Assessment of individual radionuclide distributions from the Fukushima nuclear accident covering central-east Japan

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A tremendous amount of radioactivity was discharged because of the damage to cooling systems of nuclear reactors in the Fukushima No. 1 nuclear power plant in March 2011. Fukushima and its adjacent prefectures were contaminated with fission products from the accident. Here, we show a geographical distribution of radioactive iodine, tellurium, and cesium in the surface soils of central-east Japan as determined by gamma-ray spectrometry. Especially in Fukushima prefecture, contaminated area spreads around Iitate and Naka-Dori for all the radionuclides we measured. Distributions of the radionuclides were affected by the physical state of each nuclide as well as geographical features. Considering meteorological conditions, it is concluded that the radioactive material transported on March 15 was the major contributor to contamination in Fukushima prefecture, whereas the radioactive material transported on March 21 was the major source in Ibaraki, Tochigi, Saitama, and Chiba prefectures and in Tokyo.

gamma-ray spectrometry | individual radioactivity | deposition

A great earthquake with a magnitude of 9.0 occurred in the northwest Pacific off northeastern Japan on March 11, 2011. Until the earthquake occurred, units 1, 2, and 3 in the Fukushima No. 1 nuclear power plant, located in Futaba, Fukushima, Japan, had been supplying electric power under normal commercial operations. However, cooling functions for the reactors were lost because of damage by the earthquake. As a result of water decomposition by contact with hot fuel rods, hydrogen gas was evolved and accumulated in a reactor building. The resulting hydrogen explosion caused the collapse of outer walls of reactor buildings for units 1, 2, and 3. In addition, on March 12 exhaust ventilation from a nuclear reactor container was carried out to decrease pressure. The details of the timing and causes of the radioactive releases are described elsewhere* (http://www.mext.go.jp/english/incident/1304796.htm). The majority of the airborne fission product releases are reported to have been released through hydrogen explosions, venting, and leakage over March 2011. Consequently, high volatility fission products including ^{129}mTe, ^{131}I, ^{134}Cs, ^{136}Cs, and ^{137}Cs were discharged into the environment. These radioactive elements were carried together with the air parcel, and subsequent wet and dry depositions caused accumulation of them on the ground. Radioactive iodine, strontium, and cesium, which have large fission yields and low boiling points, present a large risk for internal radiation exposure via ingestion of contaminated agricultural crops. These predictions also require an estimation of the emission inventory as well as knowledge of the distributions of various radionuclides and their transport and deposition on the ground. Therefore, we carried out gamma-ray spectrometry of surface soil samples collected in central-east Japan to contribute to this scientific enterprise.

Results and Discussion

Geographical distributions of radionuclides in a unit area are shown in Fig. 1 together with a topographic map. Distributions of activity ratios are also shown. Focusing on geographical features in Fukushima prefecture, Iitate is in a hill district 300 m in altitude. Naka-Dori is a basin-shaped valley that extends from north to south in Fukushima prefecture with a mountainous district lying between it and the Fukushima No. 1 nuclear power plant. Very high accumulation was observed in the Iitate and Naka-Dori regions for all of the radionuclides. In the other regions, ^{131}I was rather homogeneously distributed. However, low accumulations of Cs isotopes were found in the region between northern Ibaraki and eastern Saitama. For ^{129}mTe, the amount of accumulation was high in Fukushima but low in Ibaraki, Tochigi, Saitama, and Chiba prefectures and in Tokyo. The activity ratio of ^{134}Cs/^{137}Cs was found to be 0.8–0.9 in the whole investigated region, including Fukushima prefecture. No different behavior was found among the Cs isotopes. The amount of fuel burnup was similar for units 1, 2, and 3, and therefore the radioisotope mix for the cesium isotopes is similar for the three units. However, different geographical distributions among I, Te, and Cs were found, as seen in Fig. 1.

During the major radionuclide emission periods from the accident in March 2011, two rainfall events took place over central-eastern Japan. It is well-known that most aerosols and water-soluble gases are washed out by a rainfall (1–3). According to the automated meteorological data acquisition system (AMeDAS) (http://www.jma.go.jp/jma/indexe.html), continual rainfall occurred between 1700 hours on March 15 and 0400 hours on March 16 in northern Fukushima prefecture and between 0800 hours on March 21 and 0600 hours on March 23 in Ibaraki, Chiba, Tochigi, and Saitama prefectures and in Tokyo (Fig. 2). Based on a simulation using the hybrid single particle Lagrangian integrated trajectory model, version 4.9 (HYSPLIT) with meteorological data from the global data assimilation system (4), the air parcel from the Fukushima No. 1 nuclear power plant, with an altitude of 0–1,000 m, stayed in Fukushima at 1700 hours on

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N.K. notes that sampling and measurement were done while at University of Tsukuba, and data analyses (calculation of radioactivity and drawing of contour map of radioactivity) were performed while at Shimizu Corporation.

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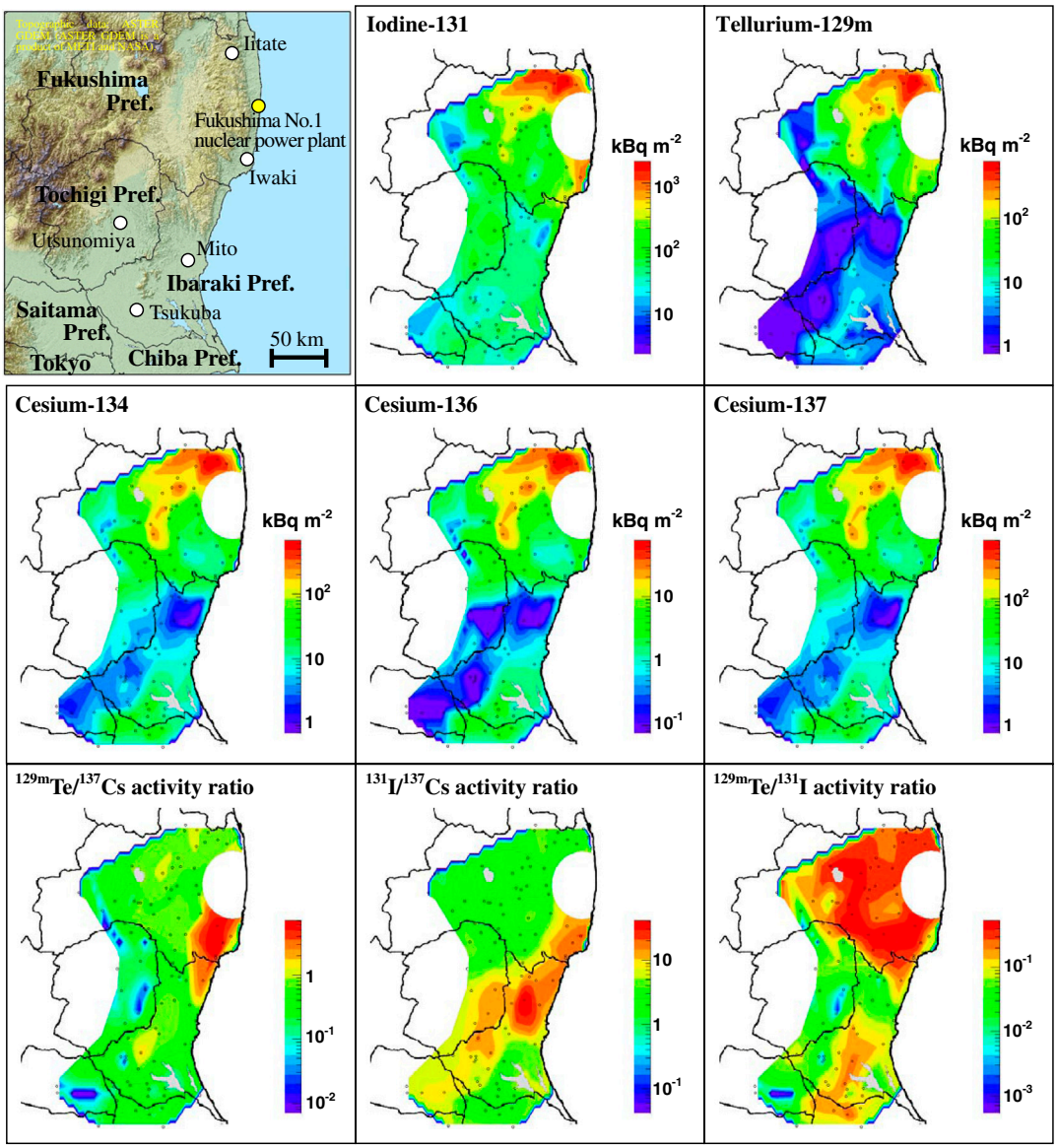

Fig. 1. Topographic map of the surveyed area including Fukushima and its adjacent prefectures; contour maps of depositions for ^{131}I, ^{129m}Te, and ^{134,136,137}Cs; and activity ratios for ^{129}mTe/^{137}Cs, ^{131}I/^{137}Cs, and ^{129}mTe/^{131}I. Activities on March 29, 2011 are shown. Open circle indicates position of sampling point.

March 15 and in Ibaraki and Chiba at 1100 hours on March 21. The air parcel from the plant remained in the relevant region before raining. Thus, radionuclides in the air were washed out by the rainfall. Most of the radionuclides were deposited on March 15 in Fukushima prefecture and on March 21 in Ibaraki, Tochigi, Saitama, and Chiba prefectures and in Tokyo. In Naka-Dori, it is believed that it was difficult to move the air parcel because of the basin-shaped valley; deposition tended to be higher. However, the higher accumulation in Iitate and Naka-Dori cannot be explained by the HYSPLIT model alone. More precise model analysis combined with detailed local meteorology is required for the local-scale simulation.

On the other hand, nonvolatile elements such as Te and Cs adsorb onto aerosols (5). Both particulate and gaseous states have been recognized for atmospheric iodine (6). There is a mountain area with an altitude of approximately 500 m between northern and central Ibaraki prefecture. According to the HYSPLIT simulation, the air parcel moved from the Fukushima No. 1 nuclear power plant to the southwest. It is considered that transport of particulate material from the accident to eastern Saitama prefecture was blocked by the mountainous area. Gaseous materials were more easily transported than particulates, even through the mountainous area.

Cesium-137 deposition continuously occurred for more than 30 y as a result of atmospheric nuclear tests (7). The annual deposition rate of ^{137}Cs in Japan was measured at the Meteorological Research Institute (8, 9). Annual ^{137}Cs flux of 100–1,000 Bq/m^2 per year was observed during the 1950s and 1960s, whereas 10 Bq/m^2 per year was observed during the 1970s. Approximately 7 kBq/m^2 of ^{137}Cs (simple summation of the monthly flux) had been deposited in the Pacific side of Japan before the Fukushima accident. The Fukushima accident produced 0.4–900 kBq/m^2 of deposition for ^{137}Cs within the present survey area; these values are 0.06–130 times the cumulative deposition from the atmospheric nuclear test. At the beginning of July 2011, a dose rate of 0.6 μSv/h was detected 23 km south from the Fukushima No. 1 nuclear power plant, where deposition of 140 kBq/m^2 of ^{131}I and 80 kBq/m^2 of ^{137}Cs were determined on March 29. The environmental external radiation dose rate before the accident was approximately 0.05 μSv/h. Dominant

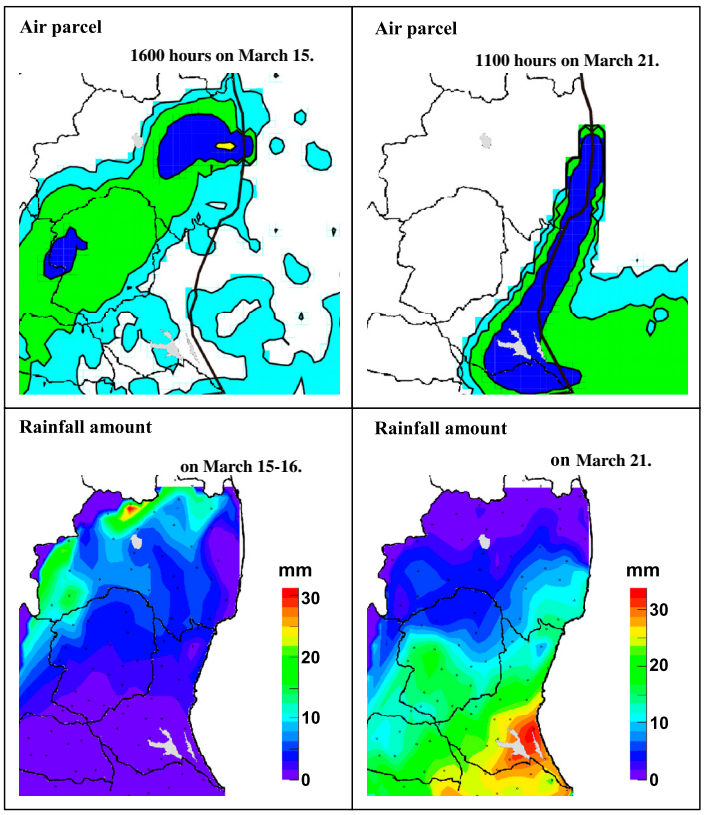

Fig. 2. Distributions of the air parcel that passed through the Fukushima No. 1 nuclear power plant by HYSPLIT model and amount of rainfall based on AMeDAS.

activities for the dose at the beginning of July 2011 are from ^{134}Cs and ^{137}Cs, the activity ratio (^{134}Cs/^{137}Cs) of which is comparable in the whole investigated area. Based on the relationship between the radiation dose rate of 0.6 μSv/h and ^{137}Cs deposition of 80 kBq/m^2 at 23 km south from the Fukushima No. 1 nuclear power plant, the annual dose can be estimated using the following equation:

$$\text{Annual dose (mSv/y)} = 0.06 \times {}^{137}\text{Cs deposition (kBq/m}^2\text{)}.$$

Conversion factors at 1 m away from the planar source, which estimate external radiation doses from individual radionuclides, are reported to be 5.4×10^{-6} and 2.1×10^{-6} for ^{134}Cs and ^{137}Cs, respectively (http://www-pub.iaea.org/MTCD/Publications/PDF/te_1162_prn.pdf). Using these conversion factors and 0.9 as the ^{134}Cs/^{137}Cs activity ratio, the annual dose also can be estimated with the same equation. This dose rate assumes an outside exposed dose directly from the contaminated surfaces, which is not mixed. An indoor exposed dose would be less than the outside exposed dose. In addition, if sources were mixed in the top 5 cm of soil, the resulting dose rate would be about half of a surface plane source. The outside annual radiation dose due to the radionuclides from the Fukushima accident is estimated to be 10 mSv in Naka-Dori, 40 mSv in Iitate, 0.2 mSv in the region between northern Ibaraki and eastern Saitama, and 2 mSv in southern Ibaraki and northern Chiba prefectures (note that the present estimate does not include the doses from short-lived radionuclides). No internal dose contribution is assumed in these estimations.

Further research on the radionuclides deposited on the land surface will bring us valuable knowledge on the consequences of the radioactive contamination in Japan, which has rather different climate and ecological conditions from the Chernobyl region.

Methods

The surface soil samples were collected in 2011 at the end of March in Ibaraki and Chiba, the middle of April in Fukushima, and the beginning of May in Tochigi, Saitama, and Tokyo. Soil samples of 1-cm thick from surface with 10×10 cm for an area were collected in Ibaraki, 5-cm thick with 5 cmφ for a diameter were collected in Chiba, Fukushima, Tochigi, Saitama, and Tokyo. The distance between adjacent sampling points was 20–30 km. It is thought that more than 90% of the fission products from Fukushima remained in the soil samples, when the soil sampling was performed (10, 11). The soil was transferred in a 100-mL plastic bottle for gamma-ray spectrometry using a high pure germanium detector. Concentrations of $^{129\,m}$Te ($T_{1/2} = 33.6$ d), ^{131}I ($T_{1/2} = 8.02$ d), ^{134}Cs ($T_{1/2} = 2.06$ y), ^{136}Cs ($T_{1/2} = 13.16$ d), and ^{137}Cs ($T_{1/2} = 30.07$ y) were determined by referring to a standard sample that has the same shape as the soil sample. Statistical error of 1σ in an individual radionuclide's activity was less than 5% for ^{131}I, ^{134}Cs, and ^{137}Cs and 10–30% for $^{129\,m}$Te and ^{136}Cs. All activities were decay-corrected as of March 29, 2011, at which time the major discharge was considered to be terminated.

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1. Pranesha T-S, Kamra A-K (1997) Scavenging of aerosol particles by large water drops 3. Washout coefficients, half-lives, and rainfall depths. *J Geophys Res* 102:23947–23954.
2. Maqua M, Bonka H, Horn H-G (1987) Deposition velocity and washout coefficient of radionuclides bound to aerosol particles and elemental radioiodine. *Radiat Prot Dosim* 21:43–49.
3. Muramatsu Y, Sumiya M, Ohmono Y (1987) Iodine-131 and other radionuclides in environmental samples collected from Ibaraki/Japan after the Chernobyl accident. *Sci Total Environ* 67:149–158.
4. Draxler R, Rolph G (2003) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT4) model. (NOAA Air Resources Laboratory, Silver Spring, MD), Available at: http://www.arl.noaa.gov/ready/hysplit4.html.

5. Reineking A, Becker K-H, Porstendörfer J, Wicke A (1987) Air activity concentration and particle size distributions of the Chernobyl aerosol. *Radiat Prot Dosim* 19:159–163.
6. Hou X, et al. (2009) A review on separation of iodine-129 in the environmental and biological samples. *Anal Chim Acta* 632:181–196.
7. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (2000) *Sources and Effects of Ionizing Radiation* (United Nations, New York), pp 195–204.
8. Igarashi Y, Otsuji-Hatori M, Hirose K (1996) Recent deposition of ^{90}Sr and ^{137}Cs observed in Tsukuba. *J Environ Radioact* 31:157–169.
9. Igarashi Y, et al. (2009) Possible change in Asian dust source suggested by atmospheric anthropogenic radionuclides during the 2000s. *Atmos Environ* 43:2971–2980.
10. Schimmack W, Bunzl K, Bachhuber H (1987) Variability of the sorption of Cs, Zn, Sr, Co, Cd, Ce, Ru, Tc, and I at trace concentrations by a forest soil along a transect. *Environ Int* 6:427–436.
11. Gil-Garcia C, Tagami K, Uchida S, Rigol A, Vidal M (2009) New best estimates for radionuclide solid-liquid distribution coefficients in soils. Part 3: Miscellany of radionuclides (Cd, Co, Ni, Zn, I, Se, Sb, Pu, Am, and others). *J Environ Radioact* 100:704–715.