福島県立医科大学 学術機関リポジトリ



Title	Difference in the Cesium Body Contents of Affected Area Residents Depending on the Evacuation Timepoint Following the 2011 Fukushima Nuclear Disaster
Author(s)	Igarashi, Yu; Kim, Eunjoo; Hashimoto, Shozo; Tani, Kotaro; Yajima, Kazuaki; Iimoto, Takeshi; Ishikawa, Tetsuo; Akashi, Makoto; Kurihara, Osamu
Citation	Health physics. 119(6): 733-745
Issue Date	2020-12
URL	http://ir.fmu.ac.jp/dspace/handle/123456789/1592
Rights	© 2020 Health Physics Society. This is a non-final version of an article published in final form in "Health Phys. 2020 Dec;119(6):733-745".
DOI	10.1097/HP.000000000001249
Text Version	author

This document is downloaded at: 2021-12-22T07:12:58Z

DIFFERENCE IN THE CS BODY CONTENTS OF AFFECTED AREA RESIDENTS $_1^1$ DEPENDING ON THE EVACUATION TIMEPOINT FOLLOWING THE 2011 FUKUSHIMA NUCLEAR DISASTER Yu Igarashi,^{*,†} Eunjoo Kim,^{*} Shozo Hashimoto,^{*} Kotaro Tani,^{*} Kazuaki Yajima,^{*} Takeshi Iimoto,^{*,†} Tetsuo Ishikawa,[‡] Makoto Akashi[§] and Osamu Kurihara^{*} *National Institutes for Quantum and Radiological Science and Technology, 4-9-1 Anagawa, Inage-ku, Chiba-city, Chiba, Japan [†]The University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa-city, Chiba, Japan [‡]Fukushima Medical University, 1-Hikarigaoka, Fukushima-city, Fukushima, Japan [§]Rurgasaki Public Health Center, 2983-1, Ryugasaki-shi, Ibaraki, Japan Corresponding author: Dr. Eunjoo Kim, Department of Radiation Measurement and Dose Assessment, Center for Advanced Radiation Emergency Medicine, Quantum Medical Science Directorate, National Institutes for Quantum and Radiological Science and Technology, 4-9-1 Anagawa, Inage-ku, Chibacity, Chiba, 263-8555, Japan. Tel: +81-43-206-4734, Fax: +81-43-284-1769. Email: kim.eunjoo@gst.go.jp

65

ABSTRACT

Estimating the internal thyroid dose received by residents involved in the 2011 Fukushima Daiichi Nuclear Power Plant (FDNPP) accident has been a challenging task because of the shortage of direct human measurements related to the largest contributing radioisotope to the dose, ¹³¹I. In a previous dose estimation, we used the results of whole-body counter (WBC) measurements targeting ¹³⁴Cs and ¹³⁷Cs, based on the assumption that these radioisotopes were incorporated at the same time as ¹³¹I in the early phase of the accident. The main purpose of this study was to clarify whether the trace of the early intake remained in the WBC measurements that were started several months after the accident. In the present work, WBC data of 1,639 persons from Namie town, one of the heavily contaminated municipalities, were analyzed together with their evacuation behavior data. The results demonstrated that the Cs detection rate in the WBC results was several times higher in the late evacuees (who evacuated outside the 20-km radius of the FDNPP at 3:00 p.m. [Japanese Local Time] on 12 March or later) compared to the prompt evacuees (who evacuated before 3:00 p.m. on 12 March). Among the adults, the Cs detection rates (and the 90th percentile values of the ¹³⁷Cs intake) of the prompt and late evacuees were about 20% (5.4×10^3 Bq) and 60% (1.6×10^4 Bq), respectively. Approximately 20% of the individuals analyzed were categorized as late evacuees. These differences in Cs would be caused by exposure to the radioactive plume in the afternoon on 12 March, which was likely to influence the late evacuees. On the other hand, the intake on 15 March when the largest release event occurred was expected to be relatively small for Namie town's residents. In conclusion, the trace of the early intake remained in the WBC measurements, although this would not necessarily be true for all subjects. The results obtained from this study would provide useful information for the reconstruction of the early internal thyroid doses from radioiodine in the future.

Key words: Fukushima, nuclear accident, internal dose, cesium, whole-body counter, evacuation

INTRODUCTION

The Fukushima Daiichi Nuclear Power Plant (FDNPP) run by Tokyo Electric Power Company suffered from a massive earthquake following the destructive tsunami that occurred on 11 March 2011. Consequently, the FDNPP reactors in operation (Units 1, 2, and 3) lost all cooling functions for the cores' heating after a normal shutdown, resulting in the release of huge amounts of radionuclides into the surrounding environment (National Diet of Japan 2012). The estimated release amounts of ¹³¹I and ¹³⁷Cs were 151 PBq and 14.5 PBq, respectively, which were roughly one-tenth and one-fifth of those in the Chernobyl accident (Katata et al. 2015). The FDNPP accident has been tentatively ranked as the highest level (Level 7) on the International Nuclear Event Scale (INES) (Nuclear Emergency Response Headquarters Government of Japan 2011).

The Japanese government repeatedly issued evacuation orders while expanding the on-alert area (Fukushima Prefecture 2018a). Approximately 78,000 residents who lived within a 20-km radius of the FDNPP (designated as the restricted area as of April 22, 2011) were requested to evacuate outside by an order at 6:25 p.m. (Japanese Local Time, hereinafter the same) on 12 March shortly after the hydrogen explosion event at Unit 1 (at 3:36 p.m.). Regarding Namie town focused in the present study, one of the heavily contaminated municipalities with the radionuclides (mainly ¹³⁴Cs, ¹³⁷Cs) due to this accident, the municipal government independently instructed about 19,600 residents living within the 20-km radius to evacuate in the morning on the same day (National Diet of Japan 2012). The eastern part of Namie town is located within the 20-km radius (**Fig. 1**).

Many studies have been performed to determine how much radiation dose the residents of Fukushima Prefecture received due to the accident (Ishikawa 2017; Kurihara 2018b). One common view of the various publications by Japanese experts was that following the FDNPP accident, both the external and internal doses of the residents were generally low, and the future radiation-induced health effects would be undetectable. However, there have been many concerns, particularly regarding the thyroid exposure to young children mainly from intake of ¹³¹I. This would have been related to the fact that many malignant cases have been unexpectedly identified in thyroid ultrasound examinations that

have been conducted as a part of the Fukushima Health Management Survey (FHMS) (Yasumura et al. 2012). The target subjects of these thyroid ultrasound examinations are all prefectural residents aged 0-18 years of March 11, 2011 (~370,000), and about 200 malignant cases (including suspected malignancies) were identified as of December 2017 (Fukushima Prefecture 2018b). The Exploratory Committee of the FHMS stated in their interim report that these malignant cases were unlikely to be caused by radiation exposure because of (1) the much lower doses compared to those in the Chernobyl NPP accident, (2) the earlier detection than the latent period of thyroid cancer, (3) no malignant cases in children aged ≤ 5 years, and (4) the absence of a significant regional difference in the detection rate compared to the companion studies conducted at other prefectures in Japan, although further studies were required (Fukushima Prefecture 2018c).

The internal thyroid doses of subjects have been estimated by Japanese scientists (Tokonami et al. 2012; Matsuda et al. 2013a; Kim et al. 2016a). The results of these studies indicated that the majority of individual thyroid doses were <20–30 mSv, although there might be a very limited number of subjects who received higher doses (Kamada et al. 2012). The major obstacle for the thyroid dose estimation is the shortage of direct human measurements of ¹³¹I with a physical half-life of 8.02 d. These measurements were available only for about one month after the FDNPP accident because of decay out. The number of these measurements totaled only-about 1,300 subjects of the public: direct thyroid measurements of 62 subjects by Tokonami et al. (2012), whole-body counter (WBC) measurements of 173 subjects by Matsuda et al. (2013a), and a screening survey of 1,080 subjects for internal thyroid exposure by the Nuclear Emergency Response Local Headquarters (NERLH) (Kim et al. 2016a). The NERLH screening survey covered relatively a large number of subjects, but it was conducted at only three municipalities located mostly outside of the 30-km radius of the FDNPP: Kawamata town, Iwaki city, and litate village. Residents who evacuated from the restricted area were not targeted in that survey.

To overcome the difficulty in the thyroid dose estimation, Hosoda et al. (2013) and Kim et al. (2016c) used the results of late WBC measurements by the National Institute of Radiological Sciences

(NIRS) and the Japan Atomic Energy Agency (JAEA), and they applied the derived intake ratios of ¹³¹I to ¹³⁴Cs (or ¹³⁷Cs). The numbers of subjects measured by the NIRS and the JAEA were 174 and 9,927 (by the end of January in 2012), respectively. These measurements were started several months after the accident (27 June at the NIRS and 11 July 2011 at the JAEA). As a result, only ¹³⁴Cs and/or ¹³⁷Cs could be detected. The thyroid dose estimation in the above studies were performed based on the assumption that the Cs body contents detected in the subjects came from the subjects' intake via inhalation at the same time in the early phase of the accident as well as ¹³¹I.

On the other hand, Matsuda et al. (2013b) analyzed their WBC measurement results of 372 subjects who were dispatched to Fukushima Prefecture at any timepoint from April 2011 to March 2012, and those authors proposed that the major route of intake of Cs would be inhalation until May in 2011, and then ingestion in June and later months. Nomura et al. (2016) assessed the relationship between selfprotection measures at the initial stage (i.e., evacuation, indoor sheltering) and the Cs body contents for 525 subjects who were examined with WBCs at Minami-soma Municipal General Hospital. These subjects were enrolled from among the residents who lived in Minami-soma city at the time of the accident; the southern part of the city was included in the restricted area. Based on their analyses of the WBC measurements, Nomura et al. deduced that the effect of the self-protection measures was insignificant. Although our study cannot be directly compared with the Matsuda and Nomura studies because of the differences of subjects and measurements, the conclusions from these two studies seem to pose a critical problem regarding the thyroid dose estimations by Hosoda et al. and Kim et al.; namely, the trace of the early intake might not have remained in the WBC measurements by the NIRS and the JAEA because of the possible ingestion of Cs from the daily diet and the relatively short biological half-life of Cs, i.e., ~100 days for adults and ~30 days for children (5 yrs) (ICRP 1990). We therefore conducted the present study to clarify this issue, based on analyses of the relationship between the Cs content and the evacuation times of the Namie residents. The thyroid dose estimation was not fully addressed in this paper; however, the present study expects to provide useful information for the reconstruction of the early internal thyroid doses by using Cs as a tracer of radioiodine.

MATERIALS AND METHODS

Subjects and their behavior data

The individuals analyzed in this study were all residents of the Namie town, Japan who underwent WBC measurements at the JAEA by the end of January 2012 (Momose et al. 2012) and responded to self-administered questionnaires about their behaviors for the first 4 months after the March 2011 accident as part of the Basic Survey for the external dose estimation (Ishikawa et al. 2015), one of the main components in the FHMS. The total of 1,639 subjects and their composition by age and gender are shown in **Fig. 2**. Personal behavior data were generated from answer sheets of the above questionnaires and contained the whereabouts (the place name and its latitude and longitude), the time spent indoors/outdoors or moving, and the type of the building where the person stayed (e.g., a wooden house, a concrete building), although not all of the items were analyzed in this study. These data were provided hourly until 25 March and daily from 26 March to 11 July (only for representative places to stay and commute). We found that the data for April and later months were missing for most of the 1,639 subjects.

The distance from the subject at each time to the FDNPP (latitude 37.421071 N, longitude 141.032755 E), one of the important indexes in the analyses, was calculated by using a program package of the Python language (Geod) (Python 2019). The locations of the subjects were visualized on maps using our own program written in the Python language as necessary. For the two group analyses described below in the results, the locations of individuals when they moved residences were determined assuming a uniform linear motion between the places of departure and destination. The use of the personal behavior data in this study was approved by the Research Ethics Committee of Japan's National Institutes for Quantum and Radiological Science and Technology (QST) – National Institute of Radiological Science (NIRS) QST-NIRS (13-011).

Whole-body counter measurements

Details of the WBC measurements of Fukushima Prefecture residents by the JAEA (for the first year after the accident) are described elsewhere (Kurihara et al. 2018a). Briefly, the JAEA started the measurements on 11 July 2011 (2 months after the accident). The WBC units that were mainly used were two standing-type units and one chair-type unit at two JAEA sites in Ibaraki Prefecture because no WBCs were available for measurements of the residents in Fukushima Prefecture at that time. Surface contamination check was performed using a closed-end Geiger-Muller (GM) survey meter before the WBC measurements, which would have minimized false-positive detection due to the remained contaminant on clothes. The nominal minimum detectable activity (MDA) values for ¹³⁴Cs and ¹³⁷Cs were 300–400 Bq for a counting time of 2 min. These values were evaluated taking into account the variation of background counts (including the component from ⁴⁰K in the human body) and were increased by about 100 Bg compared to those before the accident. The statistical uncertainties in the net peak area corresponding to 300Bq were about 20-45%. Subjects were selected by the Fukushima prefectural government; residents living in the municipalities near the FDNPP were prioritized. Residents of Namie town were measured at relatively early times (mostly during the period from July to September in 2011). Children aged under 3 years (at that measurement timepoint) were excluded from the subject populations because of difficulties in the measurements with the WBC units.

Calculations of intake and committed effective dose

The calculations of intake and the committed effective dose (CED) based on the results of the WBC measurements were performed using the following equations:

$$I_{i,134} = \frac{M_{134}}{R_{i,134}(t)} \tag{1}$$

$$I_{i,137} = \frac{M_{137}}{R_{i,137}(t)} \tag{2}$$

where, the subscript *i* is the age group defined in the ICRP publications: 5 yr, 10 yr, 15 yr, and Adult (ICRP 1995); note that 0 yr and 1 yr age groups are not included in subjects of the WBC measurements. The subscripts 134 and 137 indicate ¹³⁴Cs and ¹³⁷Cs, respectively; *M* is the Cs body content determined by the WBC measurements; *R* is the whole-body retention rate as a function of the elapsed time after intake (until measurement); *e* is the effective dose per unit intake (DPUI) in the case of Type F compounds with an activity median aerodynamic diameter (AMAD) of 1 μ m.

The retention rate and DPUI values were taken from the database of the MONDAL system developed by Ishigure et al. (2004) and the ICRP database of dose coefficients (ICRP 1998). The intake scenario was assumed to be acute intake via inhalation on 12 March 2011, as in previous studies (Kim et al. 2016c; Kurihara et al. 2018a). This intake scenario was set to avoid underestimation of individual doses rather than to obtain realistic dose estimates on the assumption that dietary intake would have be minimized due to prompt regulations for contaminated food and drink. Persons who occasionally consumed highly contaminated foodstuffs such as wild animals or home-grown vegetables were found to be very few in number (Hayano et al 2013; Tsubokura et al. 2013).

In the present analyses, the intake was treated as zero for the subjects with negative detection. Here, the negative detection is the cases where the Cs body content was neither detected nor exceeded the nominal MDA values, whereas the positive detection (described later) is the cases where the Cs content was larger than the nominal MDA values. Several percentile values of intake and the CED were obtained using the Excel PERCENTILE function. For explanation, the Cs intake is obtained using the Cs whole-body retention rate, as shown in the above equations. It should be noted that the Cs whole-body retention rates are considerably different between the age groups and rapidly decreases for the younger age groups, which means that even a small amount Cs content can be converted into a very large Cs intake (or a very large CED) for children with increasing the elapsed time. This needs to be carefully considered as described in Discussion.

RESULTS

Cs detection rate in the WBC measurements

Table 1 provides the numbers of subjects with positive detection of 134 Cs or 137 Cs for each age and gender group along with the Cs detection rate. As shown, the Cs detection rate was higher the group is older, and the gender difference in the Cs detection rate was significant in the Adult group. The highest Cs detection rates were for the Adult male group (n=91): 51.6% for 134 Cs and 56.0% for 137 Cs.

The relationship between the movement of resident after the accident and the Cs detection rate

Fig. 3 illustrates the whereabouts of the 1,639 residents of Namie town on the first few days after the March 2011 accident; note that the locations of some individuals are overlapped at the same places. Note that the locations of residents when they were moving were determined on this figure assuming a uniform linear motion between the whereabouts of departure and destination. As demonstrated in the figure, the majority of the Namie residents originally stayed near the coastal area of Namie town within the 20-km radius and then started to evacuate rapidly on the morning of 12 March. The residents remaining near the FDNPP were sparse as of 12:00 a.m. on 15 March. It was found that the major evacuation route from Namie town was twofold: a Tsushima route (toward the northwest from the FDNPP) and a Minami soma route (toward the north from the FDNPP). Tsushima is the west district in Namie, and we defined the Tsushima area as the areas outside the 20-km radius of Namie town in this study (**Fig. 1**).

Fig. 4 shows the trends of the numbers of individuals (the Adult group) staying within the 20-km radius (<20-km), those outside the 20-km radius (\geq 20-km), and those who moved during the period between 12:00 a.m. on 12 March and 12:00 a.m. on 16 March. The number of individuals (<20-km) started decreasing in the morning of 12 March, suggesting that the evacuation order by the municipal government functioned well. The number of people moving fluctuated on 13 March and later, indicating that evacuations were repeatedly performed during the daytime. It was also clear that some

individuals returned to inside the 20-km radius after they had evacuated outside the radius. The same figures for the other age groups and all of the 1,639 individuals are provided in **Appendix 1**.

Fig. 5 illustrates the trends of the 317 individuals in the Adult group, separating those who stayed within the 20-km radius and those who moved outside the 20-km radius. Here the distances from the whereabouts of individuals moving to the FDNPP at each timepoint were determined in a manner similar to that shown in **Fig. 3**. In the figure, 116 individuals with positive detection and 201 individuals with negative detection regarding ¹³⁷Cs (see **Table 1**) are also provided separately, along with the trends of the ¹³⁷Cs detection rate for these two groups. This analysis was performed in reference to Nomura et al. (2016). As demonstrated, the individuals with positive detection moved to remote places at relatively early times; however, the time of evacuation for these individuals appeared to be somewhat late compared to those with negative detection.

One remarkable finding was that the detection rate for the subjects who remained within the 20-km radius started increasing at around noon on 12 March and reached the maximum (77%, 27 of 35 individuals) at 8:00 p.m. on the same day, and then gradually decreased afterwards. A similar tendency was seen in the other age groups and all of the 1,639 individuals (**Appendix 2**). The detection rate for the individuals who stayed outside the 20-km radius did not significantly change with time; note that the ¹³⁷Cs detection rate in the Adult group is 36.6% (**Table 1**), which is indicated in the figure's lower panel.

Difference in the internal dose due to the timepoint of evacuation

Fig. 6 shows the results of the comparison of the ¹³⁷Cs body content between two subject groups in box-and-whisker plots for each age group; one group is the Namie residents who evacuated outside the 20-km radius before 3:00 p.m. on 12 March (G1: the prompt evacuation group), and the other group is those who evacuated outside the 20-km radius on 3:00 p.m. on 12 March or later (G2: the late evacuation group). The individuals with negative detection are marked as nearly zero in the figure.

As shown in Fig. 6, the distribution of the ¹³⁷Cs body content values was clearly different between

the two groups; the ¹³⁷Cs body content values of the G2 group were significantly higher than those of the G1 group. One important finding (in **Fig. 6**) was that all of the subjects with significant ¹³⁷Cs detection among the G1 group were categorized as outliers (in accordance with the definition of a box-and-whisker plot); namely, all of the individuals other than outliers were those with negative detection. The number of such outliers was small in the G2 group.

Table 2 provides a comparison of numbers of the G1 and G2 groups. The number of G2 group subjects was a total of 388 in all age groups; this was 23.7% of the 1,639 individuals. **Figs 7** and **8** provide the results of our comparisons of the ¹³⁷Cs intake and the CED from ¹³⁴Cs and ¹³⁷Cs for the G1 and G2 groups, respectively. The data corresponding to these two figures are provided as different percentiles in **Tables 3** and **4**. The maximum CED was 2.3 mSv, which was detected in a subject in the 5 yr age group and the G1 group, whereas 0.72 mSv was the maximum CED in the Adult group. The 50th percentile (median) CED was determined in the 15 yr and Adult groups of the G2 group, but not for the other groups (**Table 4**).

Fig. 9 compares the ¹³⁷Cs body content, the ¹³⁷Cs intake, and the CED from ¹³⁴Cs and ¹³⁷Cs for the 15 yr and Adult groups in the following four groups; the first two groups are prompt and late evacuees who moved toward the northwest (Tsushima): G1(T) and G2(T), and the last two groups are prompt and late evacuees toward the north (Minami-soma): G1(M) and G2(M). The numbers of G1(T), G2(T), G1(M), and G2(M) subjects were 231, 37, 64, and 24, respectively. Here, the definitions of G1 and G2 are the same as those described above. The evacuees to Tsushima and Minami-soma were categorized by the duration of time (>24 hr during the period between 12:00 a.m. on 12 March and 12:00 a.m. on 14 March) staying in either of the two areas. **Table 5** provides the subject numbers and difference percentiles of CEDs for each of these groups. The younger age groups (5 yr and 10 yr) were not included because of the low Cs detection rates not enough to provide the corresponding data in the table.

DISCUSSION

As mentioned in the Introduction, it is crucial to clarify whether late WBC measurements taken by the NIRS and the JAEA have a trace of the early intake of radionuclides. If not, these measurements will result in unavailable in the thyroid dose estimation for Fukushima residents. This study focused on residents of Namie town, one of the most affected municipalities by the FDNPP accident and analyzed the relationship between results of the WBC measurements and the timepoint of evacuation using available data of 1,639 residents of Namie town. The most important finding of this study was the definite difference in the CED values between the prompt and late evacuees. Our results also indicate that the most decisive time for the Namie residents was the afternoon of 12 March (**Fig. 5**).

Major release events of radionuclides from the FDNPP occurred on 12, 15-16, and 20-21 March, and so on (Tsuruta et al. 2017). Among these events, the radioactive plume caused by the hydrogen explosion at Unit 1 on the afternoon of 12 March would be related to our above-described findings. Several investigations determined the precise behavior of this plume; it flowed toward the northnorthwest (NNW) direction at the beginning, which was evident from the result of soil sampling adjacent to the FDNPP site, and the plume was then expected to change its direction toward the northnortheast (NNE) (Chino et al. 2016). The time trends of the airborne concentration of Cs near the FDNPP were also revealed by analyses of radionuclides collected on filter tapes installed in suspended particulate matter (SPM) monitors, demonstrating that the airborne concentration of Cs at sites located in the northern area along the coast increased shortly after the explosion event and reached the maximum at night: >100 Bq m⁻³ for 7 hr after 9:00 p.m. with a maximum concentration of about 575 Bq m^{-3} for ¹³⁷Cs at the site located 25 km north of the FDNPP (Minami-soma city) (Tsuruta et al. 2014). Another study using the same SPM technique demonstrated that the highest ¹³⁷Cs airborne concentration of 13,600 Bq m⁻³ was observed at the site located 3.2 km west-northwest (WNW) (Futaba town) at 3:00 p.m. before the explosion, as a result of a vent operation at Unit 1 (Tsuruta et al. 2018). The first peak in the 137 Cs airborne concentration (55.7 Bg m⁻³) was also observed at 9:00 a.m. at this site.

The Cs detection rate was several times higher in our present G2 group compared to the G1 group

(**Table 2**). The Cs detection rate was >60% in the G2 group adults but only ~20% in the G1 group adults. A similar observation to this was found in our previous study (Kunishima et al. 2017). This result may also indicate that the intake of radionuclides due to the largest release event on March 15–16 was small in the residents of Namie town thanks to their evacuation to remote places (**Fig. 3**). The 90th-percentile values of the ¹³⁷Cs intake for the G1 and G2 groups were 5.4×10^3 Bq and 1.6×10^4 Bq, respectively (**Table 3**). This difference (~10⁴ Bq) would be an index for the magnitude of the ¹³⁷Cs intake on 12 March (for individuals with relatively high internal doses), considering that most of the G2 group evacuated outside the 20-km radius, as did most of the G1 group (**Fig. 4**), resulting in similar internal exposure for the two groups on 15 March and later.

The evidence that the CED values of the evacuees who moved to Minami-soma were generally higher than those of the evacuees who moved to Tsushima district (**Fig. 9**, **Table 5**) contributes to our understanding of the intake on 12 March together with the studies described in the paragraph above. The intake on 12 March is also important in terms of evaluating the contribution of the internal thyroid dose from the other short-lived radionuclides (e.g., ¹³²Te/¹³²I, ¹³³I) (Shinkarev et al. 2015; Ohba et al. 2017).

To estimate the internal thyroid dose from the WBC measurements which were performed since July 2011, it is necessary to determine the intake ratio of ¹³¹I to ¹³⁴Cs (or ¹³⁷Cs). Our previous study derived the intake ratio (¹³¹L^{/137}Cs) from two independent direct measurements of residents in Kawamata and Iitate (see **Fig. 1**), namely, direct thyroid measurements of children (the screening survey by the NERLH) and the WBC measurements of adults by the JAEA (Kim et al. 2016b). The time trend of the ambient dose equivalent rate suggested that the radioactive plume first reached Kawamata town and Iitate village in the afternoon of 15 March, not on 12 March. Therefore, the difference in the radionuclide composition in the plumes on these two days should be examined, when considering the possible intake by late evacuees from Namie town. The relevant information will be obtained from SPM samples in the future. The derived intake ratio was 3, which was much smaller than the radioactivity ratio in the core inventory, nearly 10 (Katata et al. 2015). It was considered that

the probable main reason for this discrepancy lay in the relatively-low thyroid iodine uptake in Japanese compared to the corresponding parameter used in the iodine biokinetic model for internal dose assessment, although further studies are needed on this regard (Kim et al. 2016b).

Finally, it should be addressed that late WBC measurement data of children are considered difficult for use in the reconstruction of the early internal dose in the FDNPP accident at this moment. The intake amount of radionuclides is expected to be larger in adults than in children as long as the same intake scenario is applied to both groups. In the case of Cs, the dose coefficient due to intake is relatively similar in the different age groups. As a result, the CED should have been higher in the Adult group than in the Child group in the WBC measurements; however, they were not higher (**Table 4**). We suspect that the CEDs of a relatively small number of children with positive Cs detection were likely to be overestimated due to their accidental intake via ingestion during the several months after the accident and/or contamination on their clothes at the time of the WBC measurements (Kurihara et al. 2018a).

Such possibilities were also suggested by our findings that the intakes and the CEDs of the outliers were not related to the timepoint of evacuation, in particular for the 5 yr age group (**Figs. 7, 8**). Moreover, although food safety regulations were enforced shortly after the FDNPP accident (Hamada and Ogino 2012), daily intake via ingestion might occur in subjects. Several surveys have reported that dietary intake amount of Cs was 1 Bq or less per day on average in the first year after the accident (Koizumi et al. 2012; Harada et al. 2013). Even such a small intake amount could partly contribute to the detected Cs contents if continued, resulting in overestimations of CEDs in particular for children by applying the acute intake scenario described before. A more appropriate intake scenario is thus needed to interpret the late WBC measurements, focusing on the different age groups with each unique Cs biological half-life. In conclusion, it was deduced that the trace of the early intake remained in the WBC measurements, although this would not be necessarily true for all the individuals examined. Further analyses on the data presented in this study are essential to find out the critical group more

precisely.

CONCLUSION

We analyzed the WBC measurement results of 1,639 residents of Namie town together with their behavioral data after the 11 March, 2011 nuclear power accident at the FDNPP, and our findings revealed that the timepoint of evacuation influenced the individual internal dose from Cs. We categorized the individuals into two groups depending on the timepoints at which they evacuated to outside of the 20-km radius of the FDNPP. As a result, here was found an evident difference in the detection rate and intake of Cs between the prompt and late evacuees. It was deduced that this difference would be caused by exposure to the radioactive plume released in the afternoon of 12 March, in particular for the late evacuees who remained within the 20-km radius as of 3:00 p.m. on that critical day. Regarding the adults, the Cs detection rate exceeded 60% for the late evacuees, was only about 20% for the prompt evacuees. Further studies are necessary to appropriately interpret the results of the available WBC measurements for estimating the internal thyroid doses of Fukushima Prefecture's residents due to their intake of radioiodine. However, the results obtained from this study would provide useful information for the reconstruction of the early internal thyroid doses from radioiodine in the future.

FUNDING

This study was funded by the Ministry of the Environment of Japan.

REFERENCES

Chino M, Terada H, Nagai H, Katata G, Mikami S, Torii T, Saito K, Nishizawa Y. Utilization of ¹³⁴Cs/¹³⁷Cs in the environment to identify the reactor units that caused atmospheric releases during the Fukushima Daiichi accident. Sci Rep 6: 31376; 2016.

Fukushima Prefecture. Transition of evacuation area [online]. 2018a (in Japanese). Available at *http://www.pref.fukushima.lg.jp/site/portal/cat01-more.html*. Accessed 6 August 2019.

Fukushima Prefecture. Result of thyroid ultrasound examinations [online]. 2018b (in Japanese) Available at *http://www.pref.fukushima.lg.jp/site/portal/43-7.html*. Accessed 6 August 2019.

Fukushima Prefecture. The interim report on the Fukushima Health Management Survey. [online].
2018c (in Japanese). Available at *http://www.pref.fukushima.lg.jp/uploaded/attachment/158522.pdf*.
Accessed 6 August 2019.

Hamada N, Ogino H. Food safety regulations; what we learned from the Fukushima nuclear accident. J Environ Radioact 111: 83–99; 2012.

Harada K H, Fujii Y, Adachi A, Tsukidate A, Asai F, Koizumi A. Dietary intake of radiocesium in adult residents in Fukushima Prefecture and neighboring regions after the Fukushima Nuclear Power Plant accident: 24-h food-duplicate survey in December 2011. Environ Sci Technol 47: 2520–2526; 2013.

Hayano R S, Tsubokura M, Miyazaki M, Satou H, Sato K, Misaki S, Sakuma Y. Internal radiocesium contamination of adults and children in Fukushima 7 to 20 months after the Fukushima NPP accident as measured by extensive whole-body-counter surveys. Proc Jpn Acad Ser B89: 157–163; 2013.

Hosoda M, Tokonami S, Akiba S, Kurihara O, Sorimachi A, Ishikawa T, Momose T, Nakano T, Mariya Y, Kashiwakura I. Estimation of internal exposure of the thyroid to ¹³¹I on the basis of ¹³⁴Cs accumulated in the body among evacuees of the Fukushima Daiichi Nuclear Power Station accident. Environ Inter 61:73–76; 2013.

International Commission on Radiological Protection. Age-dependent Doses to Members of the Public from Intake of Radionuclides – Part 1. Oxford: ICRP; Publication 56, Ann ICRP 20(2); 1990.

International Commission on Radiological Protection. Age-dependent Doses to Members of the Public from Intake of Radionuclides: Part 4 Inhalation Dose Coefficients. Oxford: ICRP; Publication 71, Ann ICRP 25(3–4); 1995.

International Commission on Radiological Protection. ICRP Database of Dose Coefficients: Workers and Members of the Public version 3. ICRP CD1; 1998.

Ishigure N, Matsumoto M, Nakano T, Enomoto H. Development of software for internal dose calculation from bioassay measurements. Radiat Protect Dosim 109:235–242; 2004. [MONDAL3 (updated version), online]. Available at *https://www.nirs.qst.go.jp/db/anzendb/RPD/mondal3.php*. Accessed 6 August 2019.

Ishikawa T, Yasumura S, Ozasa K, Kobashi G, Yasuda H, Miyazaki M, Akahane K, Yonai S, Ohtsuru A, Sakai A, Sakata R, Kamiya K, Abe M. The Fukushima Health Management Survey: estimation of external doses to residents in Fukushima Prefecture. Sci Rep 5:12712; 2015.

Ishikawa T. Radiation Doses and Associated Risk Form the Fukushima Nuclear Accident: A Review of Recent Publications. Asia Pac J Public Health 29(2S): 18S-28S; 2017.

Kamada N, Saito O, Endo S, Kimura A, Shizuma K. Radiation doses among residents living 37 km northwest of the Fukushima Dai-ichi Nuclear Power Plant. J Environ Radioact 110:84–89; 2012.

Katata G, Chino M, Kobayashi T, Terada H, Ota M, Nagai H, Kajino M, Draxler R, Hort M. C, Malo A, Torii T, Sanada Y. Detailed source term estimation of the atmospheric release for the Fukushima Daiichi Nuclear Power Station accident by coupling simulations of an atmospheric dispersion model with an improved deposition scheme and oceanic dispersion model. Atoms Chem Phys 15:1029–1070; 2015.

Kim E, Kurihara O, Kunishima N, Momose T, Ishikawa T, Akashi M. Internal thyroid doses to Fukushima residents—estimation and issues remaining. J Radiat Res 57: i118–i126; 2016a.

Kim E, Kurihara O, Tani K, Ohmachi Y, Fukutsu K, Sakai K, Akashi M. Intake ratio of ¹³¹I to ¹³⁷Cs derived from thyroid and whole-body doses to Fukushima residents. Radiat Protect Dosim 168:408–418; 2016b.

Kim E, Kurihara O, Kunishima N, Nakano T, Tani K, Hachiya M, Momomse T, Ishikawa T, Tokonami S, Hosoda M, Akashi M. Early Intake of Radiocesium by Residents Living Near the TEPCO Fukushima Dai-ichi Nuclear Power Plant after the Accident. Part 1: Internal Doses Based on Whole-body Measurements by NIRS. Health Phys 111: 451–464; 2016c.

Koizumi A, Harada K H, Niisoe T, Adachi A, Fujii Y, Hitomi T, Kobayashi H, Wada Y, Watanabe T, Ishikawa H. Preliminary assessment of ecological exposure of adult residents in Fukushima Prefecture to radioactive cesium through ingestion and inhalation. Environ Health Prev Med 17: 292–298; 2012.

Kunishima N, Kurihara O, Kim E, Ishikawa T, Nakano T, Fukutsu K, Tani K, Furuyama K, Hashimoto S, Hachiya M, Naoi Y, Akashi M. Early Intake of Radiocesium by Residents Living Near the TEPCO Fukushima Dai-ichi Nuclear Power Plant After the Accident. Part 2: Relationship Between Internal Dose and Evacuation Behavior in Individuals. Health Phys 112: 512-525; 2017.

Kurihara O, Li C, Lopez M A, Kim E, Tani K, Nakano T, Takada C, Momose T, Akashi M. Experiences of Population Monitoring Using Whole-Body Counters in Response to the Fukushima Nuclear Accident. Health Phys 115: 259-274; 2018a.

Kurihara O. Review: External and internal dose assessments of Fukushima residents after the 2011 nuclear disaster. J Natl Inst Public Health 67: 11-20; 2018b. Available at

https://www.niph.go.jp/journal/data/67-1/201867010003.pdf. Accessed 6 August 2019.

Matsuda N, Kumagai A, Ohtsuru A, Morita N, Miura M, Yoshida M, Kudo T, Takamura N, Yamashita S. Assessment of Internal Exposure Doses in Fukushima by a Whole Body Counter Within One Month after the Nuclear Power Plant Accident. Radiat Res 179:663–668; 2013a.

Matsuda N, Morita N, Miura M, Yamauchi M, Kudo T, Usa T. Internal radioactivity of temporary residents in Fukushima within one year after the radiological accident. J Environ Occup Sci 2: 123–130; 2013b.

Momose T, Takada C, Nakagawa T, Kanai K, Kurihara O, Tsujimura N, Ohi Y, Murayama T, Suzuki T, Uezu Y, Furuta S. Whole-body Counting of Fukushima Residents after the TEPCO Fukushima Daiichi Nuclear Power Station Accident. In: Proceedings of the 1st NIRS Symposium on Reconstruction of Early Internal Dose in the TEPCO Fukushima Daiichi Nuclear Power Station Accident. Chiba, Japan: National Institute of Radiological Sciences; NIRS-M-252; 2012: 67-82.

Available at *https://www.nirs.qst.go.jp/publication/irregular/02.html*. Accessed 6 August 2019.

National Diet of Japan. The official report of The Fukushima Nuclear Accident Independent Investigation Commission [online]. 2012.

Available at *http://warp.da.ndl.go.jp/info:ndljp/pid/3856371/naiic.go.jp/en*. Accessed 6 August 2019.

Nomura S, Tsubokura M, Gilmour S, Hayano RS, Watanabe YN, Kami M, Kanazawa Y, Oikawa T. An evaluation of early countermeasures to reduce the risk of internal radiation exposure after the Fukushima nuclear incident in Japan. Health Policy and Planning 31: 425–433; 2016.

Nuclear Emergency Response Headquarters Government of Japan. Report of Japan Government to the IAEA Ministerial Conference on Nuclear Safety — The Accident at TEPCO's Fukushima Nuclear Power Stations [online]. 2011.

Available at *http://japan.kantei.go.jp/kan/topics/201106/iaea_houkokusho_e.html*. Accessed 6 August 2019.

Ohba T, Hasegawa A, Kohayagawa Y, Kondo H, Suzuki G. Body Surface Contamination Levels of Residents under Different Evacuation Scenarios after the Fukushima Daiichi Nuclear Power Plant Accident. Health Phys 113: 175-182; 2017.

Python. Available at *https://www.python.org/about/*. Accessed 6 August 2019.

Shinkarev SM, Kotenko KV, Granovskaya EO, Yatsenko VN, Imanaka T, Hoshi M. Estimation of the contribution of short-lived radioiodines to the thyroid dose for the public in case of inhalation intake following the Fukushima accident. Radiat Prot Dosim 164: 51-56; 2015.

Tokonami S, Hosoda M, Akiba S, Sorimachi A, Kashiwakura I, Balonov M. Thyroid doses for evacuees from the Fukushima nuclear accident. Sci Rep 2:507; 2012.

Tsubokura M, Kato S, Nihei M, Sakuma Y, Furutani T, Uehara K, Sugimoto A, Nomura S, Hayano R, Kami M, Watanabe H, Endo Y. Limited internal radiation exposure associated with resettlement to a radiation-contaminated homeland after the Fukushima Daiichi nuclear disaster. PLOS ONE 8: e81909; 2013.

Tsuruta H, Oura Y, Ebihara M, Ohara T, Nakajima T. First retrieval of hourly atmospheric radionuclides just after the Fukushima accident by analyzing filter-tapes of operational air pollution monitoring station. Sci Rep 4: 6717; 2014.

Tsuruta H, Oura Y, Ebihara M, Moriguchi Y, Ohara T, Nakajima T. Spatio-Temporal Distribution of Atmospheric Radiocesium in Eastern Japan just after the TEPCO Fukushima Daiichi Nuclear Power Plant Accident – Analysis of Used Filter-Tapes of SPM Monitors in Air Quality Monitoring Stations. Earozoru Kenkyu 32: 244-254; 2017 (in Japanese).

Tsuruta H, Oura Y, Ebihara M, Moriguchi Y, Ohara T, Nakajima T. Time-series analysis of atmospheric radiocesium at two SPM monitoring sites near the Fukushima Daiichi Nuclear Power Plant just after the Fukushima accident on March 11, 2011. Geochemical J 52: 103-121; 2018.

Yasumura S, Hosoya M, Yamashita S, Kamiya K, Abe M, Akashi M, Kodama K, Ozasa K. Study Protocol for the Fukushima Health Management Survey. J Epidemiol 22:375–383; 2012.

Footnotes

 *National Institutes for Quantum and Radiological Sciences and Technology, 4-9-1 Anagawa, Inage-ku, Chiba-city, Chiba, Japan **Fig. 1** The locations of Fukushima Prefecture, Namie town, and the FDNPP in Japan with the 20-km radius from the FDNPP.

Fig. 2 Composition of the 1,639 subjects by age and gender. The ages are as of 12 March 2011.

Fig. 3 The whereabouts of the 1,639 residents at 3:00 a.m. on 12 March (1/6), 9:00 a.m. on 12 March (2/6), 3:00 p.m. on 12 March (3/6), 9:00 p.m. on 12 March (4/6), 12:00 a.m. on 13 March (5/6), and 12:00 a.m. on 15 March (6/6). The concentric circles denote 10, 20 (*thick line*), 30, 50, and 70 km-radii of the FDNPP from the inside.

Fig. 4 Trends of the numbers of individuals (the Adult group) staying within or outside the 20-km radius or moving during the period from 12:00 a.m. on 12 March to 12:00 a.m. on 16 March.

Fig. 5 Trends of the 317 individuals belonging to the Adult group by separating those staying within the 20 km radius (*upper*) and outside the 20-km radius (*lower*) for the period from 12:00 a.m. on 12 March to 12:00
 a.m. on 16 March.

Fig. 6 Comparison of the ¹³⁷Cs body content (kBq) between the G1 and G2 evacuation groups. The boxes cover the 25th percentile (not seen) to the 75th percentile of the distribution. The right panel expands the vertical axis of the left figures for easy to see the distributions in the small range. Same in Figs 7 and 8. The *lines* in the boxes are the 50th percentile (median) of the distribution (only seen in the Adult group and the 15 yr group (G2)). The *whiskers* cover the first quantile $-1.5 \times IQR$ (interquartile range) to the third quantile + 1.5 × IQR unless outliers (*crosses*) exist. The following figures with box-and-whisker plots use the same format.

Fig. 7 Comparisons of the ¹³⁷Cs intake (kBq) between the G1 and G2 evacuation groups.

ьз

Fig. 8 Comparisons of the CED (mSv) from ¹³⁴Cs and ¹³⁷Cs between the G1 and G2 evacuation groups.

⁴ **Fig. 9** Comparisons of the (a) ¹³⁷Cs body content (kBq), (b) ¹³⁷Cs intake (kBq) and (c) CED (mSv) from ¹³⁴Cs ⁶ and ¹³⁷Cs for the 15 yr and Adult group in the G1(T), G2(T), G1(M), and G2(M) groups. G1(T): the prompt ⁹ evacuees toward the northwest, i.e. Tsushima; G2(T): the late evacuees toward the northwest, G1(M): the ¹¹ prompt evacuees toward the north, i.e., Minami-soma; G2(M): the late evacuees toward the north.

APPENDIX 1

Trends of the numbers of individuals (5 yr, 10 yr, 15 yr, and All groups) staying within or moving outside $\frac{1}{2}$ the 20-km radius or moving during the period from 12:00 a.m. on 12 March to 12:00 a.m. on 16 March.

APPENDIX 2

⁹ Trends of individuals with positive ¹³⁷Cs detection in the 5 yr, 10 yr, 15 yr, and All groups and the detection ¹¹ $_{12}^{11}$ rate for the period from 12:00 a.m. on 12 March to 12:00 a.m. on 16 March.

	5 yr	10 yr	15 yr	Adult			
Number of subjects (Male and Female)	347	486	489	317			
With ¹³⁴ Cs detection	26 (7.5%) [*]	50 (10.3%)	108 (22.1%)	97 (30.6%)			
With ¹³⁷ Cs detection	40 (11.5%)	79 (16.3%)	145 (29.7%)	116 (36.6%)			
Males	179	239	241	91			
With ¹³⁴ Cs detection	16 (8.9%)	27 (11.3%)	70 (29.0%)	47 (51.6%)			
With ¹³⁷ Cs detection	21 (11.7%)	44 (18.4%)	89 (36.9%)	51 (56.0%)			
Females	168	247	248	226			
With ¹³⁴ Cs detection	10 (6.0%)	23 (9.3%)	38 (15.3%)	50 (22.1%)			
With ¹³⁷ Cs detection	19 (11.3%)	35 (14.2%)	56 (22.6%)	65 (28.8%)			

Table 1. The number of subjects with the positive detection of ¹³⁴Cs or ¹³⁷Cs for each age and gender group

* Figures in parentheses are the detection rate in percentage.

	5 yr	10 yr	15 yr	Adult	Total
Number of subjects	347	486	489	317	1,639
G1 evacuation group	279	387	363	222	1,251
With ¹³⁴ Cs detection	13 (4.7%)*	24 (6.2%)	52 (14.3%)	39 (17.6%)	128 (10.2%)
With ¹³⁷ Cs detection	19 (6.8%)	42 (10.9%)	80 (22.0%)	53 (23.9%)	194 (15.5%)
G2 evacuation group	68	99	126	95	388
With ¹³⁴ Cs detection	13 (19.1%)	26 (26.3%)	56 (44.4%)	58 (61.1%)	153 (39.4%)
With ¹³⁷ Cs detection	21 (30.9%)	37 (37.4%)	65 (51.6%)	63 (66.3%)	186 (47.9%)

Table 2. The numbers of subjects in the G1 and G2 evacuation groups

* Figures in parentheses are the detection rate in percentage.

	¹³⁴ Cs intake (Bq)			¹³⁷ Cs intake (Bq)				
	5 yr	10 yr	15 yr	Adult	5 yr	10 yr	15 yr	Adult
G1 evacuation group:								
Max.	2.2×10^{5}	1.0×10^5	4.5×10^4	1.4×10^4	3.1×10^5	1.1×10^5	4.7×10^4	1.7×10^4
95th percentile	N.D.	1.1×10^4	6.9×10^3	7.2×10^{3}	$4.7 imes 10^4$	1.4×10^4	7.9×10^3	8.6×10^{3}
90th percentile	N.D.	N.D.	5.2×10^{3}	$5.5 imes 10^3$	N.D.	9.8×10^{3}	5.9×10^{3}	5.4×10^{3}
75th percentile	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
50th percentile (median)	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
G2 evacuation group:								
Max.	1.3×10^5	6.7×10^4	2.6×10^4	$5.8 imes 10^4$	1.6×10^5	8.2×10^4	3.4×10^4	7.3×10^4
95th percentile	$9.7 imes 10^4$	3.2×10^4	1.5×10^4	1.7×10^4	1.1×10^5	3.3×10^4	$1.8 imes 10^4$	1.9×10^4
90th percentile	6.4×10^4	2.4×10^4	1.2×10^4	1.3×10^4	$8.7 imes 10^4$	2.7×10^4	1.4×10^4	1.6×10^4
75th percentile	N.D.	1.1×10^4	7.5×10^3	7.4×10^3	4.2×10^4	1.6×10^4	$8.9 imes 10^3$	8.7×10^3
50th percentile (median)	N.D.	N.D.	N.D.	3.9×10^3	N.D.	N.D.	4.0×10^{3}	4.8×10^{3}

Table 3. The Cs intake (Bq) between the G1 and G2 evacuation groups

N.D.: not detected in the WBC measurements

Age range: 5 yr: more than 2 years to 7 years, 10 yr: more than 7 years to 12 years, 15 yr: more than 12 years to 17 years, Adult: more than 17 years (ICRP1995)

	5 yr	10 yr	15 yr	Adult
G1 evacuation group:				
Max.	$2.3 imes 10^{0}$	9.7×10^{-1}	4.9×10^{-1}	1.7×10^{-1}
95th percentile	2.1×10^{-1}	1.0×10^{-1}	8.0×10^{-2}	8.2×10^{-2}
90th percentile	N.D.	3.8×10^{-2}	5.8×10^{-2}	6.2×10^{-2}
75th percentile	N.D.	N.D.	N.D.	N.D.
50th percentile (median)	N.D.	N.D.	N.D.	N.D.
G2 evacuation group:				
Max.	$1.2 imes 10^{0}$	6.6×10^{-1}	3.0×10^{-1}	7.2×10^{-1}
95th percentile	8.8×10^{-1}	2.9×10^{-1}	1.6×10^{-1}	2.0×10^{-1}
90th percentile	5.5×10^{-1}	2.1×10^{-1}	1.3×10^{-1}	1.6×10^{-1}
75th percentile	2.2×10^{-1}	1.0×10^{-1}	8.7×10^{-2}	8.8×10^{-2}
50th percentile (median)	N.D.	N.D.	2.0×10^{-2}	4.5×10^{-2}

Table 4. The CEDs (mSv) of the G1 and G2 evacuation groups

N.D.: not detected in the WBC measurements

Subjects : 15 yr and Adult groups	G1(T)	G2(T)	G1(M)	G2(M)
Number of persons	231	37	64	24
CED:				
Max	4.9×10^{-1}	6.1×10^{-1}	1.2×10^{-1}	7.2×10^{-1}
95th percentile	1.2×10^{-1}	2.9×10^{-1}	1.0×10^{-1}	3.0×10^{-1}
90th percentile	6.4×10^{-2}	1.5×10^{-1}	7.7×10^{-2}	2.8×10^{-1}
75th percentile	N.D.	8.9×10^{-2}	4.4×10^{-2}	1.4×10^{-1}
50th percentile (median)	N.D.	6.2×10^{-2}	N.D.	4.1×10^{-2}

Table 5. The CEDs (mSv) for the 15 yr and Adult subjects among the G1(T), G2(T), G1(M) and G2(M) groups

N.D.: not detected in the WBC measurements

Figure1

























Figure9 F1g. 9



Appendix1 Appendix 1



Appendix2 Appendix 2



