Detailed deposition density maps constructed by large-scale soil sampling for gamma-ray emitting radioactive nuclides from the Fukushima Dai-ichi Nuclear Power Plant accident

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Article info

Article history:
Received 27 November 2013
Received in revised form 5 February 2014
Accepted 19 February 2014
Available online 2 April 2014

Keywords:
Deposition density maps
Soil sampling
Gamma-ray emitting radioactive nuclides
Gamma-ray spectrometry
Fukushima Dai-ichi NPP accident

Abstract

Soil deposition density maps of gamma-ray emitting radioactive nuclides from the Fukushima Dai-ichi Nuclear Power Plant (NPP) accident were constructed on the basis of results from large-scale soil sampling. In total 10,915 soil samples were collected at 2168 locations. Gamma rays emitted from the samples were measured by Ge detectors and analyzed using a reliable unified method. The determined radioactivity was corrected to that of June 14, 2011 by considering the intrinsic decay constant of each nuclide. Finally the deposition maps were created for $^{134}$Cs, $^{137}$Cs, $^{131}$I, $^{129}$mTe and $^{110}$mAg. The radioactivity ratio of $^{134}$Cs to $^{137}$Cs was almost constant at 0.91 regardless of the locations of soil sampling. The radioactivity ratios of $^{131}$I and $^{129}$mTe to $^{137}$Cs were relatively high in the regions south of the Fukushima NPP site. Effective doses for 50 y after the accident were evaluated for external and inhalation exposures due to the observed radioactive nuclides. The radiation doses from radioactive cesium were found to be much higher than those from the other radioactive nuclides.

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1. Introduction

Enormous amounts of radioactive nuclides were released into the atmosphere from the Fukushima Dai-ichi Nuclear Power Plant (NPP) accident (Prime Minister of Japan and His Cabinet, 2011). The released radioactive nuclides were deposited on the ground, trees, houses and other objects over a wide area. Monitoring data obtained by various organizations after the accident indicated that large amounts of radioactive nuclides were deposited in the environment, which significantly increased the radiation levels. In the early stage after the accident, the general public was exposed to significant levels of radiation from $^{131}$I with a half-life of 8 d. Hereafter, several radioactive nuclides with long half-lives remain on the ground for a long time and continue to expose the public to radiation, as was the case in the Chernobyl accident (IAEA, 1991, 2006; MUE, 2011).

In the face of this serious crisis, many researchers insisted that comprehensive large-scale environmental monitoring of the contaminated areas should be conducted as soon as possible. As a result of the great efforts made by many people in different positions, the mapping project to investigate environmental contamination conditions and migration of radioactive nuclides in the environment was officially begun on June 6, 2011 with the support of the Strategic Funds for Promotion of Science and Technology. Many people and organizations contributed to the considerable preparations and discussions for starting the project despite the limited time.

One of the main goals of the mapping project was to produce precise distribution maps of dominant gamma-ray emitting...
radioactive nuclides deposited on the ground. There are following two methods for that purpose: 1) collecting soil samples and measuring gamma rays using Ge detectors in the laboratories; and 2) in situ spectrometry using portable Ge detectors. The latter method has the advantage of quantifying the average deposition density of radioactive nuclides by detecting the gamma rays coming from a wide area around the measurement location (ICRU, 1994). However, in the beginning of the project, we could not prepare sufficient numbers of portable Ge detector systems for in situ measurements and technicians who could properly operate the systems, since the importance of in situ spectrometry had not been, so far, sufficiently recognized in Japan. On the other hand, collecting soil samples dose neither need complex equipment nor a high level of skill. Furthermore, soil samples are essential because they can give precious information on deposited radioactive nuclides such as chemical forms or soil characteristics.

Thus, we decided to collect a large number of soil samples for analysis with gamma-ray spectrometry using Ge detectors at many laboratories. The purpose of this paper is to provide a detailed description on how radioactive nuclide deposition density maps were constructed in the first campaign of the mapping project. The mapping of deposition densities of gamma-ray emitting radioactive nuclides were completed for $^{134}\text{Cs}$, $^{137}\text{Cs}$, $^{131}\text{I}$, $^{129}\text{mTe}$, and $^{110}\text{mAg}$. Small amounts of other radioactive nuclides were detected in multiple soil samples; however, the number of detected samples for these nuclides was not sufficient to be shown on a map. On the basis of the constructed maps, the features of the regional distributions of the radioactivity ratios among observed radioactive nuclides were examined to get basic information for understanding deposition pathways of radioactive nuclides. The maps for strontium and plutonium will be presented elsewhere (Ikeuchi et al., submitted for publication). The maximum effective doses for 50 y were conservatively estimated for all of the radioactive nuclides mapped in the first campaign including strontium and plutonium.

2. Materials and methods

2.1. Selection of sampling locations

The soil sampling locations were selected on the basis of the following criteria. Based on the radiation levels roughly clarified by prior environmental monitoring including air-borne survey (MEXT, 2011), the sampling locations were selected within a 100 km radius of the Fukushima NPP site and throughout the rest of Fukushima Prefecture. The region within 80 km of the site was divided into 2 km square grids, and the rest was divided into 10 km square grids. A suitable location was chosen within each grid considering the ground condition and other conditions as described below. In the case that two municipalities were in one grid, two sampling locations were selected to cover both municipalities. Non-inhabitable areas were avoided since our direct concern was to check the contamination levels of living conditions.

Inhomogeneously contaminated locations were avoided in order to obtain sample data representing regional contamination;
Thus, flat fields of a certain width and little vegetation were selected. The intention was to continue the periodical environmental monitoring at identical locations to investigate the time-dependent changes in the contamination conditions due to weathering effects. Thus, locations where the geographic conditions were expected not to change for a long period of time were chosen: for example, farm fields were avoided since they would be often plowed. We also avoided riverside locations since floodwaters may significantly change the deposition conditions. With regard to land-ownership, public lands were preferred to private lands, because it was easier to obtain permission for soil sampling.

Since there was insufficient time to directly check the sampling locations, they were selected by carefully checking maps overlaid 2 or 10 km square grids. After the sampling locations were determined, we obtained permission for soil sampling from all municipalities with related jurisdiction. When the conditions of the sampling location were not clear, staff of the project visited the location beforehand to examine the conditions with the help of Fukushima University.

2.2. Soil sampling

More than 400 volunteers from more than 90 organizations took part in the soil sampling from June 4 to July 8, 2011. Huge numbers of soil sampling kits and consumable goods were prepared by Osaka University before the project officially started. Each soil sampling team had two or three members including at least one specialist in radiation or radioactivity. Each team collected soil samples at several locations per day. Every day, up to 30 teams were engaged in soil sampling. In the 20 km zone where entry was restricted, we asked the Federation of Electric Power Company of Japan, which was responsible for routine monitoring in the zone, to support the soil sampling.

The soil sampling procedures, being described in detail in another paper (Onda et al., submitted for publication), are briefly outlined here. To ensure the quality of the soil sampling and spectrometry, the procedures were carefully pre-examined beforehand. The protocol describing the standard methods for soil sampling and gamma-ray spectrometry was determined after the critical discussions with specialists and was approved by the Committee on the Construction of Maps for Radiation Dose Distribution (MEXT, 2012). In the campaign, all of the soil sampling and spectrometry were carried out according to the protocol. Some of the planned sampling locations could not be reached because the access roads had been severely damaged by the earthquake or tsunami. Overall, soil sampling was performed at 2138 locations.

After the preliminary soil sampling and analysis, most of the deposited radioactive nuclides were concluded to exist less than 5 cm in depth from the ground surface (Onda et al., submitted for publication). Therefore, the surface soil was dug up to 5 cm depth.

Fig. 2. Deposition density map for $^{137}$Cs. The radioactivity per unit ground area is shown by the colored mark at the soil sampling location.
by a specific tool. Collected soil was sufficiently mixed to ensure reliable measurements since the detection systems were calibrated assuming that radioactivity was uniformly distributed in the container. The sampled soil was put into a plastic container with a diameter of 5 cm and a height of 5 cm (U-8 type). In principle, five soil samples were collected at one location in order to compensate for large deviation in the deposited radioactivity with the sampling point. At locations where sampling was not easy, soil samples less than five were collected. In total, 10,915 soil samples were collected.

2.3. Spectrometry and data analysis

Twenty-one organizations performed gamma-ray spectrometry with Ge detectors. Some of the organizations were not expertized in measuring environmental samples. Therefore, preliminary tests using mock-up samples were carried out to check the fundamental workability of measurement systems. In addition, the measured results for same soil samples were compared by the participating organizations and found to be in good agreements; thus the reliability of the measurements was ensured.

The spectrometry systems were calibrated using the standard sources provided by the International Atomic Energy Agency (IAEA) and the Japan Chemical Analysis Center (JCAC). Observed peak counts in the spectrometry were converted to radioactivity according to the calibration data if they were above the detection limits. All organizations were required to always report the measurement results for $^{134}\text{Cs}$, $^{137}\text{Cs}$ and $^{131}\text{I}$. The results for other radioactive nuclides were supposed to be reported when they were detected. The determined radioactivities were averaged over all of the soil samples collected at one location, which was usually five samples. The averaged radioactivity was converted to radioactivity per unit ground area (Bq/m$^2$).

Radioactive nuclides $^{134}\text{Cs}$ and $^{137}\text{Cs}$ were detected at every sampling location. In other radioactive nuclides, statistically significant data were obtained from soil samples at a limited number of locations because of short half-lives and deposited radioactivity levels. When statistically significant data were obtained for at least one soil sample at one location, deposition was judged to have occurred at the location, and the average deposition density was estimated.

2.4. Air dose rate measurements

Air dose rates were measured using survey meters at the same locations where the soil samples were collected. A NaI(Tl) scintillation survey meter TCS-171B/172B manufactured by Hitachi-Aloka Medical, Ltd. was used. This survey meter can accurately measure the ambient dose equivalent rates even if the photon spectrum

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**Fig. 3.** Deposition density map for $^{131}\text{I}$. The radioactivity per unit ground area is shown by the colored mark at the soil sampling location.
varies by utilizing a spectrum-dose conversion function (G(E) function) developed at JAEA (Tsutsumi et al., 1991) based on a reliable detector response function (Saito and Moriuchi, 1981, 1987). At locations where the radiation levels were higher than 30 μSv/h which was the upper limit for accurate measurements by the NaI(Tl) survey meter, an ionization chamber-type survey meter was used. The survey meters were required to have been calibrated within the last year.

2.5. Mapping

The average radioactivity per unit ground area was obtained as described in Section 2.2 and plotted on a map based on positional data obtained by the Global Positioning System (GPS). The data were not interpolated to make contour maps since such interpolation sometimes produces inappropriate artificial data. In addition, soil sampling locations are often not explicitly indicated on the contour maps. Maps issued by the Geospatial Information Authority of Japan were used as base maps.

Ranges of the nuclide deposition density were indicated on the map by different colors at sampling locations. When the analysis of the whole collected samples showed statistically significant radioactivity, the average value was plotted with a circle. While, in case that a part of the collected soil samples showed gamma-ray intensity below the detection limit, the average value was plotted with a triangle.

3. Results and discussion

3.1. Deposition density maps

Figs. 1–5 show the maps for deposition densities of 134Cs, 137Cs, 131I, 129mTe and 110mAg, respectively. The radioactivity per unit ground area (Bq/m²) on June 14, 2011 is indicated by a colored mark at the soil sampling locations. The major soil sampling campaign was completed on this date; thus, all radioactivities were corrected to this day based on the decay half-life of the corresponding nuclide. These numerical data have been provided through a database developed in the mapping project (NRA; Seki et al., submitted for publication).

The basic features of the 134Cs maps were quite similar to those of 137Cs, since the radioactivity ratio of 134Cs/137Cs was almost constant at 0.91 on June 14, 2011 as shown in Fig. 6. This figure suggests that the isotope ratio of 134Cs/137Cs was similar among the plumes released from the three reactors in the Fukushima accident.

Figs. 1 and 2 indicate that the area northwest of the Fukushima NPP site was most contaminated; further, the strip-shaped Fukushima Basin running from north to south in the center of Fukushima Prefecture showed relatively high contamination. High deposition densities of 131I, 129mTe and 110mAg were also observed northwest of the Fukushima NPP site. However, there were insufficient data for 131I, 129mTe and 110mAg to discuss the features of the deposition density distribution in detail.

Fig. 4. Deposition density map for 129mTe. The radioactivity per unit ground area is shown by the colored mark at the soil sampling location.
3.2. Radioactivity ratios of $^{131}$I, $^{129m}$Te and $^{110m}$Ag

Figs. 7–9 give the distribution maps of the radioactivity ratios of $^{131}$I, $^{129m}$Te and $^{110m}$Ag, respectively, to $^{137}$Cs. The regional distributions of the ratios indicated similar tendencies for $^{131}$I and $^{129m}$Te. The ratios were relatively high at the coastal regions south of the Fukushima Dai-ichi NPP site compared with the other regions.

Figs. 10 and 11 give the scatter plots of the deposition densities for $^{131}$I and $^{129m}$Te as a function of those for $^{137}$Cs. The correlation of $^{131}$I–$^{137}$Cs was examined by fitting the data points to the regression line over the whole area, the southern coastal area, and the remaining area as shown in Fig. 10(a)–(c), respectively. Fair correlations were observed in the southern coastal area and remaining area as the coefficients of the determination ($R^2$) indicated relatively high values.

For $^{129m}$Te, the scatter plot for the whole area (Fig. 11(a)) shows good correlation with $^{137}$Cs. If we enlarge the low deposition density part of Fig. 11(a), another correlation between $^{129m}$Te and $^{137}$Cs can be found as shown in Fig. 11(b). Here, the data with the ratio of $^{129m}$Te–$^{137}$Cs greater than 1.0 are plotted with circles, separately from the other data shown by the triangles. The circles in Fig. 11(b) correspond to the bright colored circles in the southern coastal area in Fig. 8. The data in this area showed a different correlation compared to the other region.

The deposition densities for the radioactive nuclides of $^{131}$I and $^{129m}$Te both showed a good correlation with that of $^{137}$Cs in the southern coastal areas. Those in the other regions showed different...
correlations. This suggests that at least two dominant pathways for contamination existed due to different radioactive plumes or deposition processes.

According to the review of atmospheric diffusion simulations carried out after the accident (Yamazawa and Hirao, 2012), the radioactive nuclide deposition in the southern coastal areas was due to plumes released on March 14 and 15 and after March 20. The monitoring data in Ibaraki Prefecture to the south indicated relatively high fractions of $^{131}$I in the radioactive plumes during these periods (Furuta et al., 2011). The observed data in the present study were consistent with the results from these simulations and observations.

The ratio of $^{110m}$Ag to $^{137}$Cs seemed to be high for the whole region along the coast. It was difficult to find specific features in the scatter plot. However, some features were clarified in the second campaign covering wide areas in eastern Japan, which will be discussed in another paper (Mikami et al., submitted for publication).

### 3.3. Air dose rate maps

The distribution of air dose rates measured from June 4 to July 10, 2011 is presented in Fig. 12. All of the measurements were made in terms of ambient dose equivalent rates. No correction was performed for the physical decay of radioactive cesium during the period, since the temporal variation of air dose rates during the measuring period was estimated to be negligible compared with other uncertainties. The distribution of the air dose rates was quite similar to that of the radioactive cesium deposition density because radioactive cesium was the dominant contributor to the air dose rates. More than 99% of the air dose rate was estimated to be due to radioactive cesium as discussed later.

The correlation of the air dose rates to $^{137}$Cs deposition densities was not so good as shown in Fig. 13. This implies that the radioactive cesium deposition was heterogeneous and greatly varied even within a small area of a few square meters. In fact, the variation coefficient of the cesium radioactivity concentration for the five soil samples collected at one location greatly varied with the average of 36% as indicated in Fig. 14. On the other hand, air dose rates were found to have a good correlation with cesium deposition densities measured by in situ measurements using portable Ge detectors as discussed in another paper (Mikami et al., submitted for publication). Consequently, the deposition density of radioactive nuclides obtained by soil sampling should be noted to have some degree of statistical fluctuation.

### 3.4. Dose evaluation

The effective doses accumulated over 50 y after the accident due to deposited radioactive nuclides were roughly estimated using the dose conversion coefficients presented by IAEA (2000).
conversion coefficients estimate the external exposure dose and internal exposure dose due to inhalation of re-suspended radioactive nuclides under the following simplified assumptions: a person stands on the ground where radioactive nuclides are deposited; the physical decay of deposited radioactive nuclides and weathering effects are roughly considered; a conservative initial re-suspension rate of $10^{-6}$ is used. Then you should note that the evaluations were very conservative and the public is unlikely to receive such high doses in a real environment. Here, we examined the postulated maximum exposures due to the deposited radioactive nuclides.

The evaluated effective doses over 50 y are given in Table 1 for the radioactive nuclides detected in the first campaign. Doses were also evaluated for plutonium and strontium; these were quantified in another study (Ikeuchi et al., submitted for publication). The radioactive nuclide $^{137}$Cs was estimated to cause the most significant exposure. The second important radioactive nuclide was $^{134}$Cs which produces the dose an order of magnitude smaller than that from $^{137}$Cs. The third was $^{110m}$Ag, which had the dose three orders of magnitude smaller than that of $^{137}$Cs. The doses due to strontium and plutonium were negligible in comparison with the doses from the other radioactive nuclides.

We evaluated the relative contributions of the observed radioactive nuclides to the external effective dose rates on June 14, 2011. The radioactive nuclides in soil were assumed to have depth profiles approximated by an exponential function with a relaxation mass per area of 1 g/cm$^2$. The dose conversion coefficients were taken from Saito et al. (2012). In this evaluation, the averaged deposition densities over the fifty typical locations with different radiation densities were used to represent typical external exposure conditions. Table 2 provides the evaluated results for June 14, 2011. About 70% of the external effective dose was due to $^{134}$Cs, and 30% was due to $^{137}$Cs. The contributions from the other nuclides were less than 1%.

4. Conclusions

The ground deposition densities of gamma-emitting radioactive nuclides were determined by collecting a large number of soil samples around the Fukushima Dai-ichi NPP site. All of the collected soil samples were carefully analyzed using Ge detector spectrometry. Special attention was paid for performing the soil sampling and analysis by reliable and standardized methods as quickly as possible in order to detect radioactive nuclides with short half-lives. In particular, $^{131}$I had the potential to expose the public to significant radiation in the early stage after the accident. Maps were constructed for the critical gamma-ray emitting nuclides of $^{134}$Cs, $^{137}$Cs, $^{131}$I, $^{129m}$Te, and $^{110m}$Ag. The radioactivity ratios for the observed radioactive nuclides indicated specific regional features. The ratios of $^{131}$I and $^{129m}$Te-$^{137}$Cs were high in the regions south of the Fukushima NPP site. These regional features of radioactivity ratios were inferred to be due to the different
deposition pathways of radioactive nuclides. Therefore, they are important for extracting essential information to clarify the deposition pathways after the accident. The effective doses received by a person from external and inhalation exposures to the deposited radioactive nuclides were conservatively estimated. The largest effective doses were derived from $^{137}\text{Cs}$, followed by $^{134}\text{Cs}$. The doses from the other radioactive nuclides were three orders of magnitude smaller than that by $^{137}\text{Cs}$. The external effective dose on June 14, 2011 was estimated to mostly be due to $^{134}\text{Cs}$ and $^{137}\text{Cs}$. The constructed deposition density maps of radioactive nuclides will provide basic information on the initial contamination conditions around the Fukushima Dai-ichi NPP site.

Fig. 9. Distribution of the radioactivity ratio between $^{110}\text{mAg}$ and $^{137}\text{Cs}$. The ratio was calculated for all locations where $^{110}\text{mAg}$ was detected.

Fig. 10. Scatter plot of $^{131}\text{I}$ and $^{137}\text{Cs}$ deposition densities: (a) in the whole area, (b) in the southern coastal area, and (c) in the rest area. The regression line fitted to the plotted data and the coefficient of determination ($R^2$) are shown in each figure.
Fig. 11. Scatter plot of $^{129m}$Te and $^{137}$Cs deposition densities: (a) in the whole area and (b) in the southern area. In (b), data are shown by closed circles if the activity ratio of $^{129m}$Te and $^{137}$Cs was greater than 1.0 and by triangles otherwise. The regression line was fitted to the data shown by the closed circles in (a) and (b). The coefficient of determination ($R^2$) is also shown in each figure.

Fig. 12. Air dose rate map at the soil sampling locations. The ambient dose equivalent rate as measured by a survey meter calibrated within one year is shown by the colored mark at the soil sampling location.
samples collected at one location. The coefficients of determination ($R^2$) are shown in the figure.

Table 1

Effect of dose rates using the dose conversion coefficients (Saito et al., 2012).

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Contribution to effective dose (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{134}$Cs</td>
<td>70.9</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>28.1</td>
</tr>
<tr>
<td>$^{129m}$Te</td>
<td>0.6</td>
</tr>
<tr>
<td>$^{110m}$Ag</td>
<td>0.3</td>
</tr>
<tr>
<td>$^{131I}$</td>
<td>0.1</td>
</tr>
</tbody>
</table>

### Acknowledgment

This project was financially supported by the Ministry of Education, Culture, Sports and Science (MEXT). We express our hearty thanks to all of the people who directly and indirectly supported the project. Without their support, this large-scale project would have been impossible to achieve within a short time after the Fukushima Dai-ichi Nuclear Disaster. We are much obliged to Dr. Nakamura and all of the members of the Committee on the Construction of Maps for Radiation of Maps for Radiation Dose Distribution for their useful and encouraging comments. We are also indebted to Dr. Takahashi, Dr. Namba and their stuff of Fukushima University for helping arrangement of soil collection locations.

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