

See discussions, stats, and author profiles for this publication at: <https://www.researchgate.net/publication/259875399>

Simultaneous Sampling of Indoor and Outdoor Airborne Radioactivity after the Fukushima Daiichi Nuclear Power Plant Accident

ARTICLE in ENVIRONMENTAL SCIENCE & TECHNOLOGY · JANUARY 2014

Impact Factor: 5.33 · DOI: 10.1021/es404691m · Source: PubMed

CITATIONS

4

READS

69

7 AUTHORS, INCLUDING:



Sarata K. Sahoo

National Institute of Radiological Sciences

97 PUBLICATIONS 551 CITATIONS

SEE PROFILE



Mirosław Janik

National Institute of Radiological Sciences

63 PUBLICATIONS 403 CITATIONS

SEE PROFILE



Masahiro Hosoda

Hirosaki University

82 PUBLICATIONS 438 CITATIONS

SEE PROFILE



Shinji Tokonami

Hirosaki University

289 PUBLICATIONS 2,004 CITATIONS

SEE PROFILE

Simultaneous Sampling of Indoor and Outdoor Airborne Radioactivity after the Fukushima Daiichi Nuclear Power Plant Accident

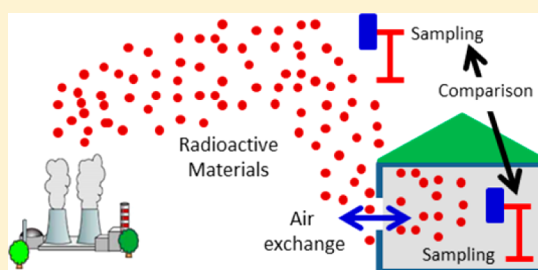
Tetsuo Ishikawa,^{*,†,‡} Atsuyuki Sorimachi,^{†,§} Hideki Arae,[‡] Sarata Kumar Sahoo,[‡] Mirosław Janik,[‡] Masahiro Hosoda,[§] and Shinji Tokonami[§]

[‡]National Institute of Radiological Sciences, 4-9-1 Anagawa, Inage-ku, Chiba 263-8555, Japan

[§]Hirosaki University, 66-1 Honcho, Hirosaki, Aomori 036-8564, Japan

Supporting Information

ABSTRACT: Several studies have estimated inhalation doses for the public because of the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident. Most of them were based on measurement of radioactivity in outdoor air and included the assumption that people stayed outdoors all day. Although this assumption gives a conservative estimate, it is not realistic. The “air decontamination factor” (ratio of indoor to outdoor air radionuclide concentrations) was estimated from simultaneous sampling of radioactivity in both inside and outside air of one building. The building was a workplace and located at the National Institute of Radiological Sciences (NIRS) in Chiba Prefecture, Japan. Aerosol-associated radioactive materials in air were collected onto filters, and the filters were analyzed by γ spectrometry at NIRS. The filter sampling was started on March 15, 2011 and was continued for more than 1 year. Several radionuclides, such as ^{131}I , ^{134}Cs , and ^{137}Cs were found by measuring the filters with a germanium detector. The air decontamination factor was around 0.64 for particulate ^{131}I and 0.58 for ^{137}Cs . These values could give implications for the ratio of indoor to outdoor radionuclide concentrations after the FDNPP accident for a similar type of building.



INTRODUCTION

After the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, a lot of radiation and radioactivity monitoring has been conducted. Airborne radionuclide concentrations have been measured at many places in Japan;¹ however, most of these measurements were performed outdoors. Some studies have estimated inhalation doses, but these were based on the assumption that people stay outside all day.^{2–4} Although such an assumption is reasonable for estimating “projected dose”,⁵ which means the dose without taking into account protective actions, it does not reflect reality. According to a common time budget, people spend more time indoors than outdoors. Then, radionuclide concentrations in indoor air are necessary for estimating more realistic doses. However, such data have never been reported in the case of the FDNPP accident.

When considering protection against atmospheric releases from nuclear power facilities in an accident, the ratio of indoor to outdoor airborne radionuclide concentrations is an important parameter. This is called the “exposure factor”⁶ or “air decontamination factor”.⁷ The ratio depends upon several factors, such as air exchange rate between indoor and outdoor environments, deposition rate of radionuclides onto walls, floors, furniture, etc. inside a building, and the nature of the radioactive materials. According to a model calculation, a typical commercial building may have an exposure factor somewhere in the range of 0.4–0.9 for $1\ \mu\text{m}$ particles, 0.05–0.4

for $5\ \mu\text{m}$ particles, and 0.02–0.2 for $10\ \mu\text{m}$ particles.⁶ The EURANOS project estimated that the air decontamination factor (hereafter, DF) over some period can be achieved as $1/2$ for particles in the $0.5\ \mu\text{m}$ range, $1/8$ for particles in the $4\ \mu\text{m}$ range, and $1/12$ for elemental iodine gas and there was no effect on non-reactive gases, such as CH_3I .⁷

Jensen⁸ reported that the inhalation dose reduction factor was in the range of 0.23–0.45 for a Finnish study and 0.40–0.47 for a Norwegian study. A DF of 0.25 was recommended by the Nuclear Safety Commission (NSC) of Japan soon after the FDNPP accident.⁹ However, this value was not based on any actual measurements.

Sampling of airborne radioactivity has been conducted simultaneously both inside and outside a workplace (a five-story building) at the National Institute of Radiological Sciences (NIRS) in Chiba Prefecture, Japan, located about 220 km south–southwest of the FDNPP. The filter sampling was started on March 15, 2011 and lasted for more than 1 year. The filter was changed to a new one almost twice a day until the end of March. After April, the sampling period was extended accordingly, because of a decrease of the activity

Received: October 21, 2013

Revised: January 21, 2014

Accepted: January 22, 2014

Table 1. Detected Radionuclides and Their Activity Concentrations for Typical Samples Taken in Outdoors

sampling period		radionuclide concentration (Bq/m ³)						
		¹³² Te	¹³¹ I	¹³⁴ Cs	¹³⁷ Cs	^{129m} Te	⁹⁵ Nb	¹³⁶ Cs
start	stop	(228 keV)	(364 keV)	(605 keV)	(662 keV)	(696 keV)	(766 keV)	(819 keV)
(1) NIRS Outdoor								
2011/3/15, 18:00	2011/3/16, 9:00	1.43 ± 0.02	4.65 ± 0.02	0.207 ± 0.005	0.214 ± 0.005	0.23 ± 0.06	ND	0.039 ± 0.003
2011/3/16, 9:00	2011/3/16, 18:00	7.47 ± 0.06	15.96 ± 0.06	0.91 ± 0.01	0.92 ± 0.01	1.1 ± 0.2	ND	0.151 ± 0.009
2011/3/20, 18:01	2011/3/21, 9:00	9.6 ± 0.2	13.41 ± 0.06	10.11 ± 0.03	10.24 ± 0.03	3.8 ± 0.2	0.020 ± 0.006	1.21 ± 0.02
2011/3/21, 9:02	2011/3/21, 17:58	4.4 ± 0.2	3.93 ± 0.04	4.36 ± 0.03	4.48 ± 0.03	2.1 ± 0.2	ND	0.52 ± 0.02
2011/3/21, 18:00	2011/3/22, 8:57	0.41 ± 0.05	0.87 ± 0.01	0.122 ± 0.003	0.123 ± 0.003	0.31 ± 0.05	ND	0.013 ± 0.003
2011/3/22, 8:58	2011/3/22, 17:58	0.7 ± 0.1	14.7 ± 0.1	0.214 ± 0.006	0.210 ± 0.005	0.48 ± 0.09	0.013 ± 0.004	ND
2011/3/22, 17:59	2011/3/23, 9:00	3.6 ± 0.1	16.1 ± 0.1	0.461 ± 0.007	0.448 ± 0.006	2.3 ± 0.1	ND	0.034 ± 0.008
(2) JCAC Outdoor								
2011/3/15, 6:25	2011/3/16, 10:00	5.7 ± 0.01	33 ± 0.04	0.70 ± 0.006	0.87 ± 0.006			0.13 ± 0.004
2011/3/16, 10:05	2011/3/17, 10:00	0.80 ± 0.005	7.4 ± 0.02	0.10 ± 0.0038	0.12 ± 0.004			0.016 ± 0.0023
2011/3/20, 10:05	2011/3/21, 11:40	6.6 ± 0.01	33 ± 0.05	6.1 ± 0.03	7.5 ± 0.03			0.89 ± 0.013
2011/3/21, 11:45	2011/3/22, 9:50	0.57 ± 0.004	3.5 ± 0.02	0.38 ± 0.006	0.46 ± 0.006			0.053 ± 0.0027
2011/3/22, 9:55	2011/3/23, 9:25	2.6 ± 0.01	47 ± 0.06	0.29 ± 0.007	0.33 ± 0.006			0.036 ± 0.0037

concentration. On the basis of these measurements, changes in indoor and outdoor radionuclide concentrations with time were estimated. Also, the DF and its changes with time were estimated for ¹³¹I, ¹³⁴Cs, and ¹³⁷Cs. The present study is the first and probably the sole report on the ratio of indoor to outdoor radionuclide concentrations after the FDNPP accident. While the DFs reported in the present study are not typical values, they could be useful for estimating indoor radionuclide concentrations for similar types of buildings from outdoor monitoring data obtained after the FDNPP accident.

MATERIALS AND METHODS

Two sets of sampling equipment were assembled soon after the FDNPP accident started. Each consisted of a 47 mm filter holder and a pump connected with the holder through a tube. The first set was placed on the roof of the five-story building at NIRS. This installation site was about 20 m from the ground. The filter holder was set on a tripod around 1 m above the roof. The second set was placed in a fifth-floor room of the same building. The filter holder was fixed on the top of a shelf (about 2 m above the floor). Windows of the room were always closed. The door of the room was sometimes opened during working hours on weekdays, but it was closed at night and on weekends. The room was basically used for measuring natural radioactivity for environmental samples. Because of this activity, it was sometimes occupied by a few people during the working hours.

Nitrocellulose filters were mainly used, although Whatman glass microfiber filters (grade GF/F) were used for the first two samplings. The sampling period began at 18:00 on March 15 and lasted for more than 1 year. Until the end of March, the sampling was performed twice a day: around 9:00 to 18:00 and 18:00 to 9:00 of the next day. After April, the sampling time was extended to a few days or 1 week because of a decrease of radionuclide concentrations. A flow rate of 20 L/min was used

for the samples taken until April 28, except for the first two samples (25 L/min). After that, the flow rate was decreased to 10 L/min to prevent overheating of the pump because of the extended sampling time. Consequently, the total sampling volume for each filter was around 10–20 m³ until the beginning of April, around 60 m³ from the middle of April to the beginning of May, and around 100 m³ after that. The differences in the sampling flow rate may affect collection efficiency for particles. However, the data (concentrations and DF) for which the same flow rate was used (20 L/min, from March 16 to April 28) can be compared on the same sampling conditions.

The filter samples were measured with a high-purity germanium detector (HPGe) at NIRS. The calibration for the 47 mm filter source was performed by an intercomparison to the HPGe of Hiroaki University.¹⁰ The calibration was performed in the following way. The HPGe at Hiroaki University was calibrated using standard sources with multiple radionuclides (Japan Radioisotope Association). The amounts of ¹³⁴Cs (604 and 796 keV) and ¹³⁷Cs (662 keV) for a few 47 mm filter samples were determined with HPGe at Hiroaki University. When the same samples were measured with HPGe at NIRS, calibration factors for 604, 662, and 796 keV for the NIRS HPGe were estimated.

When these three points were interpolated, a calibration factor for a peak for ¹³¹I (636 keV) was estimated. Then, using the intensity ratio of γ rays, calibration factors for other peaks for ¹³¹I (81, 284, 364, and 723 keV) were estimated from filter samples taken in March. From these points, a calibration curve against γ -ray energy was established. Correction for the sum peak of ¹³⁴Cs was also performed.

The counting time ranged from 10 000 to 30 000 s, depending upon the predicted level of activity. Small amounts of ¹³⁴Cs and ¹³⁷Cs were detected from the background

spectrum because of dust contamination in the room where the HPGe was located.

A detection limit of ^{134}Cs and ^{137}Cs for each filter sample was calculated using the following equation based on a method by Currie:¹¹

$$N_D = \frac{k^2}{t_s} + 2k \sqrt{\left(\frac{1}{t_s} + \frac{1}{t_b}\right) N_B} \quad (1)$$

where N_D is the net counting rate corresponding to the detection limit, k is a constant (1.645), t_s is the counting time for a sample, t_b is the counting time for the background, and N_B is the background counting rate.

Because of the tight use schedule of the HPGe, background counts were measured after a series of sample measurements. The first background measurement with a counting time of 80 000 s was performed in the middle of April, and the second background measurement was performed in the middle of May (300 000 s). The first background measurement was used for the background for samples measured in March and April, and the second background measurement was used for that in May and June. Even if the same background counting rate was applied to eq 1 for a series of samples, the detection limit in the unit of Bq/m^3 could be different for each, depending upon the sampled air volume and counting time.

As a result, the detection limit for ^{134}Cs and ^{137}Cs was estimated to be around $2\text{--}4 \times 10^{-3} \text{ Bq}/\text{m}^3$ for the samples measured in March 2011, and that decreased to around $3 \times 10^{-4} \text{ Bq}/\text{m}^3$ for the samples in June 2011. Because ^{131}I was not detected from the background spectrum, the net counts for filter samples without subtracting the background were used for estimation of radioactivity.

The activity concentration at the time sampling was stopped was estimated using the decay-corrected net count rates, calibration coefficients, and total sampling volume. From these data, (1) outdoor concentrations of ^{131}I , ^{134}Cs , and ^{137}Cs and their changes with time and (2) changes in the ratio of $^{134}\text{Cs}/^{137}\text{Cs}$ with time were discussed for comparison of the present results to other studies. Then, (3) DF for ^{131}I , ^{134}Cs , and ^{137}Cs and (4) their changes with time were analyzed.

RESULTS AND DISCUSSION

Detected Radionuclides. For the first sample taken from March 15, 18:00 to March 16, 9:00, the following radionuclides were detected: ^{132}Te , ^{131}I , ^{129}Te , ^{134}Cs , ^{137}Cs , ^{132}I , ^{129m}Te , and ^{136}Cs . In addition, ^{95}Nb and ^{140}La were detected for the sample taken from March 20, 18:00 to March 21, 9:00. ^{132}Te (half-life of 78.2 h), ^{129}Te (69.6 min), ^{132}I (2.30 h), ^{129m}Te (33.6 days), ^{136}Cs (13.2 days), ^{95}Nb (35.0 days), and ^{140}La (40.3 h) were detected for (some of) the samples taken until March 22 but not detected after that because of their short half-lives. ^{131}I was detected for the samples taken until March 31, while ^{134}Cs and ^{137}Cs were detected for the samples taken until the end of June, 2011. Table 1 shows the detected radionuclides and their activity concentration (particulate type only) for typical filter samples. Japan Chemical Analysis Center (JCAC), which is located near NIRS (around 3 km away), also measured airborne radionuclides.² Their results are provided in Table 1 for comparison.

A brief description of the measurements by JCAC is as follows:² sampling of airborne particles and gaseous chemical species was carried out at a height of 1.5 m above ground level

at JCAC. Airborne particles and gaseous iodine were collected continuously using a cellulose glass-fiber filter (Toyo HE 40T) in combination with an activated charcoal cartridge containing 10% triethylenediamine (TEDA) (Toyo CHC-50). The filter and cartridge were changed every day (around 9:30 with a several minutes break for change) and measured daily starting on March 14, 2011. Air sampling was performed using a low-volume air sampler, and the air flow rate was regulated at around 90 L/min. Radioactivity of the filter and cartridge was directly measured together by γ -ray spectrometry with a Ge detector.

As for measurements at NIRS, because of the urgency of the situation following the nuclear accident, the Ge detector was in heavy use with many samples of other researchers to be measured; therefore, more than 1 week passed from the end of sampling to the start of measurement with the Ge detector. Thus, the detected radionuclides with short half-life (^{129}Te , ^{132}I , and ^{140}La) were considered to be produced from their parent nuclides (^{129m}Te , ^{132}Te , and ^{140}Ba , respectively) that were collected on the filters. For this reason, these short-lived radionuclides are not shown in Table 1.

Outdoor Concentrations of ^{131}I , ^{134}Cs , and ^{137}Cs . Changes in outdoor concentrations of ^{131}I , ^{134}Cs , and ^{137}Cs until the end of March 2011 are shown in Figure 1. The data

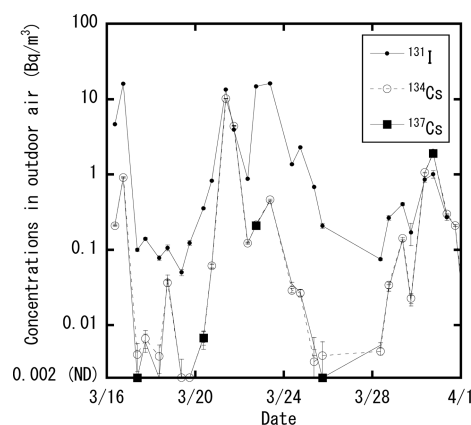


Figure 1. Changes of concentrations of ^{131}I , ^{134}Cs , and ^{137}Cs in outdoor air in March 2011.

below the detection limit ($2\text{--}4 \times 10^{-3} \text{ Bq}/\text{m}^3$, depending upon the sampled volume) are plotted on the line of $0.002 \text{ Bq}/\text{m}^3$ (ND). The pattern of changes was in accordance with the model calculation of atmospheric dispersion of radioactive materials.^{12,13} That is, the peaks of outdoor concentrations (March 15–16, 20–21, 22–23, and 30–31 in Figure 1) seemed to correspond to those of the dry deposition pattern. Precipitation and wind speed data were obtained from the weather-monitoring station nearest NIRS, and their relationship with outdoor concentrations was considered; however, no relationship was found.

According to the measurements by JCAC,² the highest concentration of ^{137}Cs ($7.5 \text{ Bq}/\text{m}^3$) was observed on March 20–21. This was consistent with the highest value ($10.2 \text{ Bq}/\text{m}^3$, sampled outdoors from March 20, 18:00 to March 21, 9:00) in the present study (Table 1). Also, a similar ^{137}Cs activity was observed for the sample taken on March 16: around $0.87 \text{ Bq}/\text{m}^3$ at JCAC² and $0.92 \text{ Bq}/\text{m}^3$ at NIRS. It is assumed that the radionuclides associated with aerosols (without gaseous radionuclides) were collected by the filter sampling in the

present study. On the other hand, JCAC measured both gaseous and aerosol-associated radioactivities and estimated the ratio of gaseous cesium to total cesium to be 0.016 on March 15–16 and 0.025 on March 20–21. Then, it was reasonable that a similar level of ^{137}Cs was observed at NIRS as well. The concentration level of ^{134}Cs was almost the same as that of ^{137}Cs .

JCAC reported that (1) the estimated ratios of gaseous iodine to total iodine were 0.71 on March 15–16, 0.52 on March 20–21, and 0.68 on March 22–23 and (2) the highest concentration of ^{131}I (47 Bq/m^3) was observed on March 22–23. Considering these values, the aerosol-associated ^{131}I was estimated to be 15 Bq/m^3 on March 22–23. This was in good agreement with the estimated concentration (16.1 Bq/m^3) for the sample taken from March 22, 18:00 to March 23, 9:00 in the present study (Table 1).

$^{134}\text{Cs}/^{137}\text{Cs}$ Ratio. Changes in the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio can be used as time markers. ^{134}Cs and ^{137}Cs were detected for the filter sampling until June 2011. ^{134}Cs and ^{137}Cs were not detected for most of the samples taken after June (the detection limit of around $3 \times 10^{-4} \text{ Bq}/\text{m}^3$). The ratio of $^{134}\text{Cs}/^{137}\text{Cs}$ activity outdoors and its changes with time are shown in Figure 2. To decrease the fluctuation, weekly averages of the ratio were

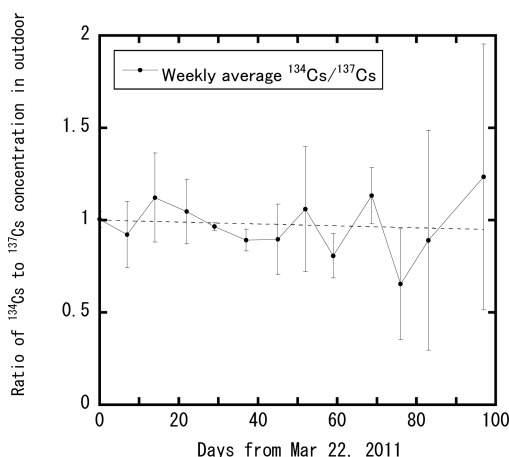


Figure 2. Ratio (weekly average) of $^{134}\text{Cs}/^{137}\text{Cs}$ activity outdoors and its changes with time.

plotted. Theoretically,¹⁴ the logarithmic slope is $-0.000860 \text{ day}^{-1}$, which corresponds to a half-life of 2.21 years. The present results gave a slope of $-0.000529 \text{ day}^{-1}$, which corresponded to a half-life of 3.59 years. The difference probably resulted from the lowered accuracy of the activity concentration associated with the decrease of the outdoor concentration after April.

DFs for ^{131}I , ^{134}Cs , and ^{137}Cs . Changes of ^{131}I concentrations in outdoor and indoor air are shown in Figure 3a, and those of ^{137}Cs are shown in Figure 3b. The data below the detection limit ($2\text{--}4 \times 10^{-3} \text{ Bq}/\text{m}^3$) were plotted on the line of $0.002 \text{ Bq}/\text{m}^3$ (ND). The indoor concentration seemed to change following changes in the outdoor concentration. Also, the indoor concentration was similar to or less than the outdoor concentration in most cases. These findings were reasonable. However, there were some data points where the indoor concentration was higher. Such a case was clearly seen for the data on March 21 from the evening (just after the second peak in panels a and b of Figure 3). The outdoor concentration of ^{131}I decreased from $13.4 \text{ Bq}/\text{m}^3$ (March 21,

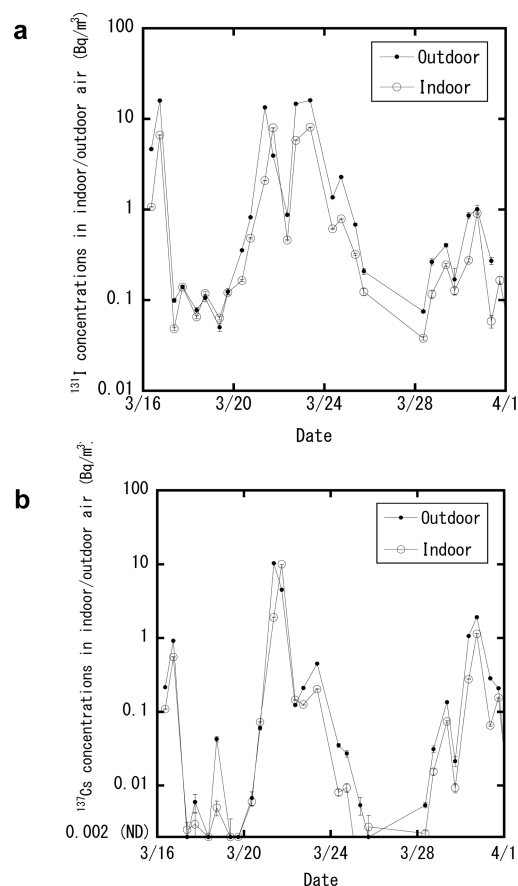


Figure 3. Changes of (a) ^{131}I and (b) ^{137}Cs concentrations in outdoor and indoor air.

9:00 sampling stop) to $3.9 \text{ Bq}/\text{m}^3$ (March 21, 18:00 sampling stop). As shown in Figure 3a, the indoor ^{131}I concentration responded to the decrease with time delay because of the air exchange rate between outdoors and indoors. Similar changes could also be seen for ^{137}Cs (Figure 3b).

Figure 4a shows changes in the DF for ^{131}I and ^{137}Cs until the end of March 2011. After that, ^{131}I was not detected. In the case of ^{137}Cs data below the detection limit (either indoors or outdoors), DF was plotted as 0. The ratio for ^{134}Cs was almost the same as that for ^{137}Cs . The ratio ranged from 0.16 to 2.0 (arithmetic mean of 0.64) for ^{131}I and from 0.08 to 2.2 (arithmetic mean of 0.58) for ^{137}Cs . The differences in sampling flow rate may affect collection efficiency for particles. Even if the first two samples taken with a different flow rate (25 L/min) were excluded, the results were similar; the arithmetic mean of DF for ^{131}I was 0.67, and that for ^{137}Cs was 0.59.

In some cases, the ratio exceeded 1.0, which was due to the decrease of the indoor concentration with a time delay, following the decrease of the outdoor concentration. The time delay depends upon the air exchange rate between outdoor and indoor. When the outdoor concentration drastically decreased, the indoor concentration was temporally higher than the outdoor concentration, as mentioned before. The delay would be smaller with the increasing air exchange rate. Figure 4b shows changes in the DF for ^{137}Cs for a longer period (the sampling flow rate was not the same for the whole period, as mentioned before). After the beginning of April, the DF seemed to be more stable (range of 0.22–1.1 and arithmetic mean of 0.52 for the data in April).

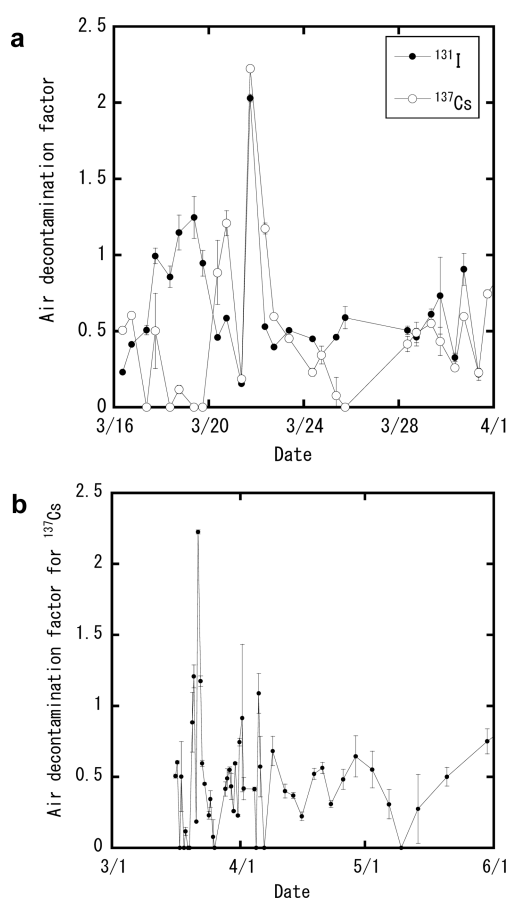


Figure 4. Changes of the ratio of indoor to outdoor air concentrations (DF) for (a) ^{131}I and ^{137}Cs and (b) ^{137}Cs for a longer period.

The DFs obtained in the present study are compared to other literature values in Table 2. As mentioned above, the ratio of gaseous cesium to total cesium was less than 0.1. Thus, the DF for cesium estimated above (arithmetic mean of 0.58) would be similar to the DF for total cesium. For iodine, however, the DF would differ between aerosols and gaseous radionuclides. EURANOS estimated a DF of $1/12$ for elemental iodine gas but a DF of almost unity (no effect of decontamination) for non-reactive gases, such as CH_3I .⁷

Only a few data have been reported for chemical forms of iodine released by the FDNPP accident. In the case of the Chernobyl accident, however, the proportions of ^{131}I species identified in Japan were 19% particulate iodine, 5% I_2 , 6%

hypoiodous acid (HOI), and 70% organic iodides.¹⁵ Assuming that most of the iodine is in the form of organic (non-reactive) gases in the present case, there would be almost no effect of decontamination for gaseous iodine. Considering that (1) DF for particulates was 0.64 and (2) the ratio of gaseous iodine to total iodine was around 0.6, the DF for total iodine could be around 0.85. This was much higher than the recommended value (0.25) set by the NSC of Japan.⁹

The activity median aerodynamic diameter of ^{137}Cs was estimated to be $0.53\ \mu\text{m}$ for one April 28–May 12 sample and $0.64\ \mu\text{m}$ for one May 12–26 sample, both taken at Tsukuba City.¹⁶ As shown in Table 2, the DF increased with a decreasing particle size [see Lawrence Berkeley National Laboratory (LBNL) and EURANOS entries]. The LBNL suggested a DF value somewhere in the range of 0.4–0.9 for $1\ \mu\text{m}$ particles,⁶ and EURANOS proposed a DF of 0.5 for $0.5\ \mu\text{m}$ particles.⁷ The present results were generally consistent with these values.

The DF discussed above is the ratio of outdoor/indoor concentrations at the same time. From the viewpoint of dosimetry, the use of time-integrated concentrations might be useful, because the total intake by inhalation (which is proportional to the dose) can be estimated by multiplying the breathing rate by the time-integrated concentration. That is, the ratio of the area under the outdoor concentration line (in the graph of Figure 3a) to that under the indoor concentration line (in the graph of Figure 3b) could be another index [hereafter called DF (integrated)].

From panels a and b of Figure 3, the DF (integrated) was estimated at 0.72 for ^{137}Cs . The DF (integrated) for ^{131}I was calculated to be 0.48 for particulates. The DF (integrated) was calculated to be 0.79, considering the gaseous ^{131}I (DF = 1); this was almost the same as the DF (integrated) for ^{137}Cs mentioned above (0.72).

Although the DF is important for estimating realistic doses to the public because of the FDNPP accident, such data were lacking. The DF can differ, depending upon the air exchange rate, resuspension rate indoor, etc.⁶ Thus, the DF values estimated in the present study may not be typical values. The majority of Japanese dwellings are wooden; for example, the number ratio of wooden dwellings to the total dwellings was around 60% according to a 2008 government survey.¹⁷ Some dwellings in Japan are made of concrete. Also, similar building types (concrete type) can often be seen in workplaces.

In this respect, the present data will make a significant addition to the data on indoor and outdoor measurements especially for concrete-type buildings with similar human activity, such as workplaces (occupied on weekdays and closed

Table 2. Comparison of the DF in Different Studies

reference	air decontamination factor (DF)		
	^{137}Cs	gaseous ^{131}I	particulate ^{131}I
this study (range and average, data in March)	0.08–2.2 (0.58)	no data	0.16–2.0 (0.64)
this study (range and average, data in April)	0.22–1.1 (0.52)	no data	no data
LBNL ⁶	0.4–0.9 ($1\ \mu\text{m}$) 0.05–0.4 ($5\ \mu\text{m}$) 0.02–0.2 ($10\ \mu\text{m}$)		0.4–0.9 ($1\ \mu\text{m}$) 0.05–0.4 ($5\ \mu\text{m}$) 0.02–0.2 ($10\ \mu\text{m}$)
EURANOS ⁷	0.5 ($0.5\ \mu\text{m}$) 0.125 ($4\ \mu\text{m}$)	1 (non-reactive gas) $1/12$ (elemental iodine)	0.5 ($0.5\ \mu\text{m}$) 0.125 ($4\ \mu\text{m}$)
Riso ⁸	0.23–0.45 (Finland) 0.40–0.47 (Norway)		0.23–0.45 (Finland) 0.40–0.47 (Norway)
NSC ⁹	0.25	0.25	0.25

on the weekend), while they are not representative values that can be used for a general recommendation.

■ ASSOCIATED CONTENT

Supporting Information

Sampling site location and its relationship to the FDNPP site (Figure S1). This material is available free of charge via the Internet at <http://pubs.acs.org>.

■ AUTHOR INFORMATION

Corresponding Author

*Telephone: +81-24-547-1932. Fax: 024-547-1244. E-mail: isikawat@fmu.ac.jp.

Present Address

[†]Tetsuo Ishikawa and Atsuyuki Sorimachi: Fukushima Medical University, Hikariga-oka 1, Fukushima 960-1295, Japan.

Notes

The authors declare no competing financial interest.

■ REFERENCES

- (1) Priest, N. D. Radiation doses received by adult Japanese populations living outside Fukushima Prefecture during March 2011, following the Fukushima 1 nuclear power plant failures. *J. Environ. Radioact.* **2012**, *114*, 162–170.
- (2) Amano, H.; Akiyama, M.; Chumlei, B.; Kawamura, T.; Kishimoto, T.; Kuroda, T.; Muroi, T.; Odaira, T.; Ohta, Y.; Takeda, K.; Watanabe, Y.; Morimoto, T. Radiation measurements in the Chiba Metropolitan Area and radiological aspects of fallout from the Fukushima Dai-ichi Nuclear Power Plants accident. *J. Environ. Radioact.* **2012**, *111*, 42–52.
- (3) Nagakawa, Y.; Suzuki, T.; Kinjo, Y.; Sakurai, N.; Sotodate, T.; Ise, H. Measurement of radioactivity in airborne dust and estimation of public dose in Tokyo after the Fukushima Daiichi Nuclear Power Plant accident. *Proceedings of the International Symposium on Environmental Monitoring and Dose Estimation of Residents after Accident of TEPCO's Fukushima Daiichi Nuclear Power Station*; Shiran Hall, Kyoto, Japan, Dec 14, 2012; http://www.rrl.kyoto-u.ac.jp/anzen_kiban/outcome/.
- (4) Takeyasu, M.; Nakano, M.; Fujita, H.; Nakada, A.; Watanabe, H.; Sumiya, S.; Furuta, S. Results of environmental radiation monitoring at the Nuclear Fuel Cycle Engineering Laboratories, JAEA, following the Fukushima Daiichi Nuclear Power Plant accident. *J. Nucl. Sci. Technol.* **2012**, *49* (3), 281–286.
- (5) International Commission on Radiation Protection. *Principles for Intervention for Protection of the Public in a Radiological Emergency*; Pergamon Press: Oxford, U.K., 1993; International Commission on Radiological Protection (ICRP) Publication 63.
- (6) *Indoor Exposures to Radiation in the Case of an Outdoor Release*; Price, P. N., Jayaraman, B., Eds.; Lawrence Berkeley National Laboratory (LBNL): Berkeley, CA, 2006; LBNL-60662.
- (7) EURANOS. *Generic Handbook for Assisting in the Management of Contaminated Inhabited Areas in Europe Following a Radiological Emergency*; EURANOS: Brussels, Belgium, 2010; <http://www.euranos.fzk.de/index.php?action=euranos&title=products>.
- (8) Jensen, P. H. *Atmospheric Dispersion and Environmental Consequences*; Risø National Laboratory: Roskilde, Denmark, 1992; RISØ-M-2849.
- (9) Nuclear Safety Commission (NSC) of Japan. *Dose Estimation Based on Emergency Monitoring Data (Material of the Meeting on March 25, 2011)*; NSC of Japan: Tokyo, Japan, 2011; [http://home.hiroshima-u.ac.jp/er/EV_TKJ\(2\)_HS_02.html](http://home.hiroshima-u.ac.jp/er/EV_TKJ(2)_HS_02.html).
- (10) Hosoda, M.; Tokonami, S.; Tazoe, H.; Sorimachi, A.; Monzen, S.; Osanai, M.; Akata, N.; Kakiuchi, H.; Omori, Y.; Ishikawa, T.; Sahoo, S. K.; Kovacs, T.; Yamada, M.; Nakata, A.; Yoshida, M.; Yoshino, H.; Mariya, Y.; Kashiwakura, I. Activity concentrations of environmental samples collected in Fukushima Prefecture immediately after the Fukushima nuclear accident. *Sci. Rep.* **2013**, *3*, 2283.
- (11) Currie, L. A. Limits for qualitative detection and quantitative determination: Application to radiochemistry. *Anal. Chem.* **1968**, *40*, 586–593.
- (12) Morino, Y.; Ohara, T.; Watanabe, M.; Hayashi, S.; Nishizawa, M. Episode analysis of deposition of radiocesium from the Fukushima Daiichi Nuclear Power Plant accident. *Environ. Sci. Technol.* **2013**, *47*, 2314–2322.
- (13) Terada, H.; Katata, G.; Chino, M.; Nagai, H. Atmospheric discharge and dispersion of radionuclides during the Fukushima Dai-ichi Nuclear Power Plant accident. Part II: Verification of the source term and analysis of regional-scale atmospheric dispersion. *J. Environ. Radioact.* **2012**, *112*, 141–154.
- (14) Kanai, Y. Monitoring of aerosols in Tsukuba after Fukushima Nuclear Power Plant incident in 2011. *J. Environ. Radioact.* **2011**, *111*, 33–37.
- (15) Noguchi, H.; Murata, M. Physicochemical speciation of airborne ¹³¹I in Japan from Chernobyl. *J. Environ. Radioact.* **1988**, *7*, 131–144.
- (16) Kaneyasu, N.; Ohashi, H.; Suzuki, F.; Okuda, T.; Ikemori, F. Sulfate aerosol as a potential transport medium of radiocesium from the Fukushima nuclear accident. *Environ. Sci. Technol.* **2012**, *46*, 5720–5726.
- (17) Statistics Bureau, Ministry of Internal Affairs and Communications. *Housing and Land Survey, Table 12, Official Statistics of Japan*; Statistics Bureau, Ministry of Internal Affairs and Communications: Tokyo, Japan, 2008; <http://www.e-stat.go.jp/SG1/estat/ListE.do?bid=000001029530&cycode=0>.