (Mikami *et al.*, 2015a). In the measurements, undisturbed flat fields where conditions are expected not to change for a long time were selected. The measurements are thought to give accurate reference values on air dose rates and how they change according to natural weathering effects.

2) Car-borne Surveys

Car-borne surveys are carried out using KURAMA (Kyoto University RAdiation MApping)-II systems developed at Kyoto University, which are compact and easy to operate (Tsuda *et al.*, 2015). The surveys cover broad contaminated areas in East Japan in collaboration with a number of local municipalities (Andoh *et al.*, 2015). The total traveling distance of the survey cars is several tens of thousands of kilometers in each campaign. A great quantity of air dose rates can be obtained on the ground which are useful for statistical analysis.

3) Person-borne Surveys

A person carrying a KURAMA-II system walks around in an area of 1 km square as long as possible including on small roads which are difficult to drive by car. This kind of survey is intended to obtain air dose rates in various environments related to human activities. These are important for revealing public exposure dose aspects. About 600 areas of 1 km square are targeted each time. A good correlation has been found among air dose rates in undisturbed fields, those obtained by car-borne surveys and those by person-borne surveys, if the measurements were taken in closely located areas.

4) Aerial Monitoring

Monitoring by helicopter has been conducted in another national project since the accident (Sanada *et al.*, 2014). As aerial surveys can cover entire targeted regions, the map obtained is useful for grasping broad distributions of air dose rates over wide areas, though the data contain some uncertainty because the original data were obtained from a height of 300 m.

2.2 Radionuclide Deposition

Two radionuclide ground deposition quantities were obtained. One was radionuclide deposition density in terms of Bq/m^2, which consisted of integrated radionuclide activity over the vertical direction in the ground. The other was depth profiles which were radionuclide concentrations as a function of soil depth in terms of Bq/kg.

1) Radionuclide Deposition Density

In the first campaign of the mapping projects, soil samples were collected at about 2,200 points and analyzed at laboratories using gamma spectrometry with a Ge detector (Saito *et al.*, 2015). Cesium (Cs)-134 and Cs-137 were detected in all samples; while, iodine(I) -131, Te-129m and Ag-110m were detected at some of the sampling points. Further, radio chemical analysis was used to examine plutonium(Pu)-238, Pu-239+240, strontium(Sr)-89 and Sr-90 in a selection of 100 soil samples. Since the start of the second campaign from December 2011, *in situ* gamma spectrometry using a portable Ge detector has been employed to quantify gamma-emitting radionuclides (Mikami *et al.*, 2015b). This method can determine the average deposition density of a location by detecting gamma rays coming from sources widely distributed there. In the second campaign, *in situ* Ge measurements were carried out at about 1,000 locations in East Japan. Since the third campaign, measurements have been conducted at about 380 locations in the 80 km zone.

2) Radionuclide Depth Profiles

Depth profiles of radiocesium were investigated by sampling soil at different depths using a scraper plate that can minimize cross-contamination of soil samples (Matsuda *et al.*, 2015). Soil was peeled off from the surface of the ground from 5 mm depth up to a few cm according to the conditions, and the radiocesium concentration was determined by Ge spectrometry.

3. General Features of Radiological Conditions

3.1 Air Dose Rates

Figure 1 shows air dose rate distribution maps within the 80 km zone from the NPP site for five different occasions through 2015. These air dose rate maps were constructed by integrating results from undisturbed flat fields with those obtained by aerial monitoring. In the areas where ground-measured data existed, those data were used; and in the areas where they did not exist, aerial monitoring data were used after some adjustment. The aerial monitoring data tended to be systematically higher than those measured on the ground, especially in various environments related to human living.

The contamination was quite heterogeneous at different scales. As the maps show, the area north-west of the NPP site was highly contaminated, and the middle part of Fukushima Prefecture, called Nakadori, had relatively high contamination. Even in a small area with a range of a few tens of meters, the air dose rate often changes drastically with location. Further, there existed locally high dose-rate points where radiocesium had accumulated due to water flow; for example, rain gutters and gullies around houses.

The heterogeneity made exposure dose evaluation difficult, because the exposure levels changed markedly with the location of the inhabitant. Therefore, if one intends to evaluate realistic exposure doses from air dose data, changes in air dose rate with location need to be precisely considered together with detailed information on the behavior of the inhabitant.

3.2 Radionuclide Deposition

In the first campaign, carried out in June 2011, the following radionuclides were detected at many mapped locations: Cs-134, Cs-137, I-131, Te-129m, Ag-110m, Sr-89, Sr-90, Pu-238+239 and Pu-240. Figure 2 shows a Cs-137 deposition map as of June 14, 2011 (MEXT, 2012; Saito *et al.*, 2015). In this respect, it is quite similar to that of the air dose rate distribution shown in Fig. 1, since

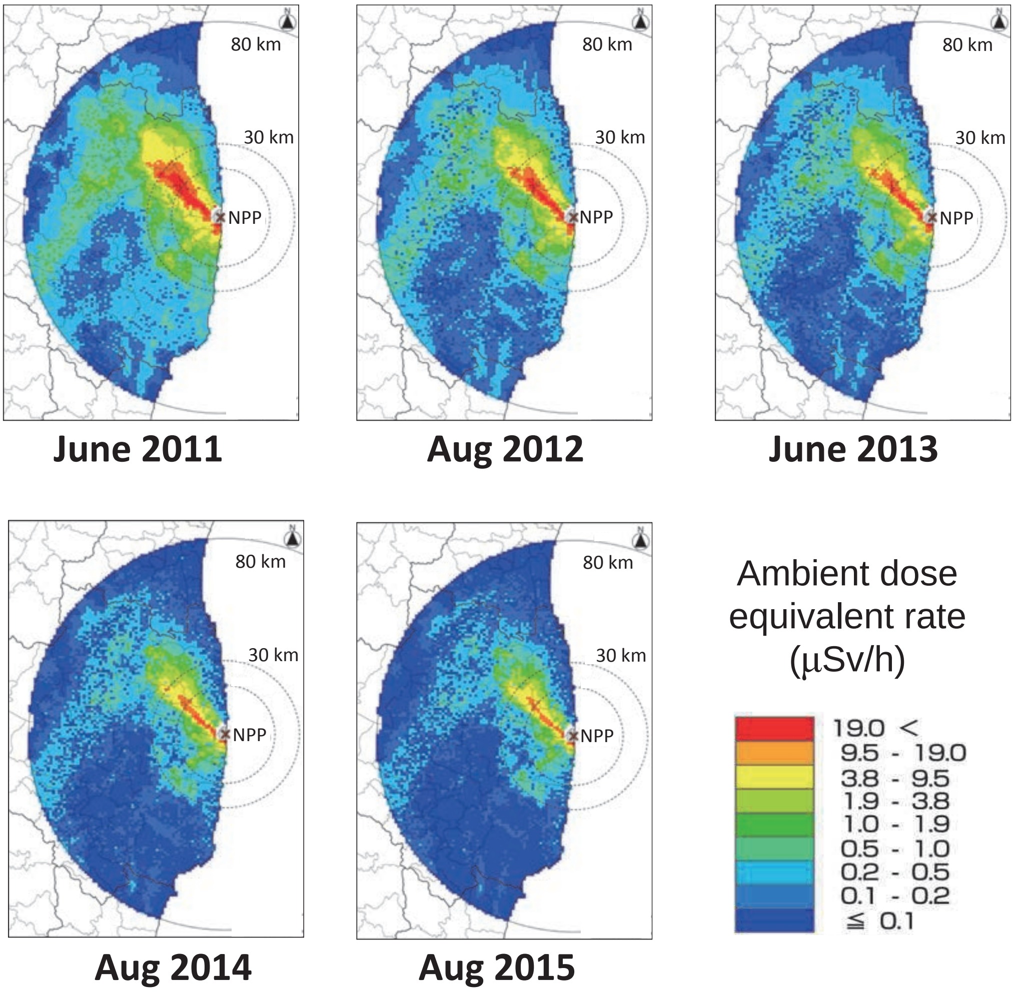

Fig. 1 Air dose rate distribution within the 80 km zone from June 2011 till August 2015. An air dose rate map in undisturbed fields was integrated with aerial monitoring data for each occasion.

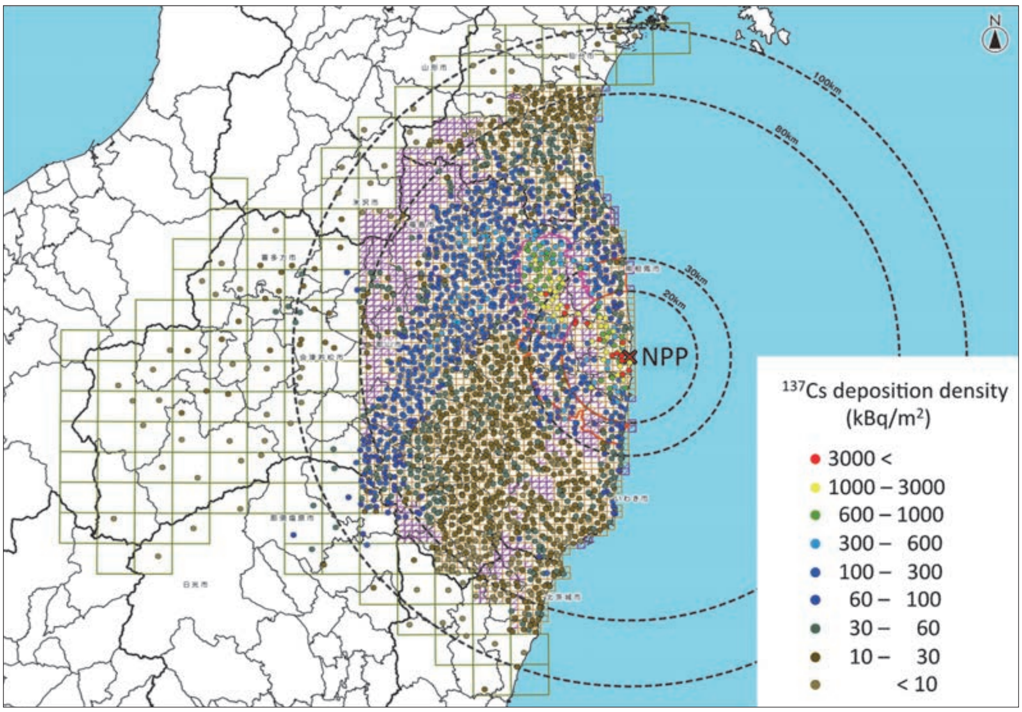

Fig. 2 Deposition density distribution of Cs-137 as of June 14, 2011, determined by soil sampling and analysis. (MEXT, 2012; Saito *et al*., 2015)

radiocesium was already the main contributor to the air dose rate in June 2011.

The air dose rate in an undisturbed flat field was found to have a strong correlation with the deposition density of radiocesium, at least after June 2011, when radiocesium became the main contributor to the air dose rate. Figure 3 shows the relation of air dose rates to deposition densities measured in November 2013. A similar strong correlation was observed in each campaign of the mapping project, though the conversion ratio from the deposition density to the air dose rate has changed with time. The relationship enabled us to create a detailed deposition density map from the air dose rate distribution.

Rough and conservative evaluations of effective doses for 50 years after June, 2011 were made for the observed radionuclides (MEXT, 2012; Saito *et al*., 2015) using the dose conversion coefficients provided by the IAEA (2000) as shown in Table 1. It was confirmed that the effective doses from radiocesium were higher than those from any other radionuclide by more than two orders. It must be noted that the high doses indicated in the table were never actually encountered, because very conservative and unrealistic assumptions were used in the evaluation. The evaluation's goal was to examine the relative importance of exposure doses among the observed radionuclides.

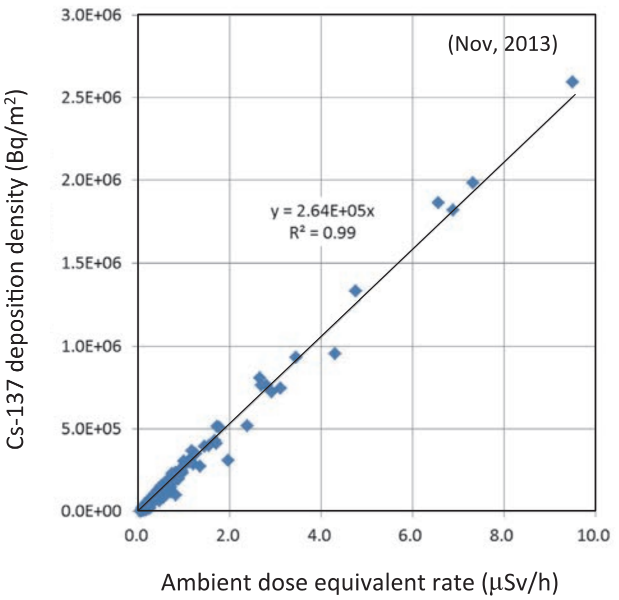

Fig. 3 Relation of air dose rates to Cs-137 deposition densities in undisturbed fields.

Table 1 Estimated maximum effective dose for 50 years from June 2011. (MEXT, 2012; Saito *et al*., 2015)

Nuclide	Maximum deposition density (Bq/m^2)	Effective dose for 50 y (mSv)
Cs−134	$1.4x10^7$	710
Cs−137	$1.5x10^7$	2000
I−131	$5.5x10^4$	0.015
Sr−89	$2.2x10^4$	0.00061
Sr−90	$5.7x10^3$	0.12
Pu−238	$4x10^0$	0.027
Pu−239+240	$1.5x10^1$	0.12
Ag−110m	$8.3x10^4$	3.2
Te−129m	$2.7x10^6$	0.6

The total amount of Cs-137 deposited on Japanese territory was calculated by integrating deposition densities observed on the ground with those from aerial monitoring. An integration method similar to that for air dose rates described in the previous section was used. The total Cs-137 deposition in the 80 km zone and in East Japan overall were estimated to be 1.6 and 2.0 PBq, respectively. In the 80 km zone, about 70 percent of radiocesium was estimated to be deposited in forested regions, 20 percent in agricultural regions and 5 percent in urban regions, which are nearly proportional to the areas.

The deposition density ratio between different radionuclides indicates specific regional features, and this may reflect differences in reactor origins and contamination pathways. The ratio of I-131 and Te-129m to Cs-137 was higher in the southern coastal region (Saito *et al*., 2015). The deposition density of Ag-110m had a good correlation with that of Cs-137 over wide areas of Fukushima, Tochigi and Gumma prefectures (Mikami *et al*., 2015b), suggesting the contamination of these areas had the same origin.

Deposition ratios between Cs-134 and Cs-137 were analyzed by Chino *et al*. (2016) with atmospheric dispersion simulation and reactor analysis data. The observed Cs-134/Cs-137 ratios differed slightly according to region, and the regional differences were thought to reflect the main source of contamination, that is, the unit 1, 2 or 3 reactor. A limited specific region north northwest from the NPP site was thought to be contaminated mainly by a release from the unit 1 reactor. Over other wide regions, the contamination came from the unit 2 and 3 reactors, with the contribution of each reactor seeming to vary according to the region.

4. Time-trend of Radiological Conditions

4.1 Air Dose Rates

Figure 4 provides the frequency and cumulative frequency of air dose rates in undisturbed fields in the 80 km zone. Each color in the figure indicates data from each monitoring period (NRA, 2016a). It is clear that areas with air dose rates below 0.2 μSv/h have increased over the course of time. In 2015, about 90 percent of the area in the 80 km zone had air dose rates below 0.5 μSv/h. In four years, the average air dose rate in undisturbed flat fields had fallen to about a quarter of that in June 2011.

The temporal change in the average air dose rate is summarized for different conditions in Fig. 5 (NRA, 2016a). The average air dose rate measured above roads by car-borne surveys within the 80 km zone fell to one fifth of that in June 2011; the air dose rate is estimated to have fallen to a little over two fifths through physical decay of radiocesium. Air dose rates above roads have decreased much faster than the physical decay rate.

The results from person-borne surveys cannot be discussed in the same manner because those surveys started in 2013. Nevertheless, the air dose rates measured in the person-borne surveys, shown as living environment data in Fig. 5, were found to be generally greater than those by the car-borne surveys and smaller than those by the undisturbed field measurements in the same areas. Thus, it was found that air dose rates in environments related to various human activities had decreased markedly faster than physical decay. On the other hand, the measurements made in purely forested environments indicated that the air dose rates had decreased almost according to physical decay (NRA, 2015).

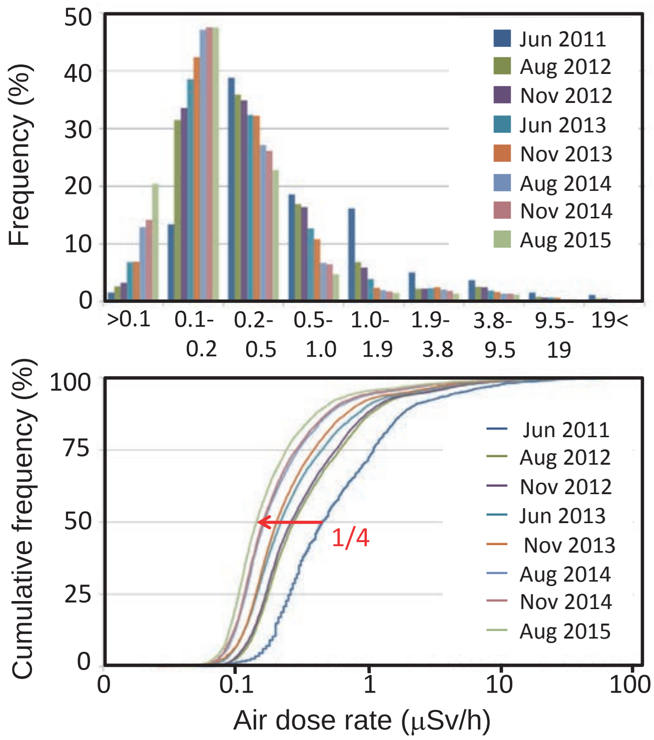

Fig. 4 Frequency distribution and cumulative frequency of air dose rates measured above flat fields in the 80 km zone. Data for eight occasions are indicated by different colors. (NRA, 2016a)

On the basis of a statistical analysis of car-borne survey data, two important factors affecting the air dose rate reduction were suggested: land use and human activities (Kinase *et al.*, 2014). Air dose rates in forests have decreased relatively more slowly, those in urban areas more quickly, and those in agricultural areas, moderately. The air dose rate reduction outside the evacuation zone has proceeded more quickly than that inside the zone even though the land use is the same. This suggests that not only decontamination but various other human activities tend to accelerate the air dose rate reduction. A detailed quantitative analysis of human activity effects on dose reduction will be a future challenge.

A predictive model of air dose rates was developed based on a statistical analysis of car-borne survey data (Kinase *et al.*, 2014). In the model, the air dose rates at one location were assumed to decrease according to an empirical formula, with two exponential functions representing a fast reduction component and a slow reduction component. The empirical formula was fitted to time-dependent air dose rates averaged over a 100 m square area. Fitting was conducted for about 14,000 areas of 100 m square, and the obtained parameters were statistically analyzed. The most suitable parameters for the prediction were determined

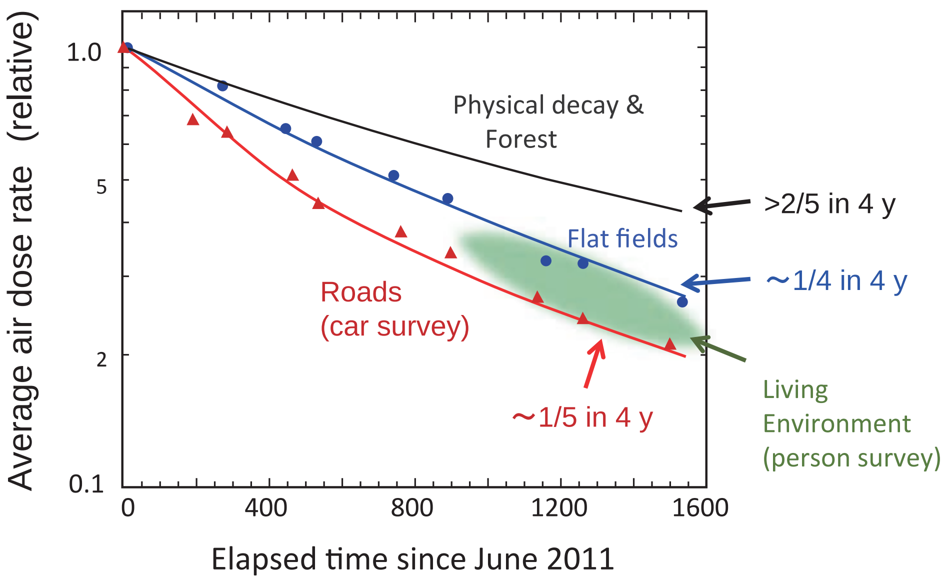

Fig. 5 Temporal change of the average air dose rate for different conditions. (NRA, 2016a)

considering the conditions in each area. Figure 6 demonstrates the differences in tendencies of predicted air dose rate reduction for two cases: one is a forested area in the evacuation zone and the other, an urban area outside the evacuation zone. The decreasing tendencies differ sharply, and the air dose rates in the latter case were predicted to decrease much faster.

4.2 Deposited Radionuclides

The time-dependent average deposition densities in undisturbed fields within the 80 km zone for Cs-134 and Cs-137 are illustrated separately in Fig. 7 (NRA, 2016a). The densities were normalized to that of Cs-137 in March 2012 when *in situ* measurements using a Ge detector started. The observed deposition densities have decreased with time in almost the same manner as the physical decay for both radionuclides. This suggests that horizontal movement of radiocesium is generally very slow in undisturbed fields. This finding coincides with the results from radiocesium migration studies carried out at several test plots in Fukushima (Yoshimura *et al.*, 2015).

Radiocesium deposited on the ground has gradually penetrated into deeper parts of the soil. There are two typical radiocesium concentration profiles in ground. One follows an exponential function, and the other is a distribution with a concentration peak at a certain depth. The latter profile was found to fit well to a hyperbolic secant function which asymptotically approached an exponential function in the deeper part (Matsuda *et al.*, 2015). Though the number of the latter profiles increased with time soon after the accident, that number seems to have hit a saturation point at present.

The 90-percent depth, defined as the depth up to which 90 percent of deposited Cs-137 is contained, is shown in Fig. 8 as a function of elapsed days (NRA, 2016a). The blue and red marks indicate whole data at about 80 locations in the 80 km zone, and the green marks indicate the geometric averages. The averages have gradually increased with time. The overall average, however, was still smaller than 5 cm in 2015, suggesting that removal of surface soil up to 5 cm is still effective for reducing air dose rates in decontamination work.

The air dose reduction in undisturbed flat fields

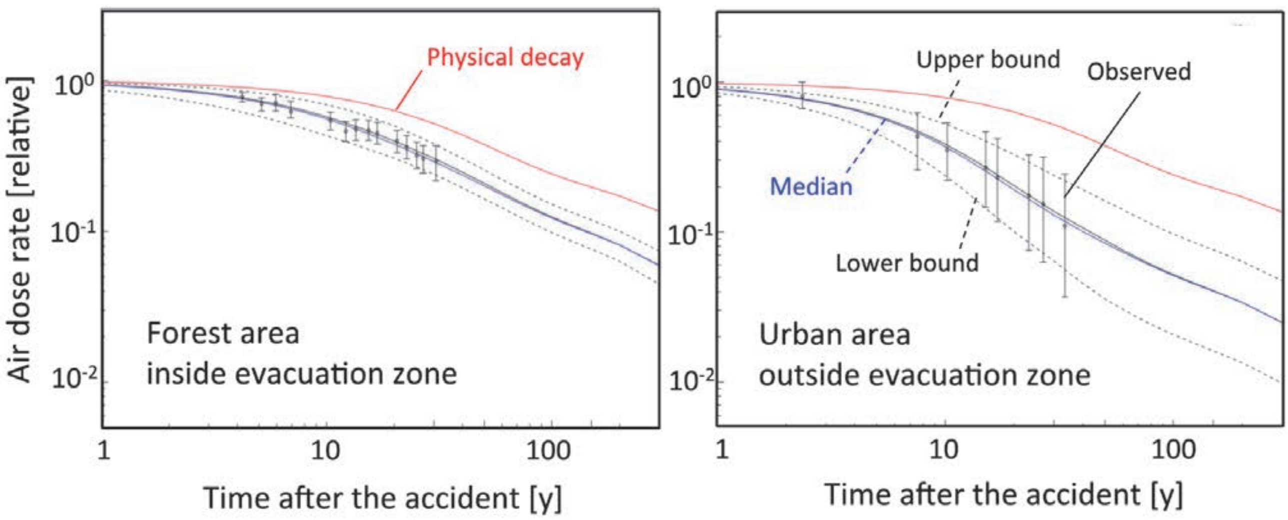

Fig. 6 Time-trends of predicted air dose rates for two different conditions. The upper bound and lower bound in the figure indicate the 90% confidential range of the prediction.

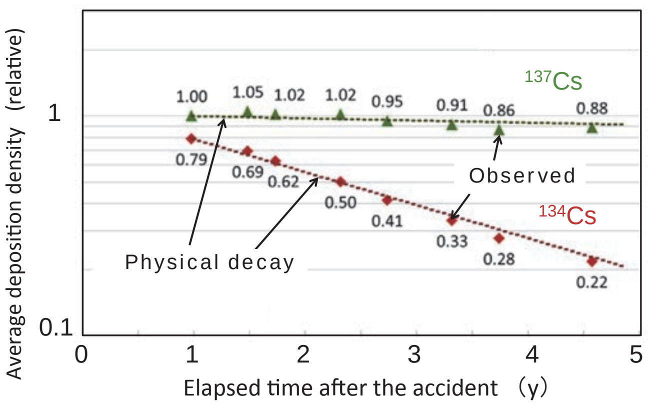

Fig. 7 Temporal change in the average deposition density in undisturbed fields within the 80 km zone. (NRA, 2016a)

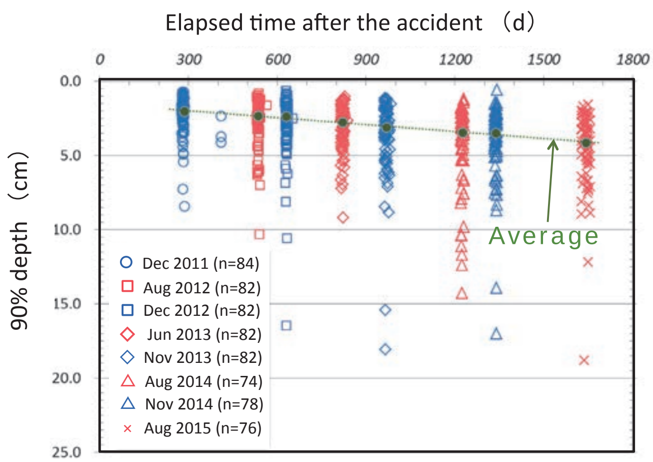

Fig. 8 Ninety percent depth of Cs-137 calculated from depth profiles observed in undisturbed fields in the 80 km zone. (NRA, 2016a)

proceeds faster than physical decay, as shown in Fig. 4, even though horizontal movement of radiocesium is very small. This can be explained by the penetration of radiocesium into the ground. The penetration results in an increased shielding effect for gamma rays, and the degree of air dose rate reduction was found to correspond to the increase of the shielding effect due to radiocesium penetration indicated in Fig. 8.

It turns out that radiocesium deposited in forest areas moves inside the forest system and hardly escapes from the system. Radiocesium deposited on tree crowns has been falling down to the ground level with throughfall, litter-fall and stemflow; and at present a large part of the deposited radiocesium is thought to exist near the ground surface (Kato *et al*., 2015). On the other hand, it was confirmed that radiocesium deposited on artificial structures such as roads and houses tends to be easily removed, and the radiocesium concentration on artificial structures has become much smaller than that in the surrounding environment (Yoshimura *et al*., 2016).

5. Summary

Since the Fukushima accident, large-scale environmental monitoring activities have been successfully implemented with the efforts of a number of people, even though preparedness for emergency monitoring over the long-term was insufficient before the accident. The monitoring resulted in an accumulation of enormous amounts of environmental monitoring data, and these data made it possible to analyze the regional and temporal characteristics of contamination conditions around the Fukushima NPP site with precision.

Air dose rates in environments related to various human activities were found to decrease at a much faster rate than physical decay; while, in pure forest, the air dose rate reduction is close to physical decay. Movement of radionuclides deposited in natural environments is generally slow; while, radiocesium deposited on artificial structures is removed faster. Further, human activities seem to accelerate the air dose rate reduction. On the basis of a statistical analysis of a large number of observation data, an empirical model for predicting air dose rates was developed. Its accuracy of prediction is expected to improve as further monitoring data are accumulated.

Accumulated environmental monitoring data have been provided to the public in different ways. The extended radiation map site on the Internet has presented distribution maps on air dose rates and radionuclide deposition densities obtained from the mapping projects and also by aerial monitoring (NRA, 2016b). JAEA developed a new database aggregating various monitoring data obtained by national projects, local governments and research institutes (JAEA, 2016). The aggregated data are presented in several different ways such as numerical data, maps and graphs indicating time trends together with some simple analytical software.

Large-scale monitoring should be continued while optimizing scale and frequency, since the radiation levels are still high in certain areas and future trends must be properly perceived. Further, the accumulated data should be thoroughly analyzed to unveil novel information on radiological environments after a severe accident.

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Kimiaki SAITO

Kimiaki Saito has been working on national projects involving large-scale environmental monitoring and characterization of radiological conditions around the Fukushima site. He started his career with measurement and evaluation of environmental radiation. His main research interest has been dosimetry using human models and Monte Carlo simulation for radiation protection purposes. In recent years, he has conducted simulation studies on biological radiation effects, also extending his activities into the medical field.

Hideaki YAMAMOTO

Hideaki Yamamoto is principal engineer at the Fukushima Environmental Safety Center, Japan Atomic Energy Agency (JAEA). He is also a member of the Japan Health Physics Society.

Satoshi MIKAMI

Satoshi Mikami is an assistant principal engineer with JAEA. Since 1992, he has worked mainly in maintenance in the field of reference calibration of radiation-protective monitoring instruments, and in airborne monitoring at the Tokai Reprocessing Plant. He has also studied photon calibration at IRSN, France for one year from September 2003. Since December 2011, Mr. Mikami has been involved in the national mapping projects after the Fukushima accident, including repeated large- scale environmental monitoring for clarifying the distributions of radionuclide deposition densities and air dose rates over wide areas and their time- dependent tendencies.

Masaki ANDOH

Masaki Andoh is a researcher with the Fukushima Environmental Research Group at JAEA. He is currently in charge of car-borne surveys using the KURAMA system.

Norihiro MATSUDA

Norihiro Matsuda is a researcher with the Fukushima Environmental Research Group at JAEA. His main fields are radiation shielding and development of radiation transport simulation code.

Sakae KINASE

Sakae Kinase is a principal scientist at JAEA, and a visiting professor at Ibaraki University.

Shuichi TSUDA

Shuichi Tsuda works with the Research Group for Radiation Transport Analysis at JAEA. His fields of specialty are microdosimetry and radiation measurement.

Tadayoshi YOSHIDA

Tadayoshi Yoshida is an assistant principal engineer with the Fukushima Safety Administration and Radiation Measurement Group at JAEA. His main duties are research on radiation dosimetry and maintenance of radiation monitoring instruments.

Shinichiro MATSUMOTO

Shinichiro Matsumoto’s experience includes fuel design for the Monju Nuclear Power Plant, performing examinations in post irradiation examination facilities, managing radioactive waste treatment facilities, and other duties for JAEA. After the Great East Japan earthquake, he helped elucidate the distribution of widely diffused radioactive materials. He is currently engaged in fuel debris extraction for IRID.

Tetsuro SATO

Tetsuro Sato is a systems engineer at Hitachi Solutions East Japan, Ltd. He is assisting JAEA research using his ICT skills. He operates their system for collecting environmental monitoring data and helps them analyze the monitoring data.

Akiyuki SEKI

Akiyuki Seki is a researcher at JAEA. He completed the master's course at the Graduate School of Science and Engineering, Ibaraki University, Japan. He worked on performance evaluation of supercomputer systems and optimization of numerical simulation codes for supercomputer systems at the Tokai Research and Development Center. After the Fukushima Daiichi nuclear accident in 2011, he developed a radionuclide distribution database system. The radionuclide distribution data, exceeding 4 hundred million data points, are contributing to countermeasure planning and providing relief to concerned residents.

Hiroshi TAKEMIYA

Hiroshi Takemiya is deputy director at the Center for Computational Science and e- Systems at JAEA. He is in charge of constructing an environmental monitoring database and analyzing the monitoring data.

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