Estimation of Tellurium-132 Distribution in Fukushima Prefecture

Keiko TAGAMI[1)]*, Shigeo UCHIDA[1)], Nobuyoshi ISHII[1)], and Jian ZHENG[1)]

1) Office of Biospheric Assessment for Waste Disposal, National Institute of Radiological Sciences, 4-9-1 Inage-ku, Chiba 2638555, Japan

*k_tagami@nirs.go.jp

The Nuclear and Industrial Safety Agency estimated that about 8.8×10^{16} Bq of tellurium-132 (^{132}Te, half-life: 3.2 d) was released into the air from the Fukushima Daiichi Nuclear Power Plant (FDNPP) which was the third largest discharged amount among the released radionuclides in the nuclear accident. ^{132}Te would be considered a contributor to the radiation dose during the early stages of an accident, that is, in March 2011. However, limited ^{132}Te concentration data were collected because of its relatively short half-life. In this study, the ^{132}Te distribution in Fukushima Prefecture in March was estimated according to the concentration ratio of ^{132}Te to ^{137}Cs by adding ^{132}Te amount estimated from ^{129m}Te. Because the data for ^{137}Cs is already fairly readily available and more data can be collected from the environment around the FDNPP, ^{137}Cs could be used as an indicator of ^{132}Te if the ^{132}Te/^{137}Cs concentration ratio is constant in a certain area. The estimation was carried out using data from the literature. The ^{132}Te/^{137}Cs concentration ratio varied in areas south of the FDNPP. However, it was almost constant near the site (within a radius of ~5 km) and in areas west and north of the plant, with an average value of 20±3. In these areas, ^{132}Te concentration in March 2011 could be roughly estimated from ^{137}Cs.

Keywords: *tellurium-132, cesium-137, concentration ratio, contour map*

1. Introduction

Large amounts of radionuclides were released into the environment after the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in March 2011, and tellurium-132 (^{132}Te, half-life: 3.2 d) was one of the radionuclides. The Nuclear and Industrial Safety Agency[1)] estimated the total amount of radionuclides discharged into the air from the FDNPP and ranked them by amount with the most released radionuclide being non-reactive xenon-133 (^{133}Xe, 11,000 PBq), followed by iodine-131 (^{131}I, 160 PBq) and ^{132}Te (88 PBq). In fact, results of high-volume air sampling carried out in March 2011 by the Center for the Promotion of Disarmament and Non-proliferation in Takasaki[2)] and the High Energy Accelerator Research Organization in Tsukuba (KEK)[3)] showed that the observed ^{132}Te activity at both institutes was as high as that of ^{131}I. Thus, ^{132}Te should be considered as a contributor to the radiation dose from the FDNPP during the early stages of accident, i.e., in March 2011. Unfortunately, however, many instruments for radionuclide determinations were destroyed during the Great East Japan Earthquake that occurred on 11 March 2011, thus limiting the data available for nuclides with short half-lives.

The Ministry of Education, Culture, Sports, Science and Technology (MEXT)[4)] has reported ^{129m}Te (half-life: 33.6 d) distribution in soil samples collected in Fukushima during June–July 2011 from meshes 2 km on each side. The MEXT committee for this project compared the results with cesium-137 (^{137}Cs) and found direction dependence for the ^{129m}Te/^{137}Cs activity ratios, particularly in areas south of the FDNPP. However, the data were collected about 3 months after the large releases in March, and it was not clear whether ^{129m}Te deposited on the ground was retained in the soil at the time of sampling. Moreover, no data were available for ^{132}Te in these samples to show if ^{129m}Te could be used as analogue of ^{132}Te, because the soil sampling was

conducted too late to detect it.

Recently, we determined that ^{132}Te and ^{129m}Te concentrations in soil samples collected in March and April 2011 showed good agreement with a high correlation factor of 0.99 ($p < 0.001$) [5]. However, the data on ^{129m}Te are still limited, and ^{129m}Te may migrate to other places; thus, in this study, we focused on using ^{137}Cs concentrations because Cs is known to be less mobile in soil and it therefore should be retained in the soil. Data for ^{137}Cs is already fairly readily available, and its activity can still be measured in the environment around the FDNPP. The concentration of ^{137}Cs could be used as an indicator of ^{132}Te if the ^{132}Te/^{137}Cs concentration ratio is constant in a certain area.

2. Data sources and data selection criteria

We have summarized data sources elsewhere[5]; briefly, the data were from accessible sources, e.g., from homepages of MEXT, as well as published literature[6,7]. All the data were from soil samples (0–5 cm depth) collected in Fukushima Pref. until the end of May 2011.

Most of these measured values were included in the data sets that we analyzed; however, values were conditionally rejected if the concentration ratio of ^{134}Cs and ^{137}Cs deviated significantly from a 1:1 ratio when the values were decay-corrected to 11 March 2011. If the ratio was far from 1:1, the data were removed from the data sets. Most data were reported on the Bq kg^{-1} basis, although some sources also provided Bq m^{-2}. However, as it was the concentration ratio of ^{132}Te/^{137}Cs that was of interest to us, and since the ratio is dimensionless, the difference in units was not considered. ^{132}Te data were available from 45 sampling sites (n = 1–23), and ^{129m}Te data were available form 139 sampling sites (n = 1–50). ^{137}Cs concentrations were measured in all the samples.

3. Results and Discussion

It was not clear what form of Te was retained in the soil where it was deposited; Te is expected to be in anionic forms ($HTeO_3^-$ or $HTeO_4^-$) in soil under oxic conditions.

We considered the possibility that ^{129m}Te might migrate with water to from its initial deposited sites and/or leach into deeper soil layers. However, in a previous study, when we collected soil samples from 20 April to 5 May 2011 in an area around the FDNPP and determined the vertical distribution of ^{129m}Te, we found

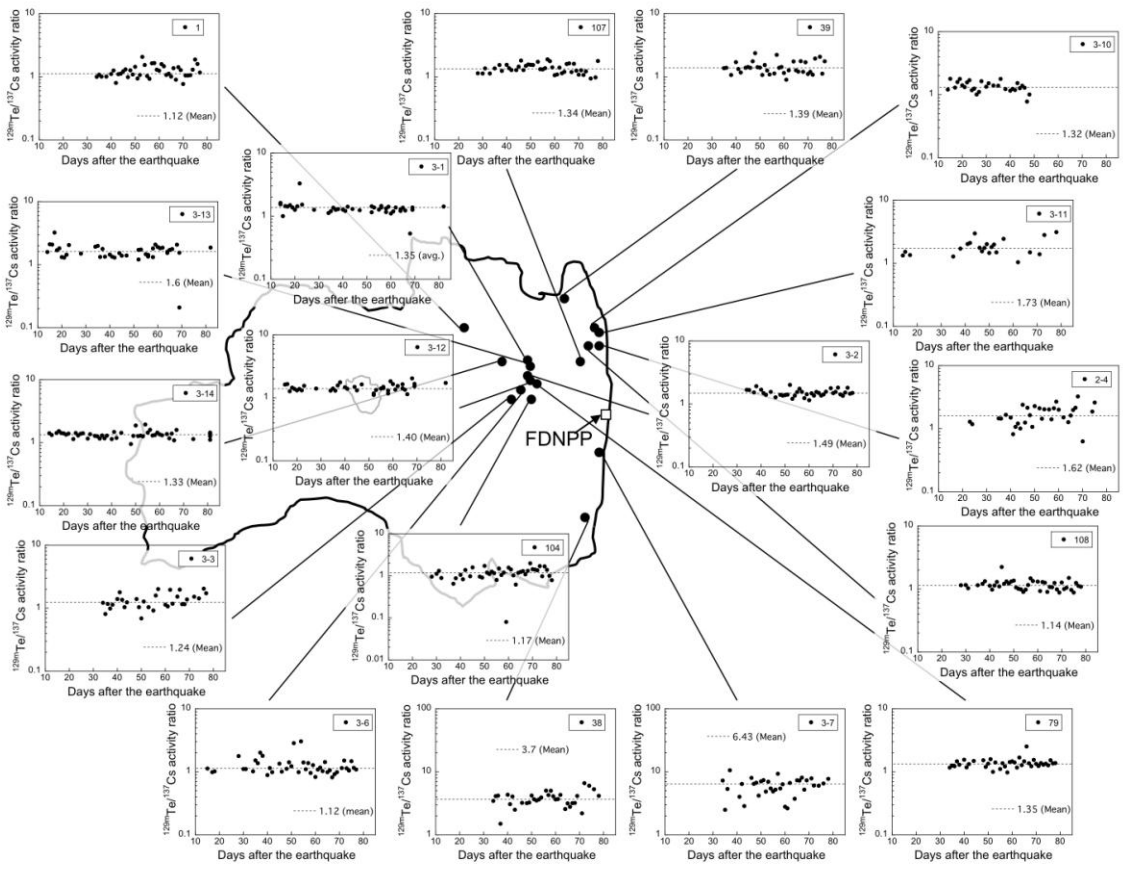

Fig.1. Change with time for the ^{129m}Te/^{137}Cs concentration ratio in soil collected in Fukushima Pref. (decay-corrected to 11 March 2011).

that most of the ^{129m}Te was distributed within 5 cm of the surface of the soil[8], although it might move horizontally with precipitation. Consequently, we analyzed concentration ratios between ^{129m}Te and ^{137}Cs with time. Data from 18 sampling sites are summarized in Fig. 1 for each site. The ratio was almost constant for the 80-d period considered, indicating that ^{129m}Te in the soil during the data collection period was relatively immobile. From ^{129m}Te concentration data, ^{132}Te concentration in the early stage of the accident could be estimated.

The arithmetic mean of the ^{129m}Te/^{137}C concentration ratio at each sampling site is also listed in Fig. 1. For the samples collected in the coastal and southern areas from the FDNPP, the mean ratio range was 3.7–6.4, while other areas showed a narrower range of the mean ratio, 1.12–1.73. Apparently, the deposition patterns of ^{129m}Te and ^{137}Cs to the coastal south was different from that to the north direction.

To show the ^{132}Te/^{137}Cs distribution pattern clearly, we prepared contour maps of the ^{132}Te/^{137}Cs concentration ratio (Fig. 2); logarithm values of ^{132}Te/^{137}Cs are plotted. Using the measured ^{132}Te results, we plotted the ratios in Fig. 2A. To provide more data, we also used ^{132}Te values estimated from ^{129m}Te in Fig. 2B. We applied a factor of 14.6 to convert from ^{129m}Te to ^{132}Te based on our previous observations[5]. Both Figs. 2A and B showed a similar tendency, i.e., a high ratio was observed south of the FDNPP. A similar trend for ^{129m}Te/^{137}Cs had already been reported by Kinoshita et al.[9] and MEXT[4]. It was noteworthy that the ^{132}Te/^{137}Cs concentration ratios near the FDNPP site were almost the same with a range of 12–25 (raw values).

Although the ^{132}Te/^{137}Cs ratio to the south was high (raw value: ~160), the ^{137}Cs amount deposited on the ground was much smaller than in the most highly contaminated area to the northwest of the plant. Accordingly, ^{132}Te amounts deposited on the ground to the south were not as high as those in the most contaminated areas.

One sampling point in Fig. 2B, west-northwest from FDNPP, showed a higher value (raw value: 40) than the other samples collected near this site. This site is in the *Naka-dori* area where higher deposition values were found, which can be attributed to the basin-shaped valley geography there as well as the weather conditions when the deposition occurred. The tendency was similar to that reported by Kinoshita et al.[9]; however, this tendency was not clear from Fig. 2A when fewer ^{132}Te data points were used. By adding estimated data for ^{129m}Te, more information could be provided to estimate ^{132}Te disposition in March 2011.

From these results, we concluded ^{132}Te could be estimated from ^{137}Cs using a conversion factor in the west and north directions from the FDNPP. The data to the

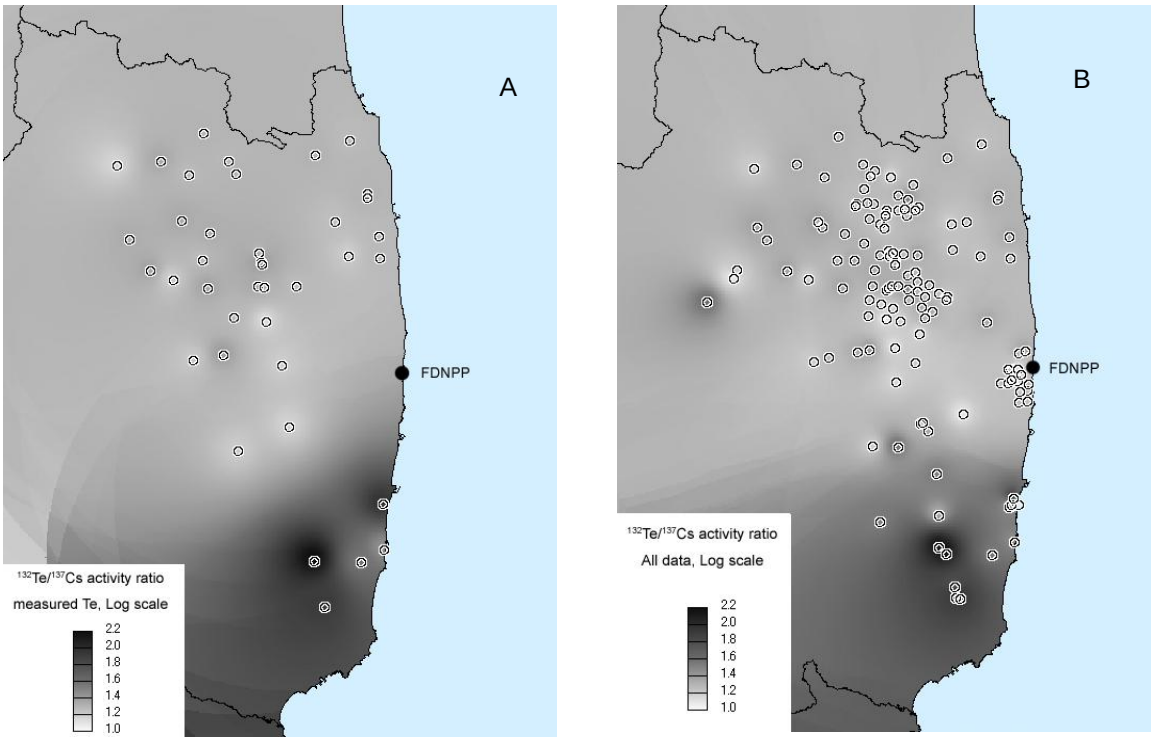

Fig.2. Contour maps of ^{132}Te/^{137}Cs concentration ratios (logarithm values) in soil collected in Fukushima Prefecture (decay-corrected to 11 March 2011).

south and one point in the west–northwest direction were removed from the data sets for the measured and estimated ^{132}Te/^{137}Cs ratios. The histograms are shown in Fig. 3. These values showed normal distributions, and the arithmetic means for measured and estimated ^{132}Te/^{137}Cs concentration ratios were 20.1 and 19.4, respectively, and these values were within the measurement error. Thus, rough estimation of ^{132}Te concentration from ^{137}Cs would be possible using a conversion factor of 20 (11 March 2011). However, collection of more detailed data is necessary to estimate concentrations in the south and *Naka-dori* areas.

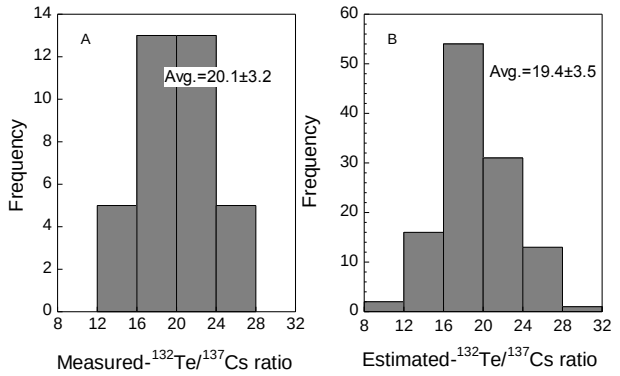

Fig.3. Histograms of ^{132}Te/^{137}Cs concentration ratios in soil samples collected in areas west and north of the FDNPP.

4. Conclusions

In order to provide more data on ^{132}Te for dose estimation in the early stage of the FDNPP accident in March 2011, we carried out a data survey from published reports and open sources. Using these data sets, we confirmed immobility of ^{129m}Te in soil up to 80 d after 11 March 2011. By applying our previously reported conversion factor[5], we estimated the ^{132}Te concentration from ^{129m}Te data and compared the value with that of ^{137}Cs. It was clear that the distribution of the ^{132}Te/^{137}Cs concentration ratio varied with the direction from the FDNPP. However, areas to the west and north of the FDNPP had almost the same ^{132}Te/^{137}Cs concentration ratio with an arithmetic mean value of 20 (11 March 2011); ^{132}Te deposition in areas to the west and north of the plant could be estimated using that mean value and ^{137}Cs concentrations.

Acknowledgements

We thank Dr. E. R. Landa (U.S. Geological Survey) for his valuable comments. This work was partially supported by the Agency for Natural Resources and Energy, the Ministry of Economy, Trade and Industry (METI), Japan.

References

1) Nuclear and Industrial Safety Agency: Data on the amount of released radioactive materials (revised 20 October 2011), 2011. http://www.meti.go.jp/press/2011/ 10/20111020001/20111020001.html (accessed 8 November 2012)
2) Center for the Promotion of Disarmament and Non-proliferation: Radionuclide monitoring data in the air at Takasaki, 2012. http://www.cpdnp.jp/013.html (accessed 12 Sept 2012)
3) KEK: Radionuclide monitoring data in the air at Tsukuba, 2012. http://www.kek.jp/ja/Research/ARL/RSC/Radmonitor/ (accessed 12 Sept 2012)
4) MEXT: Contour maps of ^{129m}Te and ^{110m}Ag in soil. 2011. http://radioactivity.mext.go.jp/ja/contents/6000/5050/view.html (accessed 23 July 2012). Data available: 5600_103120.pdf (in Japanese)
5) Tagami, K., Uchida, S., Ishii, N., Zheng, J.: Estimation of Te-132 distribution in the eastern part of Fukushima prefecture at the early stage of Fukushima Daiichi nuclear power plant accident. (Manuscript in preparation)
6) Fujiwara, T., Saito, T., et al.: Isotopic ratio and vertical distribution of radionuclides in soil affected by the accident of Fukushima Dai-ichi nuclear power plants. J. Environ. Radioactiv. 113, 37-44, 2012.
7) Yamamoto, M., Takada, T., Nagao, S., Koike, T., Shimada, K., Hoshi, M., Zhumadilov, K., Shima, T., Fukuoka, M., Imanaka, T., Endo, S., Sakaguchi, A., Kimura, S.: An early survey of the radioactive contamination of soil due to the Fukushima Dai-ichi Nuclear Power Plant accident, with emphasis on plutonium analysis. Geochem. J. 46, 341-353, 2012.
8) Tagami, K., Uchida, S., Uchihori, Y., Ishii, N., Kitamura, H., Shirakawa, Y.: Specific activity and activity ratios of radionuclides in soil collected about 20 km from the Fukushima Daiichi Nuclear Power Plant: Radionuclide release to the south and southwest. Sci. Total Environ. 409, 4885-4888, 2011.
9) Kinoshita, N., Sueki, K., Sasa, K., Kitagawa, J., Ikarashi, S., Nishimura, T., Wong, Y.S., Satou, Y., Handa, K., Takahashi, T., Sato, M., Yamagata, T.: Assessment of individual radionuclide distributions form the Fukushima nuclear accident covering central-east Japan. P. Natl. Acad. Sci. USA 108, 19526-19529, 2011.