

ESTIMATION OF THE AMOUNT OF I-129 IN THE ENVIRONMENT GENERATED DUE TO THE DECAY OF TE-129M DISCHARGED BY THE FUKUSHIMA NPS ACCIDENT

Haruo Sato¹

¹Okayam University, Okayama City, Okayama 700-8530, Japan

ABSTRACT

The accident at the Fukushima Daiichi Nuclear Power Station (1F-NPS) occurred following the Great East Japan Earthquake in March 2011, and led to the release of volatile radionuclides, which were deposited on the environment such as soils, forests, residential land, etc. in the Fukushima prefecture and the neighbouring areas. From immediately after the accident, air dose rate monitoring and analyses of radionuclides in soil, etc. were conducted in a wide range of area. Consequently, volatile radionuclides such as I-131, Cs-134, Cs-137, Te-129m, and Ag-110m were detected in a wide range of area, where was distant over 60km from 1F-NPS. Since I-131 is short-lived, it approximately disappeared after a few months. After the decay of short-lived I-131, Radiocaesium (Cs-134 and Cs-137) are the main contributors to radiation dose rate.

On the other hand, Te-129m, of which amount was approximately the same level as both Radiocaesium, was also detected although its half-life is short about 1 month. Since very long-lived I-129 is generated due to the decay of Te-129m, the accumulation of I-129 in the environment by accident is worried. Te-129m decays to I-129 by the following 2 decay series; Te-129m \rightarrow Te-129 (half-life: 69.6min) \rightarrow I-129 (64%), Te-129m \rightarrow I-129 (36%). Since the half-lives of Te-129m and Te-129 are 33.6d and 69.6min, respectively, radioactive equilibrium is established between both nuclides, but it is not established between Te-129 and I-129 and between Te-129m and I-129. In this case, the amount of I-129 increases due to the decay of Te-129m, but also the maximum value of the amount theoretically exists.

In this study, the amount of I-129 in the environment generated due to the decay of Te-129m discharged by the 1F-NPS accident was estimated based on the analysed data of Te-129m in soil. The analysis was conducted for 3 investigation locations (Okuma Town, ca. 2.7km from 1F-NPS, Naraha Town, ca. 19km from 1F-NPS, and Fukushima City, ca. 60km from 1F-NPS) among the analysed data (as of 14 June 2011) of soil samples obtained at about 2,200 investigation locations in the Fukushima prefecture and the neighbouring prefectures. The

maximum amount of I-129 was estimated 1.03×10^{-1} (Bq/m²) after about 1,000d even for the maximum deposition amount of Te-129m (2.7 (MBq/m²) as of 14 June 2011). This is approximately the same level as radioactivity concentration in the environment ($5.75 \times 10^{-3} \sim 2.96 \times 10^{-1}$ (Bq/m²)). Additionally, the results of field investigation around 1F-NPS also support this conclusion.

1. INTRODUCTION

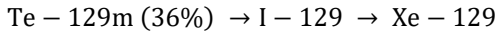
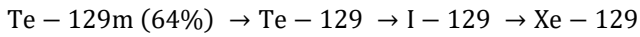
The accident at the Tokyo Electric Power Company (TEPCO) Fukushima Daiichi Nuclear Power Station (1F-NPS) occurred following the Great East Japan Earthquake on the date of 11 March 2011, and led to the release of volatile radionuclides, which were deposited on the environment such as soils, forests, residential land, etc. in the Fukushima prefecture and the neighbouring prefectures. The amounts of radionuclides discharged into the atmosphere were reported 160PBq (where, 1PBq= 1×10^{15} Bq) for I-131, 18PBq for Cs-134, 15PBq for Cs-137 and 0.14PBq for Sr-90 by the Ministry of the Environment (MOE) [1]. Chino *et al.* [2] estimated the total amounts of I-131 and Cs-137 discharged into the atmosphere by April 5, 2011 to be approximately 150PBq and 13PBq, respectively. The Nuclear and Industrial Safety Agency (NISA) within the Ministry of Economy, Trade and Industry (METI) estimated the total amounts of I-131 and Cs-137 discharged into the atmosphere by the end of April 2011 to be approximately 160PBq and 15PBq, respectively [3]. The amounts of I-131 and Cs-137 discharged into the atmosphere are the same as those that MOE reported [1]. Furthermore, the National Diet of Japan Fukushima Nuclear Accident Independent Investigation Commission (NAIIC) Reports reported that the release of amounts of I-131 and Cs-137 by May 2011 were estimated approximately 500PBq and 10PBq, respectively [4].

On the other hand, Stohl *et al.* [5] estimated the total amount of Cs-137 released to the atmosphere to be 36.6PBq (20.1-53.1PBq) on average based on the results of an atmosphere transport model (FLEXPART) and measurement data from several dozen stations in Japan, North America and other

regions. Thus, although some variations are found in the amounts of radionuclides discharged to the atmosphere, the release amounts of I-131 and Cs-137 are considered to be roughly of orders 100PBq and 10PBq, respectively. As aforementioned, since the release amounts of I-131 and Cs-137 are estimated approximately 150-500PBq and 400-1,500PBq with value converted to I-131, respectively (where, conversion factor of Cs-137 to I-131 is 40), the total sum of both nuclides is in the range of 550-2,000PBq. This amount is equivalent to approximately 1/9 to 1/3 of the 5,200PBq discharged by Chernobyl Nuclear Power Station explosion accident occurred in April 1986. The 1F-NPS accident was evaluated level 7 on the International Nuclear and Radiological Event Scale (INES), which is the worst level.

From immediately after the accident, air dose rate monitoring and analyses of radionuclides in soils and plants, etc. were conducted in a wide range of area. Consequently, volatile radionuclides such as I-131, Cs-134, Cs-137, Te-129m, and Ag-110m were detected in a wide range of area, where was distant over 60km from 1F-NPS, although their concentrations and distributions are different depending on radionuclide [eg., 6]. Since I-131 is short-lived (half-life: 8.021d) [7], it approximately disappeared after a few months. After the decay of short-lived I-131, radiocaesium (Cs-134 and Cs-137) are the main contributors to radiation dose rate.

On the other hand, Te-129m, of which amount in soil was approximately the same level as both radiocaesium, was also detected although its half-life is short about 1 month (half-life: 33.6d) [7]. Since very long-lived I-129 (half-life: 1.57×10^7 yr) [7] is generated due to the decay of Te-129m, the accumulation of I-129 in the environment by the accident is worried. Te-129m decays to I-129 by the following 2 decay series.



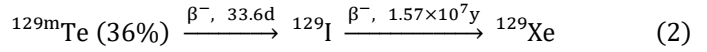
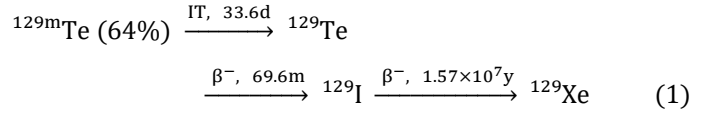
Since the half-lives of Te-129m and Te-129 are 33.6d and 69.6min, respectively, radioactive equilibrium is established between Te-129m and Te-129, but it is not established between Te-129 and I-129 and between Te-129m and I-129. The amount of I-129 increases due to the decay of Te-129m, but the maximum value of the amount also theoretically exists.

In this study, we estimated the amount of I-129 in the environment generated due to the decay of Te-129m discharged by the 1F-NPS accident based on the analysed data (deposition amount) of Te-129m in soil and compared to the radioactivity concentrations of I-129 in the environment.

2. ANALYSIS

2.1 Calculation of the Amount of I-129 Generated due to the Decay of Te-129m

As described earlier, Te-129m decays to I-129 by the following 2 decay series.



For decay series (1) and (2), we consider the following relations, respectively.

$$X_1 \xrightarrow{\lambda_1} X_2 \xrightarrow{\lambda_2} X_3 \xrightarrow{\lambda_3} X_4 \quad (3)$$

$$X_1 \xrightarrow{\lambda_1} X'_3 \xrightarrow{\lambda_3} X'_4 \quad (4)$$

Where, X_1 , X_2 , X_3 and X_4 in Eq. (3) are the numbers of nuclei of Te-129m, Te-129, I-129 and Xe-129 in decay series (1), respectively, X'_3 and X'_4 in Eq. (4) are the numbers of nuclei of I-129 and Xe-129 in decay series (2), respectively, and λ_1 , λ_2 and λ_3 are the decay constants ($= \ln 2/T$, T : half-life) of Te-129m, Te-129 and I-129, respectively.

For Eq. (3), the following relations are established.

$$\frac{dX_1}{dt} = -\lambda_1 X_1 \quad (5)$$

$$\frac{dX_2}{dt} = -\lambda_2 X_2 + \lambda_1 X_1 \quad (6)$$

$$\frac{dX_3}{dt} = -\lambda_3 X_3 + \lambda_2 X_2 \quad (7)$$

For Eq. (4), the following relations are established.

$$\frac{dX'_3}{dt} = -\lambda_3 X'_3 + \lambda_1 X_1 \quad (8)$$

Where, t is the elapsed time after the 1F-NPS accident.

When $X_1 = X_0$ and $X_2 = X_3 = X'_3 = 0$ at $t = 0$, we can respectively obtain the following analytical solutions by solving differential equations given by Eqs. (5) to (8).

$$X_1 = X_0 e^{-\lambda_1 t} \quad (9)$$

$$\begin{aligned} X_2 &= X_0 \left(\frac{\lambda_1}{\lambda_2 - \lambda_1} e^{-\lambda_1 t} + \frac{\lambda_1}{\lambda_1 - \lambda_2} e^{-\lambda_2 t} \right) \\ &= \frac{aX_0\lambda_1}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \end{aligned} \quad (10)$$

$$\begin{aligned}
X_3 &= aX_0 \left\{ \frac{\lambda_1 \lambda_2}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} e^{-\lambda_1 t} \right. \\
&\quad + \frac{\lambda_1 \lambda_2}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} e^{-\lambda_2 t} \\
&\quad \left. + \frac{\lambda_1 \lambda_2}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} e^{-\lambda_3 t} \right\} \\
&= aX_0 \lambda_1 \lambda_2 \left\{ \frac{e^{-\lambda_1 t}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} \right. \\
&\quad \left. + \frac{e^{-\lambda_3 t}}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} \right\} \quad (11)
\end{aligned}$$

$$\begin{aligned}
X'_3 &= bX_0 \left(\frac{\lambda_1}{\lambda_3 - \lambda_1} e^{-\lambda_1 t} + \frac{\lambda_1}{\lambda_1 - \lambda_3} e^{-\lambda_3 t} \right) \\
&= \frac{bX_0 \lambda_1}{\lambda_3 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_3 t}) \quad (12)
\end{aligned}$$

Where, X_0 is the initial number of nuclei of T-129m, a and b are the proportion of decay to decay series (1) and (2), respectively ($a = 64\%$ and $b = 36\%$ in this case).

Therefore, by giving initial number of nuclei of Te-129m (X_0) and the proportion of decay to Eqs. (3) and (4) for Eqs. (11) and (12), respectively, the numbers of nuclei of I-129 to respective decay series can be calculated.

The radioactivity (concentration) of each radionuclide can be calculated from the following relations based on Eqs. (9) to (12).

$$A_1 = \lambda_1 X_0 e^{-\lambda_1 t} \quad (13)$$

$$A_2 = \lambda_2 X_2 = \frac{aX_0 \lambda_1 \lambda_2}{\lambda_2 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_2 t}) \quad (14)$$

$$\begin{aligned}
A_3 = \lambda_3 X_3 &= aX_0 \lambda_1 \lambda_2 \lambda_3 \left\{ \frac{e^{-\lambda_1 t}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} \right. \\
&\quad + \frac{e^{-\lambda_2 t}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} \\
&\quad \left. + \frac{e^{-\lambda_3 t}}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} \right\} \quad (15)
\end{aligned}$$

$$A'_3 = \lambda_3 X'_3 = \frac{bX_0 \lambda_1 \lambda_3}{\lambda_3 - \lambda_1} (e^{-\lambda_1 t} - e^{-\lambda_3 t}) \quad (16)$$

Where, A_1 is the total radioactivity (concentration) of Te-129m, A_2 and A_3 are the radioactivities (concentrations) of Te-129 and I-129 in decay series (1), respectively, and A'_3 is the radioactivity of I-129 in decay series (2).

Therefore, the total sum of radioactivity of I-129 can be calculated by the following equation.

$$A = A_3 + A'_3 \quad (17)$$

Where, A is the total sum of radioactivity of I-129.

2.2 Analytical Condition

The analysis was conducted based on the data of Te-129m at 3 investigation locations (area contaminated with a high concentration (area with high air dose rate), area contaminated with a moderate concentration (area with moderate air dose rate), and area contaminated with a low concentration (area with low air dose rate)) among the analysed data (converted to as of 14 June 2011) of soil samples obtained at about 2,200 locations in the Fukushima prefecture and the neighbouring prefectures [6].

Since all data of Te-129m obtained in the field investigation were converted to as of 14 June 2011, those data were converted to as of 15 March 2011 ($t = 0$) based on its half-life in this analysis. The date of 15 March 2011 is the date that much amount of radionuclides were discharged from 1F-NPS and maximum value of the air dose rate was monitored in the

TABLE 1: INPUT DATA OF Te-129m AT 3 INVESTIGATION LOCATIONS SELECTED FOR THE ANALYSIS

Investigation location	WGS84 coordinate system	Deposition amount (as of 14 June 2011) (Bq/m ²)	Deposition amount (converted to as of 15 March 2011) (Bq/m ²)	Remarks
Okuma Town	N37, 25, 30.9 E140, 00, 18.7	2.70×10^6	1.76×10^7	Area contaminated with a high concentration (ca. 2.7km from 1F-NPS)
Naraha Town	N37, 15, 29.4 E140, 58, 09.0	4.70×10^4	3.07×10^5	Area contaminated with a moderate concentration (ca. 19km from 1F-NPS)
Fukushima City	N37, 46, 21.0 E140, 30, 54.0	6.40×10^4	4.18×10^5	Area contaminated with a low cocentration (ca. 60km from 1F-NPS)

Fukushima city. Table 1 shows the input data of Te-129m at 3 investigation locations selected for the analysis.

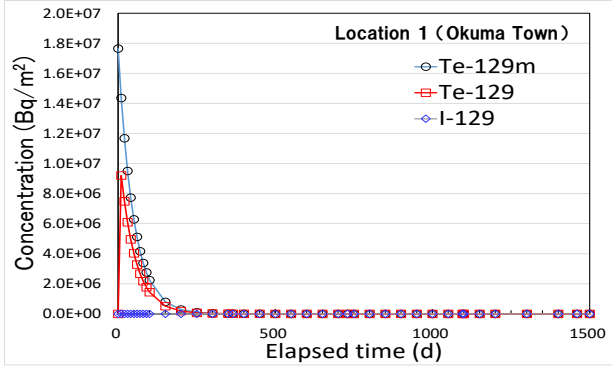


FIGURE 1: THE CALCULATED RESULTS OF TIME VARIATIONS OF THE AMOUNTS OF Te-129m, Te-129 AND I-129 AT INVESTIGATION LOCATION 1 (OKUMA TOWN, CA. 2.7km FROM 1F-NPS)

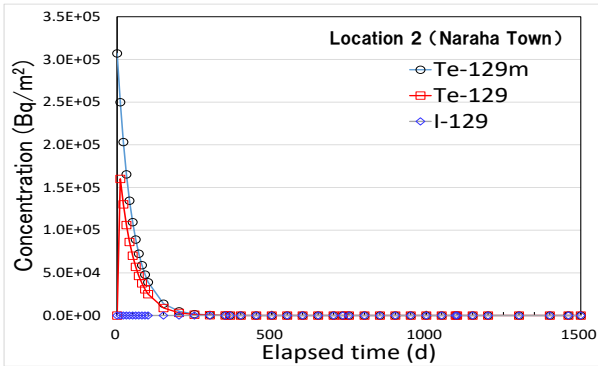


FIGURE 2: THE CALCULATED RESULTS OF TIME VARIATIONS OF THE AMOUNTS OF Te-129m, Te-129 AND I-129 AT INVESTIGATION LOCATION 2 (NARAH TOWN, CA. 19km FROM 1F-NPS)

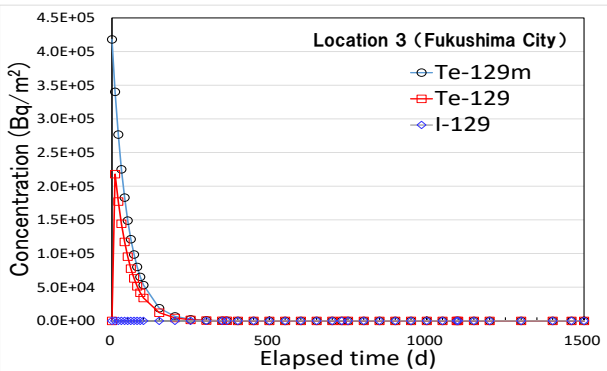


FIGURE 3: THE CALCULATED RESULTS OF TIME VARIATIONS OF THE AMOUNTS OF Te-129m, Te-129 AND I-129 AT INVESTIGATION LOCATION 3 (FUKUSHIMA CITY, CA. 60km FROM 1F-NPS)

3. RESULTS AND DISCUSSION

3.1 Time Variation of the Amount of Each Radionuclide

Figures 1-3 show the calculated results of time variations of the amounts (Bq/m^2) of Te-129m, Te-129 and I-129 at

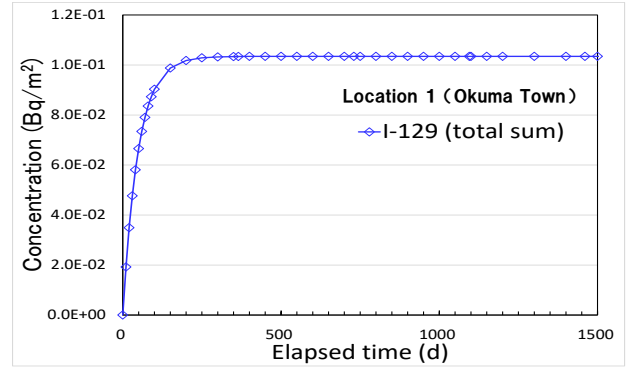


FIGURE 4: THE GRAPH ENLARGED ONLY TIME VARIATION OF THE AMOUNT OF I-129 AT INVESTIGATION LOCATION 1 (OKUMA TOWN, CA. 2.7km FROM 1F-NPS)

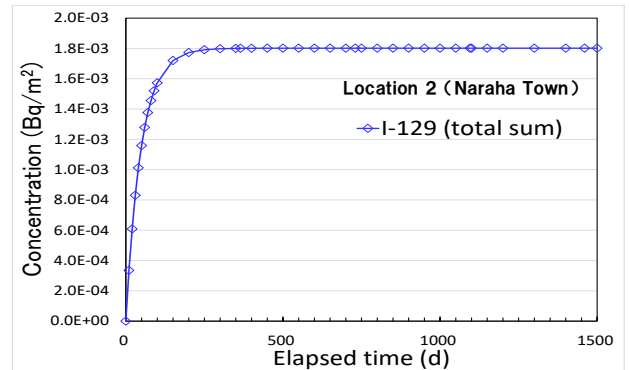


FIGURE 5: THE GRAPH ENLARGED ONLY TIME VARIATION OF THE AMOUNT OF I-129 AT INVESTIGATION LOCATION 2 (NARAH TOWN, CA. 19km FROM 1F-NPS)

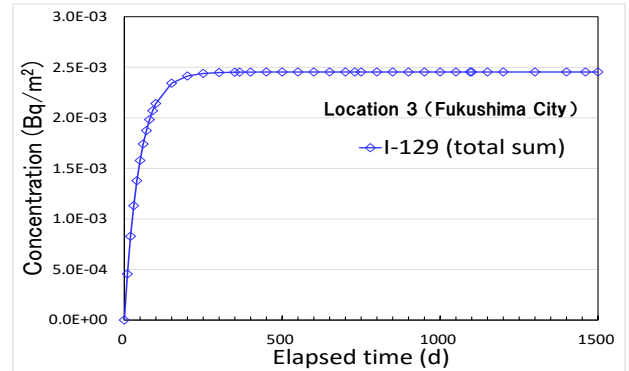


FIGURE 6: THE GRAPH ENLARGED ONLY TIME VARIATION OF THE AMOUNT OF I-129 AT INVESTIGATION LOCATION 3 (FUKUSHIMA CITY, CA. 60km FROM 1F-NPS)

investigation location 1 (Okuma Town, about 2.7km from 1F-NPS), investigation location 2 (Naraha Town, about 19km from 1F-NPS) and investigation location 3 (Fukushima City, about 60km from 1F-NPS), respectively. Additionally, Figures 4-6 show the graphs enlarged only time variations of the amounts (Bq/m²) of I-129 at investigation locations 1-3, respectively.

As shown in Figs. 4-6, the total amounts of I-129 increased with an increase of time, and asymptotically approached to a constant value. Although radioactive equilibrium is not established between Te-129 and I-129 and between Te-129m and I-129 as explained before, the total amount of I-129 theoretically has a maximum value. As a result of the analysis, the total amount of I-129 increased up to after about 1,000d from 15 March 2011 and gradually began to decrease after that.

The maximum value of the total amount of I-129 was estimated 1.03×10^{-1} (Bq/m²) after 1,000d even for the maximum deposition amount of Te-129m (2.7 (MBq/m²) as of 14 June 2011, investigation location 1 (Okuma Town, about 2.7km from 1F-NPS)). Table 2 shows the calculated results of the maximum amounts of I-129 for 3 investigation locations.

Comparing these maximum amounts of I-129 with these initial deposition amounts of Te-129m, the maximum amount of I-129 is approximately one of the 2×10^8 of the initial deposition amount of Te-129m. Thus, we can estimate the maximum amount of I-129 by knowing initial deposition amount of Te-129m in soil.

3.2 Comparison with the Amount of I-129 in the Environment

We compared the amounts of I-129 in the environment generated due to the decay of Te-129m with those (background) originally existing in the environment. Table 3 shows the amounts of I-129 (background) in the environment (soil) all over Japan [8]. As shown in Table 3, the background of I-129 in the environment is ranging $5.75 \times 10^{-3} \sim 2.96 \times 10^{-1}$ (Bq/m²), which is approximately the same level as the maximum amount of I-129 ($=1.03 \times 10^{-1}$ (Bq/m²)) in the environment generated due to the decay of Te-129m.

Honda reported the amount of fallout of I-129 in soil by the 1F-NPS accident to be $1.02 \times 10^{-2} \sim 7.20 \times 10^{-1}$ (Bq/m²) based on

TABLE 2: THE CALCULATED RESULTS OF THE MAXIMUM AMOUNTS OF I-129 FOR 3 INVESTIGATION LOCATIONS

Location	WGS coordinate system	Maximum amount of I-129 (Bq/m ²)
Okuma Town	N37, 25, 30.9 E140, 00, 18.7	1.03×10^{-1}
Naraha Town	N37, 15, 29.4 E140, 58, 09.0	1.80×10^{-3}
Fukushima City	N37, 46, 21.0 E140, 30, 54.0	2.45×10^{-3}

the results of depth distribution investigation at 11 locations [9]. Although these amounts are rather higher than background, it is approximately the same level as background. Miyake *et al.* [10] investigated I-129 concentrations in soil samples (forest, urban areas) around 1F-NPS and reported the surface deposition amount of I-129 in soil to be ranging $1.56 \times 10^{-2} \sim 7.04 \times 10^{-1}$ (Bq/m²). This range of deposition amount is also approximately the same range as the range that Honda reported [9], which is nearly background level.

Furthermore, Muramatsu *et al.* [11] also investigated the deposition density of I-129 in soil collected around 1F-NPS and reported those to be $5.5 \times 10^{-2} \sim 2.391$ (Bq/m²). In this case, some deposition densities in the town of Namie and the city of Fukushima were clearly higher than background in the environment all over Japan (≥ 1 Bq/m²). This is considered to be due to that volatile I-129 discharged from 1F-NPS directly deposited on the surface layer of soil. Also in this report, it is reported that the contribution of I-129 generated from Te-129m (by simple calculation) is approximately 0.3% of the fallout of I-129 directly deposited on the surface layer of soil. From the viewpoint of this, part of the investigation data of I-129 which Honda [9] and Miyake *et al.* [10] reported are considered to have included the fallout of I-129 directly discharged from 1F-NPS, because part of the investigation data were collected around 1F-NPS such as Namie Town.

If this contribution of the amount of I-129 generated by decay from Te-129m to the fallout of I-129 which Muramatsu *et al.* [11] estimated is valid (0.3%), the deposition density of I-129 in soil including the fallout of I-129 directly discharged from 1F-NPS is estimated 5.28×10^{-4} (Bq/m²) in the town of Okuma

TABLE 3: THE BACKGROUND OF I-129 IN THE ENVIRONMENT (ALL OVER JAPAN)

Location	I-129 (Bq/m ²)
Shintoku Hokkaido	1.46×10^{-1}
Nishiki Akita	4.81×10^{-2}
Mito Ibaraki	1.24×10^{-1}
Iwama Ibaraki	2.96×10^{-1}
Isesaki Gunma	1.78×10^{-2}
Kanazawa Ishikawa	5.90×10^{-2}
Kanmuriyama Fukui	1.48×10^{-1}
Okuekukogen Fukui	8.50×10^{-2}
Higashi-Osaka Osaka	7.05×10^{-3}
Ningyotoge Okayama	1.56×10^{-2}
Uwajima Ehime	5.75×10^{-3}
Nishiyama Nagasaki	1.61×10^{-2}
Jonan Kumamoto	1.85×10^{-2}

(investigation location 1: about 2.7km from 1F-NPS), 9.21×10^2 (Bq/m²) in the town of Naraha (investigation location 2: about 19km from 1F-NPS) and 1.25×10^2 (Bq/m²) in the city of Fukushima (investigation location 3: about 60km from 1F-NPS), for 3 investigation locations in this study. However, such high radioactivity concentrations of I-129 in soil have not been reported. The fallout of I-129 discharged from 1F-NPS is one of the future issues. At present, the effect of I-129 generated due to the decay of Te-129m discharged by the accident on the environment is considered to be negligible.

4. CONCLUSION

In this study, the amount of I-129 in the environment generated due to the decay of Te-129m discharged by the 1F-NPS accident was estimated based on the analysed data (deposition data) of Te-129m in soil obtained at about 2,200 locations in the Fukushima prefecture and the neighbouring prefectures, and compared to the radioactivity concentrations (background) of I-129 in the environment all over Japan and some field investigation data of I-129 in soil around 1F-NPS.

The maximum amount of I-129 was estimated 1.03×10^{-1} (Bq/m²) after about 1,000d even for the maximum deposition amount of Te-129m (2.7 (MBq/m²) as of 14 June 2011). The maximum amount of I-129 is approximately one of the 2×10^8 of the initial deposition amount of Te-129m. This is approximately the same level as radioactivity concentrations in the environment all over Japan ($5.75 \times 10^{-3} \sim 2.96 \times 10^{-1}$ (Bq/m²)). The results of some field investigations of I-129 in soil around 1F-NPS range $1.02 \times 10^{-2} \sim 2.391$ (Bq/m²) and some data are clearly higher than background in the environment. This is considered to include also the fallout of I-129 discharged from 1F-NPS. Therefore, the effect of I-129 generated due to the decay of Te-129m discharged by the 1F-NPS accident on the environment is considered to be negligible.

REFERENCES

- [1] Ministry of the Environment (MOE), <https://www.env.go.jp/chemi/rhm/h29kisoshiryo/h29kiso-02-02-05.html>, accessed on Nov. 1 (2018) [in Japanese].
- [2] Chino, M., Nakayama, H., Nagai, H., Terada, H., Katata, G., and Yamazawa, H., Preliminary estimation of release amounts of ¹³¹I and ¹³⁷Cs accidentally discharged from the Fukushima Daiichi Nuclear Power Plant into the atmosphere, *J. Nucl. Sci. Technol.*, Vol. 48, No. 7, pp. 1129-1134 (2011).
- [3] Nuclear Emergency Response Headquarters, Government of Japan, Report of the Japanese Government to the IAEA ministerial conference on nuclear safety –The accident at TEPCO's Fukushima Nuclear Power Stations– (2011).
- [4] National Diet of Japan Fukushima Nuclear Accident Independent Investigation Commission (NAIIC) Reports (5 July 2012), <http://warp.da.ndl.go.jp/info:ndljp/pid/385637/naiic.go.jp/en/report/>, accessed on July 21 (2014).
- [5] Stohl, A., Seibert, P., Wotawa, G., Arnold, G., Burkhardt, J. F., Eckhardt, S., Tapia, C., Vargas, A., and Yasunari, T. J., Xenon-133 and caesium-137 release into the atmosphere from the Fukushima Dai-ichi nuclear power plant: determination of the source term, atmospheric dispersion, and deposition, *Atmospheric Chemistry and Physics*, Vol. 12, pp.2313-2343 (2012).
- [6] Ministry of Education, Culture, Sports, Science and Technology (MEXT), Results of the Nuclide Analysis of Soil Sampling at 2,200 Locations in Fukushima Prefecture and Neighboring Prefectures, <https://emdb.jaea.go.jp/emdb/en/portals/1020101001/>, accessed on Dec. 7 (2018).
- [7] Japan Radioisotope Association, Radioisotope Pocket Data Book, 9th ed., Maruzen, Tokyo (2000) [in Japanese].
- [8] Takeishi, M., Namiki, A., Katagiri, H., Ishida, J., and Nomura, Y., Neutron activation analysis of ¹²⁷I and ¹²⁹I in environmental samples, *PNC ZN843 85-39* (1985) [in Japanese].
- [9] Honda, M., The Mobility of Anthropogenic Iodine-129 in the Terrestrial Environment of Fukushima, Doctoral Thesis, University of Tsukuba (2018).
- [10] Miyake, Y., Matsuzaki, H., Fujiwara, T., Saito, T., Yamagata, T., Honda, M., and Muramatsu, Y., Isotopic Ratio of Radioactive Iodine (¹²⁹I/¹³¹I) Released from Fukushima Daiichi NPP Accident, *Geochemica Journal*, Vol.46, pp.327-333 (2012).
- [11] Japan Atomic Energy Agency (JAEA), 2nd Investigation and Research on Distribution Status, etc. of Radioactive Substances by Fukushima Daiichi Nuclear Power Station Accident (Part 1): Making of Distribution Map of Radiation Dose, etc., Appendix 4.1, pp.1-149 -1-60 (2013) [in Japanese].