Natural Variation of Ambient Dose Rate in Air of Izu-Oshima Island After the Fukushima Daiichi Nuclear Power Plant Accident

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ABSTRACT

The ambient dose rate in air and radioactivity concentration in soil samples collected on Izu-Oshima Island were observed in 2012, 2013 and 2014, i.e., one, two and three years after the severe accident at the Fukushima Daiichi Nuclear Power Plant. A car-borne survey for the ambient dose rate in air was carried out for the entire island. Soil samples were collected for the radioactivity concentration measurements from 22 points. The ambient dose rates in air were 36 nGy h^{-1} in 2012, 34 nGy h^{-1} in 2013 and 29 nGy h^{-1} in 2014, respectively. The corresponding radioactivity concentrations in those years for ^{134}Cs were 53 Bq kg^{-1}, 39 Bq kg^{-1} and 29 Bq kg^{-1}, and for ^{137}Cs, 87 Bq kg^{-1}, 73 Bq kg^{-1} and 75 Bq kg^{-1}. All the values have decreased every year.

INTRODUCTION

Facilities and infrastructures in wide areas of northeastern Japan were damaged in the Great East Japan Earthquake and Tsunami that occurred on March 11, 2011. Damage was caused at the Fukushima Daiichi Nuclear Power Plant (F1-NPP), leading to the severe accident there and large amounts of artificial radionuclides such as ^{131}I, ^{134}Cs and ^{137}Cs were released into the environment. Most of the artificial radionuclides released were wet-deposited in a district located in the northwest direction from the F1-NPP, such as Iidate Village and Namie Town [(1)]. However, high-dose rate areas formed by rainfall and wind were widespread in eastern Japan [(2)].

On Izu-Oshima Island, located approximately 350 km southwest from F1-NPP, a survey was carried out in August 2011 showing that the ambient dose rates in air were 2.9 times higher than the dose rates in air before the accident [(3)]. Additionally, while ^{134}Cs had not been detected before the accident, 6.4 - 394.6 Bq kg^{-1} (n = 21) of ^{134}Cs and 7.6 - 499.3 Bq kg^{-1} (n = 21) of ^{137}Cs were detected in the island soil after the accident. To make a dynamic analysis of ^{134}Cs and ^{137}Cs in soil and examine the change in the annual effective dose calculated from the ambient dose rate in air for the island's residents, it is necessary to continue investigations, even years after the accident.

In this study, the results of sampling on Izu-Oshima in 2012, 2013 and 2014 were obtained to get a better picture of the changes in radioactivity concentration in soil and the effective dose in the three years since the F1-NPP accident.

MATERIALS AND METHODS

Ambient dose rate in air

The measurements of the ambient dose rates in air (nGy h^{-1}) and the concentrations (Bq kg^{-1}) of radiocesium (^{134}Cs and ^{137}Cs) were made in August 2012, August 2013 and August 2014 on Izu-Oshima Island (Fig. 1). A car-borne survey was carried out using a 2-in × 2-in NaI(Tl) scintillation spectrometer (Osprey, CANBERRA, Inc., Meriden, CT) in 2012 and 2013, while a 3-in × 3-in NaI(Tl) scintillation spectrometer (EMF211, EMF Japan Co., Osaka, Japan) was used in 2014. Latitude and longitude at each measurement point were measured with a global positioning system at the same time as the count rates were recorded. Measurements were made every minute in the 2012 and 2013 surveys and every 30 seconds in the 2014 survey and main roads were used wherever possible for all surveys (Fig. 2). Shielding by the car body was estimated by making measurements inside and outside of the car at six locations. Counting time was set to 2 min. For outside measurements, the scintillation spectrometer was positioned 1 m above the ground surface. A preliminary experiment was performed each year and the shielding factors were 1.60 ± 0.10 in 2012, 1.41 ± 0.09 in 2013 and 1.41 ± 0.20 in 2014. These factors were used for correction of the measured value, so as to represent the outside count rate. The pulse height distributions were also obtained outside the car for 20 min at seven locations (#1, #3, #4, #5, #9, #13, #16 in Fig. 2) in 2012 and 2013, and for 10 min at twenty-two locations (#1 - #22 in Fig. 2) in 2014. The obtained pulse height distributions were unfolded using a 22 × 22 response matrix, and the ambient dose rates in air were obtained [4]. The dose conversion factors (nGy h^{-1}/cpm) were 0.29 ± 0.05 in 2012, 0.28 ± 0.02 in 2013 and 0.13 ± 0.02 in 2014 by correlation between the total count rates and ambient dose rates in air. All obtained data from the car-borne survey were plotted on a distribution map using the inverse distance weighted technique. In this

study, the natural background radiation level (^{40}K, ^{238}U series and ^{232}Th series) was specified as 13 nGy h^{-1} [3] that had been observed before the F1-NPP accident to estimate its impact.

A fixed-point observation technique was also used to measure ambient dose rate in air. Measurements were made with a pocket-type CsI(Tl) scintillation survey meter (PDR-111, Aloka Co., Japan) at 22 locations (#1 - #22 in Fig. 2). The pocket survey meter was positioned at 1 m above the ground surface. Measurements were recorded over consecutive 30 s intervals during a total recording period of 5 min. A dose coefficient of 0.89 Sv Gy^{-1} was used to obtain the ambient dose rate in air. This dose coefficient was obtained from the intercomparison measurement with the NaI(Tl) scintillation spectrometer and CsI(Tl) scintillation survey meter at the same location and time.

Radioactivity concentration in soils

Soil samples were collected from a layer to 5 cm below the ground surface at 22 points (#1 - #22 in Fig. 2). Samples were sieved and less than 2 mm size particles were retained for this measurement. These samples were placed in cylindrical polypropylene containers (48 mm diameter × 55 mm). Radioactivity concentrations in each sample were measured with a high-purity germanium semiconductor detector (GMX10P, ORTEC, Oak Ridge, TN). The measurement time for each sample was set to 10000 s for the artificial radionuclides (^{134}Cs + ^{137}Cs). Additionally, nine soil samples collected in 2014 (#2, #4, #7, #9, #11, #13, #14, #16, #19 in Fig.2) were measured for 80000 s to identify the natural radionuclides (^{40}K, ^{238}U series and ^{232}Th series). Then, observed radioactivity concentrations from the natural radionuclides in soil samples were converted to the ambient dose rate in air at 1 m above the ground surface using the conversion factors (nGy h^{-1}/Bq kg^{-2}) [6].

RESULTS AND DISCUSSION

Figure 3 shows distribution maps of the ambient dose rate in air in 2011 (n = 95)[3], 2012 (n = 113), 2013 (n = 113) and 2014 (n = 844) measured by the car-borne surveys on Izu-Oshima Island. The distribution maps of the ambient dose rates in air for the whole island showed values were high in the northeast area located closer to the F1-NPP and low in the southwest area which may be considered as behind Mt. Mihara. On Izu-Oshima a northeast wind blows throughout the year. Mt. Mihara (764 m above sea level) is located at the center of the island. Therefore, the prevailing northeast wind changes direction, going to the southeast and northwest because of Mt. Mihara. As a result, it was assumed that the areas with observed ambient dose rates in air of 20 - 30 nGy h^{-1} had expanded toward the southeast and northwest sides of the island. Comparing surveys of three years showed that the ambient dose rates in air on the whole island were declining, though specific changes were observed in the distribution trend. Comparing the distributions of 2012 and 2013 indicated that the areas in the northeast with observed rates of 30 - 40 nGy h^{-1} had become smaller in size during 2013. The contaminated areas have been narrowed with each passing year, and areas are getting back to the background level rate (13 nGy h^{-1} [3]).

Table 1 shows the ambient dose rate in air at 1 m above the surface of the ground measured by fixed-point observation. The ambient dose rate in air in 2014 was decreased 34 % compared to that of 2011. From this, it was seen that the ambient dose rates in air were decreasing year by year with the same tendency as the distribution maps (Fig. 3). The ambient dose rate in air measured in 2005 was 13 nGy h^{-1} [3]. The value in August 2014, 3.5 y after the accident, was still 2.2 times higher than that before the accident. Here, the decay constant and environmental half-life (year) by artificial radionuclides were calculated with Equation (1) to estimate the change of ambient dose rate in air in the future:

$$D = D_L\,(exp\,(-\,\lambda_L t)),\ T = 0.693 / \lambda_L \qquad (1)$$

where D is ambient dose rate in air by artificial radionuclides (i.e., the difference between the measured ambient dose rate in air after the accident and the value, 13 nGy h^{-1}, measured in 2005[3]), D_L is the initial ambient dose rate (nGy h^{-1}) in air due to long half-life radionuclides (^{134}Cs+^{137}Cs), λ_L is decay constant, and t is elapsed years after the date that the radioactive plume reached Izu-Oshima (March 21, 2011[7]). In this paper, environmental half-life was defined as "apparent half-life" to distinguish it from physical half-life. Thus, the calculated half-life in the above included the affects of physical half-life, and the displacement of surface soil by rainfall and wind. This apparent half-life was described as the environmental half-life or ecological half-life in previous reports [8, 9]. Additionally, only the long half-life radionuclides were considered to examine the change of the dose rate in air in the future. As a result, D_L, the decay constant and the environmental half-life, were 33.9, 0.223 and 3.1 y, respectively. In consideration of the environment half-life obtained, it was expected that the ambient dose rate in air by artificial radionuclides would be the same as the level before the accident (13 nGy h^{-1} [3]) by about 4.3 y after March 2011. In the same metropolitan Tokyo governorate, Katsushika Ward (Fig. 1) had an estimated environmental half-life of 1.9 y based on changes of the ambient dose rates at 1 m above the ground surface [10]. While both Izu-Oshima and Katsushika Ward may be put into the same climatic division (i.e., high rainfall areas), there is a great divergence in their surrounding environment. On Izu-Oshima, 52% of the area is forested, but 80 to 90 % of Katsushika Ward is covered with asphalt or concrete. The surface contamination on asphalt or concrete is easily washed away by rainfall, so the environmental half-life on Izu-Oshima might be longer than that of Katsushika Ward.

Table 2 shows the radioactivity concentrations in soil samples in 2011 [3], 2012, 2013 and 2014. Those of ^{134}Cs and ^{137}Cs in 2014 decreased 72 % and 45 % compared to those of 2011, respectively. As well as the ambient dose rates in air, the radioactivity concentrations

of ^{137}Cs and ^{134}Cs in soil also showed a decreasing trend year by year. From the calculation with Equation (1), the environmental half-life (decay constant) of ^{134}Cs was 1.9 y (0.483) and that of ^{137}Cs was 2.4 y (0.304). The difference in the environmental half-lives between ^{134}Cs and ^{137}Cs which have the same chemical properties was due to the difference in their physical half-lives. In a report on environmental half-life after the Chernobyl nuclear power plant accident that occurred on April 26, 1986, it has been shown that environmental half-life is 47 y (measurement period: 10 y) in sandy soil up to 15 cm in depth [11] and 33 y (measurement period: 7 y) in pasture land [12]. Environmental half-lives obtained in this study were considerably shorter than these previously reported half-lives. While the annual rainfall around Chernobyl is very low, hand Izu-Oshima is an area of high rainfall and on the pathway of many typhoons. Izu-Oshima has the environment in which radionuclides are easily removed because of the large amounts of precipitation. Although it is difficult to directly compare environmental half-lives to that of previous reports because the measurement periods differed, it seemed that the environmental factors greatly affected the environmental half-life.

The natural radionuclides of ^{40}K, ^{238}U series (^{214}Bi, ^{214}Pb, ^{226}Ra) and ^{232}Th series (^{208}Tl) were also observed from soil samples (n = 9). The radioactivity concentrations and estimated ambient dose rates in air at 1 m above the ground surfaces are shown in Table 3. The dose conversion factors (nGy h^{-1}/Bq kg^{-1}) [6] used were 4.17×10^{-2} for ^{40}K, 4.01×10^{-1} for ^{214}Bi, 5.46×10^{-2} for ^{214}Pb, 1.25×10^{-3} for ^{226}Ra and 3.26×10^{-1} for ^{208}Tl. Total estimated natural ambient dose rate in air from soil samples was calculated to be 21 nGy h^{-1}. Since the number of soil samples was limited for identification of the natural radionuclides in this study, a slightly higher ambient dose rate in air from the artificial radionuclides was estimated in comparison to the previously reported value (i.e., 13 nGy h^{-1}, n = 137) [3].

The mean annual effective dose (mSv y^{-1}) before the accident was estimated to be 0.08 mSv y^{-1} [3]. The mean annual effective doses after the accident were estimated from the present survey results using the following equation:

$$E = D \times 10^6 \times f \times 24\ (\mathrm{h}) \times 365\ (\mathrm{day}) \qquad (2)$$

where E is mean annual effective dose (mSv y^{-1}), D is mean ambient dose rate in air (nGy h^{-1}), and f is dose coefficient (0.748 Sv Gy^{-1}) [13]. Here, the ambient dose rate indoors was assumed to be equivalent to that outdoors. Calculated annual effective doses in 2011, 2012, 2013 and 2014 were 0.29 mSv y^{-1}, 0.24 mSv y^{-1}, 0.22 mSv y^{-1} and 0.19 mSv y^{-1}, respectively. As the annual effective dose in Japan is 0.33 mSv y^{-1} [14], the annual effective doses after the accident were less than the average in Japan.

CONCLUSION

On Izu-Oshima Island, the ambient dose rate in air and radioactivity concentration in soil have been declining year by year since the F1-NPP accident. Also, change has been observed in the distribution trend of the ambient dose rate in air because of the island's environment. In consideration of the environment half-life obtained from the ambient dose rate in air (3.1 y), it was expected that the ambient dose rate in air would be the same as the level (13 nGy h^{-1}) before the accident by about 4.3 y after the accident. The environmental half-lives of ^{134}Cs (1.9 y) and ^{137}Cs (2.4 y) were shorter than their physical half-lives. Calculations of the annual effective doses for the three years after the accident showed that the doses were less than the average value in Japan (0.33 mSv y^{-1}).

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FIGURES

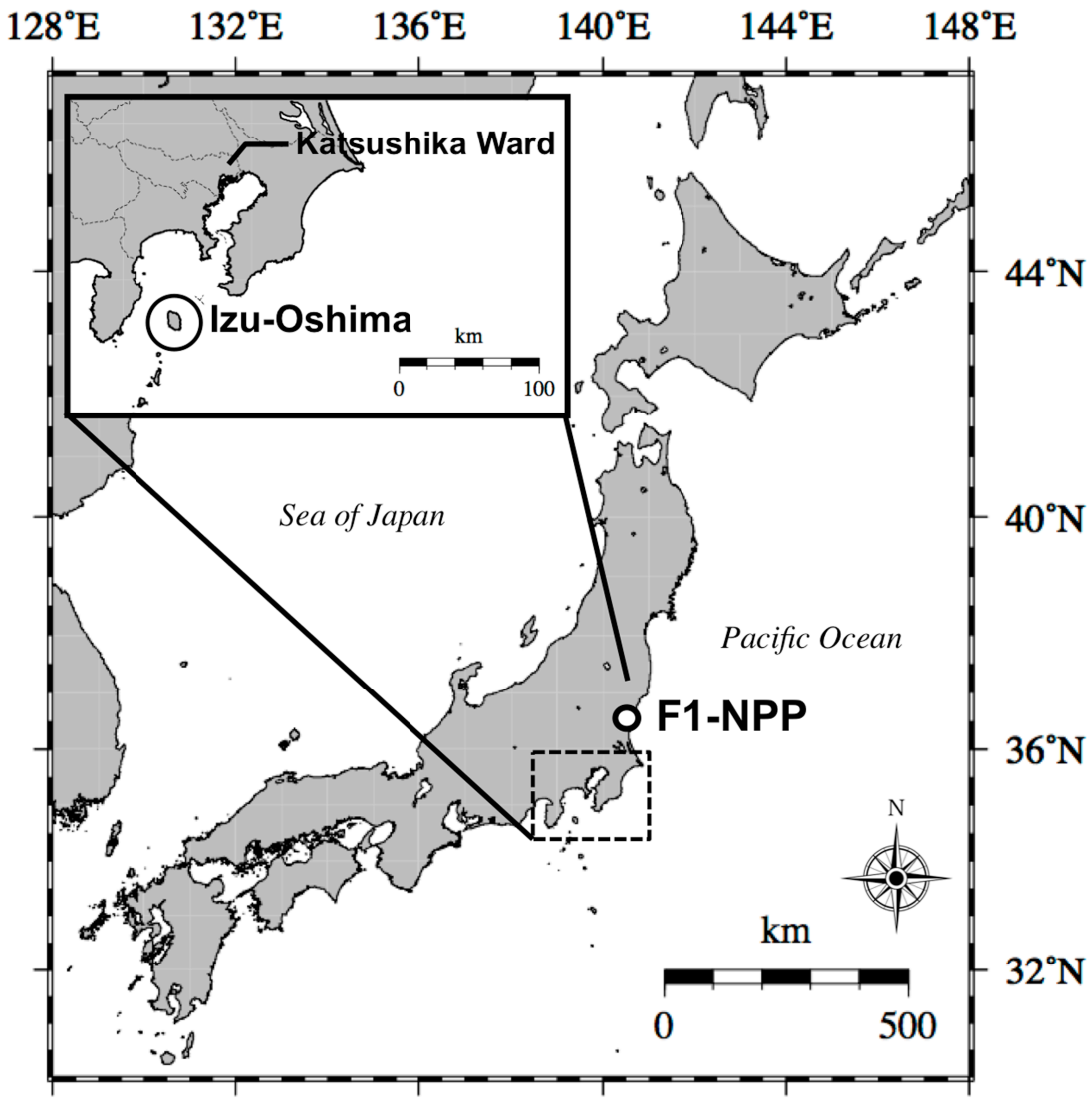

Figure 1 – The location of Izu-Oshima Island relative to the F1-NPP.

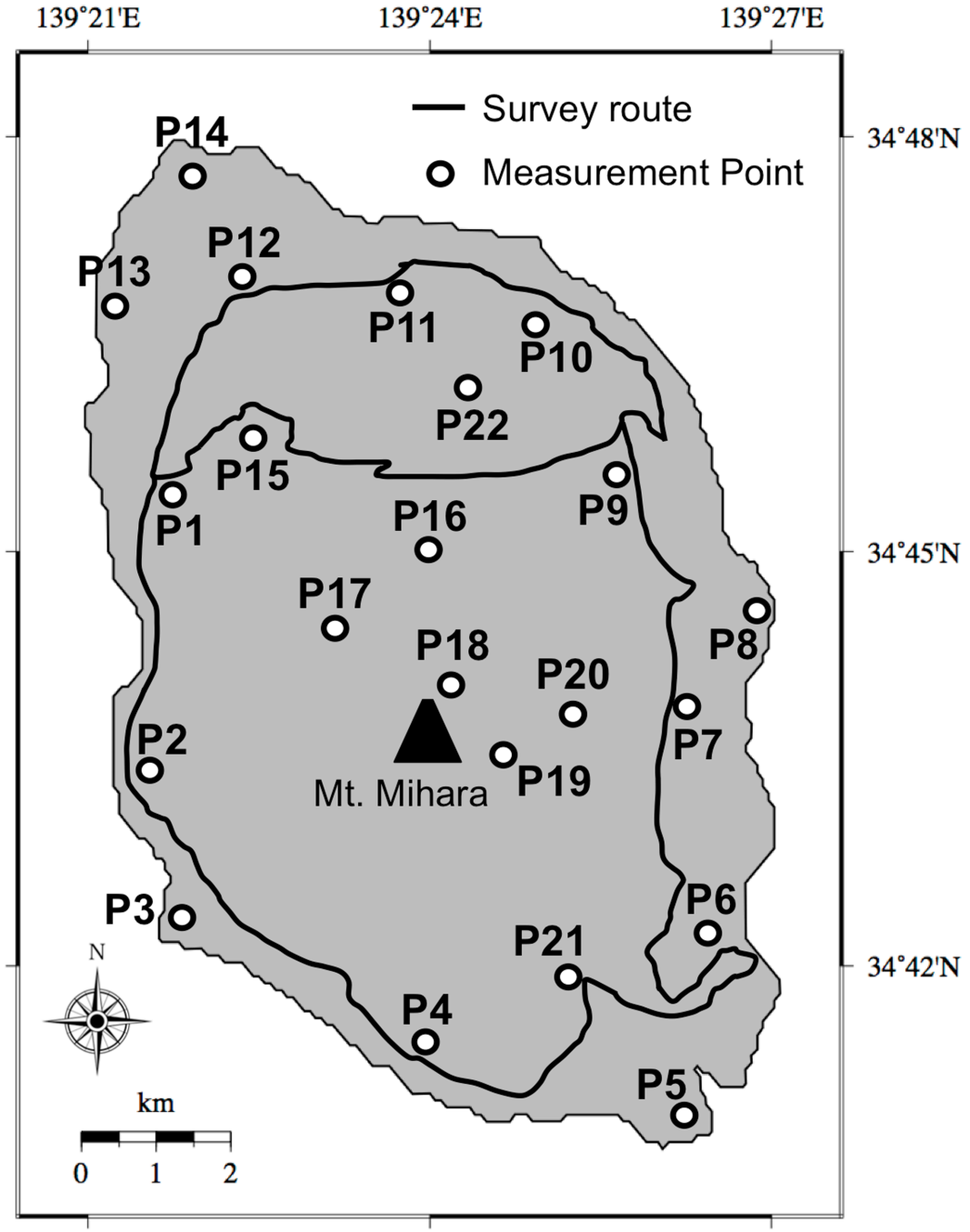

Figure 2 – Survey routes for measuring the ambient dose rates in air.

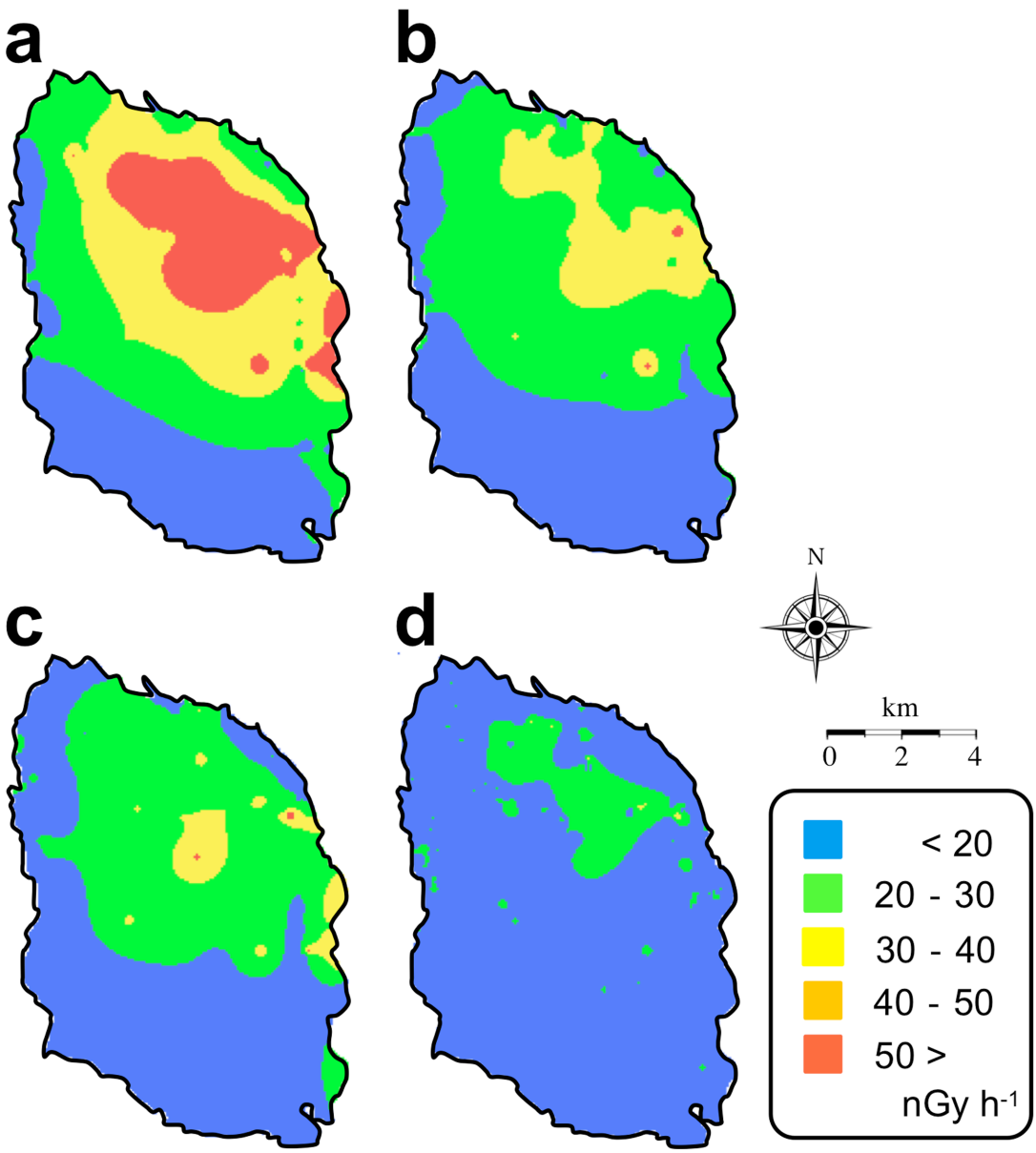

Figure 3 – The distribution maps of the ambient dose rates in air on Izu-Oshima Island obtained in 2011 (a)[3], 2012 (b), 2013 (c) and 2014 (d).

Table 1 Ambient dose rates in air (nGy h^{-1}) at 1 m above the ground surface.

	2011[3]	2012	2013	2014
Mean	44	36	34	29
Range	19 − 84	21 − 62	17 − 60	17 − 50

Table 2 Radioactivity concentrations (Bq kg^{-1}) of soil samples in 2011, 2012, 2013 and 2014.

	2011[3]	2012	2013	2014
^{134}Cs				
Mean	103	53	39	29
Range	6 - 395	8 -185	4 - 141	1 - 85
^{137}Cs				
Mean	134	87	73	75
Range	8 - 499	0 - 318	2 - 315	2 - 270

Table 3 Radioactivity concentrations (Bq kg^{-1}) of the natural radionuclides in soil samples and the estimated natural ambient dose rates in air at 1 m above the ground surface.

Natural radionuclides	Mean of radioactivity concentration (Bq kg^{-1})	Range of radioactivity concentration (Bq kg^{-1})	Mean of estimated natural ambient dose rate at 1 m above the ground surface (nGy h^{-1})	Range of estimated natural ambient dose rate at 1 m above the ground surface (nGy h^{-1})
^{40}K	364	306 – 439	15	13 – 18
^{214}Bi	8	4 – 11	3	2 – 4
^{214}Pb	8	5 – 11	0	0 – 1
^{226}Ra	45	36 – 62	0	0
^{208}Tl	6	3 – 8	2	1 – 3