

# Effect of the Fukushima nuclear power plant accident on radioiodine ( $^{131}\text{I}$ ) content in human breast milk

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## Abstract

**Background:** Environmental pollution with radioiodine (iodine-131,  $^{131}\text{I}$ ) occurred after an accident at the Fukushima nuclear power plant (FNP) on March 11, 2011, in Japan. Whether environmental pollution with  $^{131}\text{I}$  can contaminate human breast milk has not been documented.

**Methods:** The  $^{131}\text{I}$  content was determined in 126 breast milk samples from 119 volunteer lactating women residing within 250 km of the FNP, between April 24 and May 31, 2011. The degree of environmental pollution was determined based on the data released by the Japanese government.

**Results:** An  $^{131}\text{I}$  content of 210 Bq/kg in the tap water in Tokyo, which is located 230 km south of the FNP, on March 22 and of 3500 Bq/kg in spinach sampled in a city located 140 km southwest of the FNP on March 19 decreased over time to <21 Bq/kg on March 27 and 12 Bq/kg on April 26, respectively. Seven of the 23 women who were tested in April secreted a detectable level of  $^{131}\text{I}$  in their breast milk. The concentrations of  $^{131}\text{I}$  in the breast milk of the seven women were 2.3 Bq/kg (on April 24), and 2.2, 2.3, 2.3, 3.0, 3.5 and 8.0 (on April 25); the concentrations of  $^{131}\text{I}$  in the tap water available for these seven women at the same time were estimated to be <1.3 Bq/kg. None of the remaining 96 women tested in May exhibited a detectable concentration of  $^{131}\text{I}$  in their breast milk samples.

**Conclusions:** The contamination of breast milk with  $^{131}\text{I}$  can occur even when only mild environmental  $^{131}\text{I}$  pollution is present.

**Key words:** human breast milk, iodine-131, nuclear power plant accident, radioiodine.

## Introduction

On March 11, 2011, an earthquake (magnitude, 9.0) triggered a large tsunami more than 16.0 m high, which then hit the Fukushima nuclear power plant

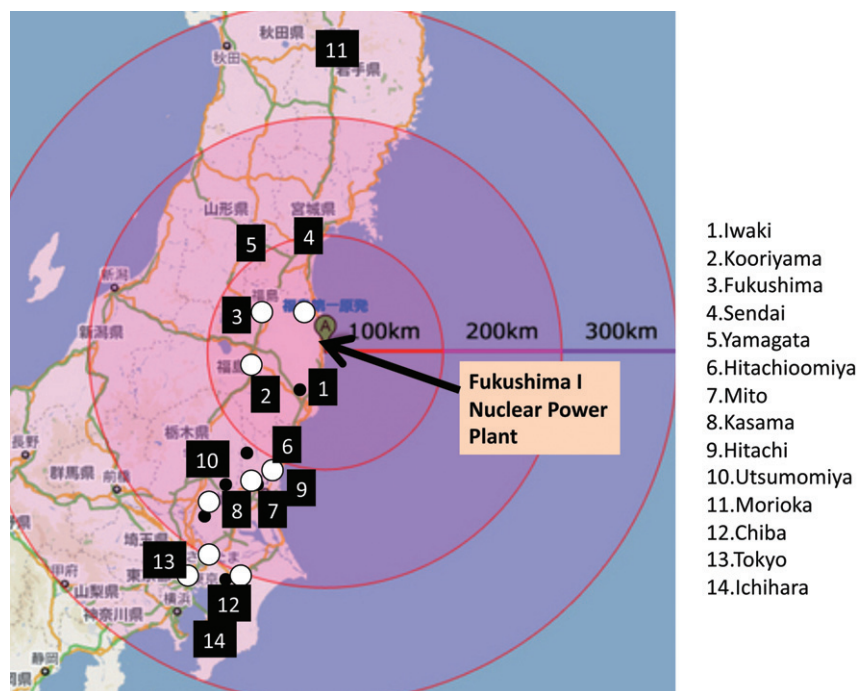
(FNP) in Japan (Fig. 1). Subsequently, the FNP explosively dispersed a massive radioactive plume on the morning of March 15, 2011. The radioactive cloud was carried by the wind, inducing widespread pollution with  $^{131}\text{I}$  and other radioactive species.

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**Figure 1** Location of the Fukushima nuclear power plant in Japan. The closed and open circles indicate the cities where women whose breast milk was or was not contaminated with  $^{131}\text{I}$  were living, respectively.



Stable iodine ingested during the consumption of daily meals is secreted in breast milk.<sup>1,2</sup> Radioactive iodine (iodine-131,  $^{131}\text{I}$ ) administered orally or intravenously for medical purposes has also been shown to accumulate in the thyroid and breast tissues and to be excreted in breast milk.<sup>3-5</sup> Beginning approximately four years after the Chernobyl reactor accident of April 1986, a sharp increase in the incidence of thyroid cancer among children and adolescents in areas covered by the radioactive plume was observed, with the risk greatest in those youngest at exposure.<sup>6,7</sup> However, whether human breast milk was actually contaminated with  $^{131}\text{I}$  after the Chernobyl reactor accident was uncertain, partly because of the short half-life of  $^{131}\text{I}$  (8 days). Nevertheless, human breast milk was regarded as a major possible contributor to the doses of  $^{131}\text{I}$  received by nursing infants in the vicinity of the Chernobyl reactor accident. Thus, breast milk contamination with  $^{131}\text{I}$  is a major concern associated with environmental  $^{131}\text{I}$  pollution. Accordingly, we investigated the  $^{131}\text{I}$  content in breast milk in collaboration with and supported by the Japanese Ministry of Health, Labour, and Welfare (JMHLW).

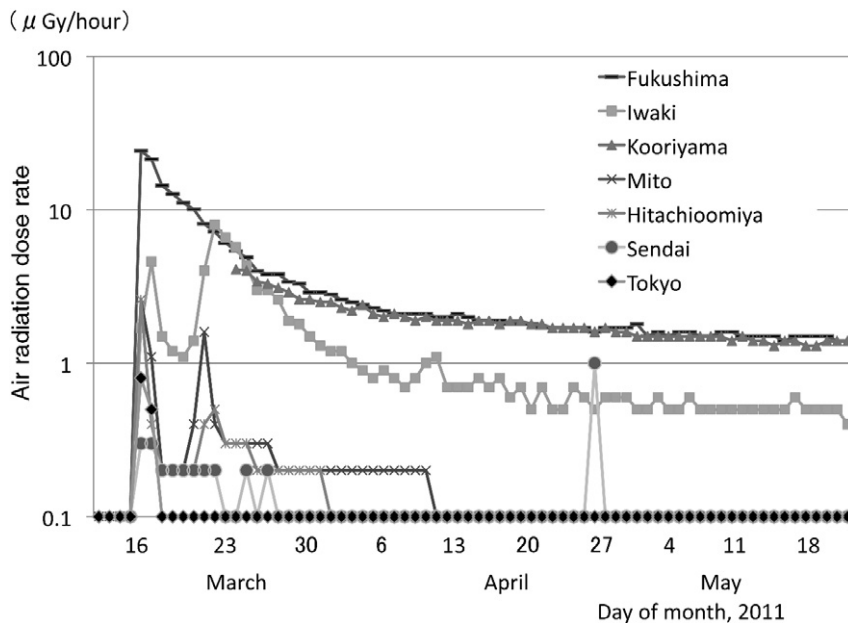
## Materials and Methods

This study was approved by the institutional review board of the Japan National Institute of Public Health.

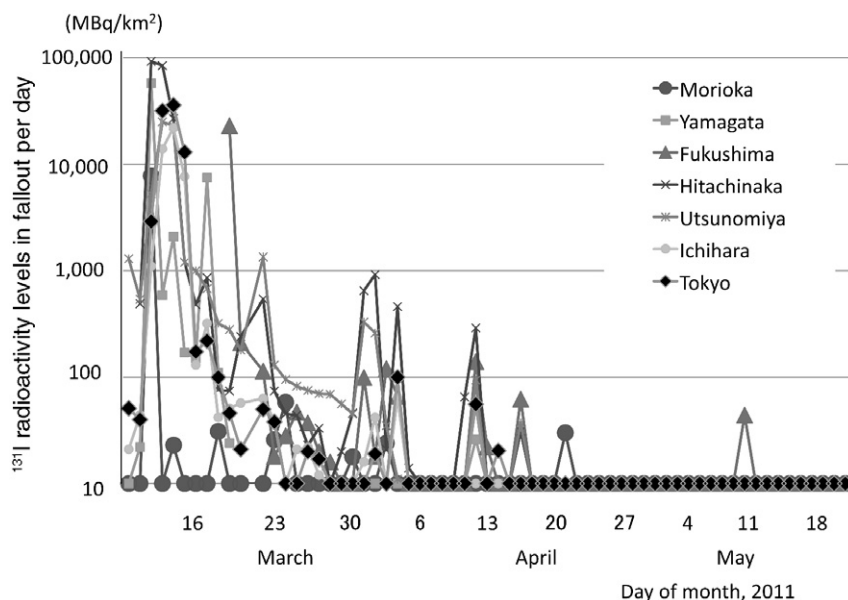
A total of 126 breast milk samples were collected from 119 volunteer lactating women; 37 women were residing within 100 km of the FNP, 60 were within 100–199 km and 22 were within 200–249 km of the FNP between April 24 and May 31. Of them, seven women who exhibited a detectable  $^{131}\text{I}$  level in their first breast milk sample provided a second breast milk sample approximately two to three weeks later. Each of the breast milk samples was placed in a cylindrical, 100-mL plastic container used to determine the  $^{131}\text{I}$  content and was monitored for two to three hours using a gamma spectrometry system equipped with high-purity germanium detectors (GR2519: Canberra, Meriden, CT, USA; EGPC20-190-R: Eurysis, Lingolsheim, France; and GEM20P4: Ortec, Oak Ridge, TN, USA) connected to multichannel analyzers and the analytical software. The energy and efficiency calibrations were performed using the standard volume radionuclide gamma sources with the same diameter of cylindrical plastic container (MX033U8 of Japan Radioisotope Association, Tokyo, Japan) composed of  $^{109}\text{Cd}$ ,  $^{57}\text{Co}$ ,  $^{139}\text{Ce}$ ,  $^{51}\text{Cr}$ ,  $^{85}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{54}\text{Mn}$ ,  $^{88}\text{Y}$  and  $^{60}\text{Co}$ .

## Other data collection

Data on the air radiation dose rate and  $^{131}\text{I}$  radioactivity in fallout in various cities were obtained from the official websites of the Japanese Ministry of Education,



**Figure 2** Chronological changes in air radiation dose rates in various cities/areas. Fukushima is located 60 km northwest of the Fukushima nuclear power plant, Iwaki is 45 km south, Kooriyama is 60 km west, Mito is 130 km south, Hitachiomiya is 110 km south, Sendai is 100 km north and Tokyo is 230 km southwest of the Fukushima nuclear power plant.



**Figure 3** Chronological changes in  $^{131}\text{I}$  radioactivity levels in fallout within 24 h in various cities. Morioka is located 255 km north of the Fukushima nuclear power plant, while Yamagata is 110 km northwest, Hitachinaka is 130 km south, Utsunomiya is 140 km southwest and Ichihara is 230 km south of the Fukushima nuclear power plant.

Culture, Sports, Science and Technology (MEXT) ('Reading of environmental radioactivity level', cited September 15, 2011; available from URL: <http://www.mext.go.jp/english/index.htm>). The  $^{131}\text{I}$  concentration in tap water, spinach, cows milk, and chicken eggs sampled in various cities were obtained from the official websites of the JMHLW ('Information on the Great East Japan Earthquake from Ministry of Health, Labour and

Welfare', cited September 15, 2011; available from URL: <http://www.mhlw.go.jp/english/index.html>) and the official websites of various cities. A Japanese citizens group represented by Kikuko Murakami independently determined the  $^{131}\text{I}$  content in the breast milk of 28 women between March 24 and April 29. Data regarding the  $^{131}\text{I}$  content in these 28 women and relevant information released by the citizens group on April

21 and May 18 were obtained from their website ('Radioactivity in breast milk', cited September 15, 2011; available from URL: <http://bonyuutyousa.net/>).

## Results

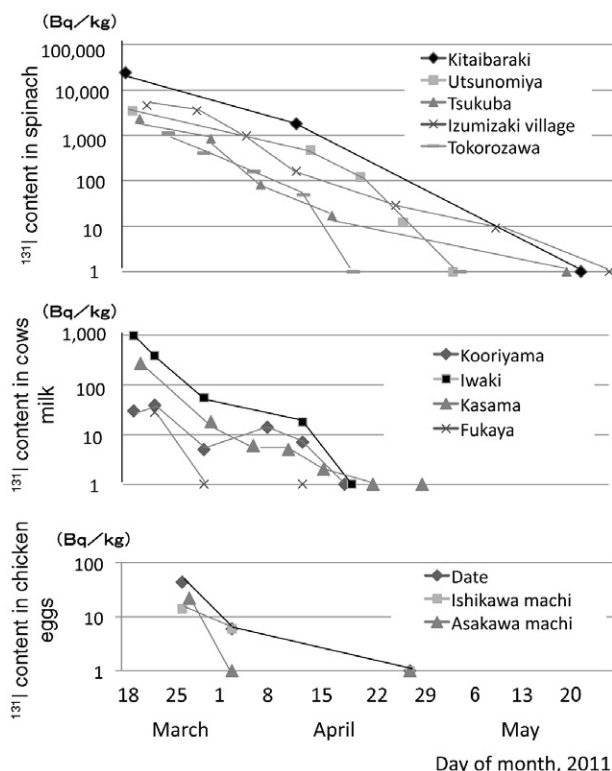
### Environmental pollution

Air pollution with radioactive materials occurred over a geographically wide area within 300 to 400 km of the FNP in the morning of March 15, 2011 (Fig. 2). Although the air radiation dose rate was  $<0.07 \mu\text{Gy/h}$  before the FNP accident in the areas shown in Figure 1, it increased sharply to  $19 \mu\text{Gy/h}$  in Fukushima city on March 15, then decreased to  $1.6 \mu\text{Gy/h}$  by the end of May. In Tokyo, located 230 km south of the FNP, the highest radiation dose rate of  $0.81 \mu\text{Gy/h}$  on March 15 decreased to  $<0.07 \mu\text{Gy/h}$  by mid-April.

The amount of  $^{131}\text{I}$  radioactivity in fallout per day reached a peak level of  $93\,000 \text{ MBq/km}^2$  in Hitachinaka city, located 130 km south of the FNP, on March 20, while it reached a peak level of  $38\,000 \text{ MBq/km}^2$  in Tokyo on March 22 (Fig. 3). Consequently, vegetables such as spinach, cows milk and chicken eggs were also contaminated with  $^{131}\text{I}$  (Fig. 4). The highest content of  $^{131}\text{I}$  was  $24\,000 \text{ Bq/kg}$ , found in spinach on March 18 in Kitaibaraki city, located 75 km south of the FNP. The  $^{131}\text{I}$  content in spinach decreased over time; for example, a level of  $3500 \text{ Bq/kg}$  was recorded in Utsunomiya city on March 19, decreasing to  $480 \text{ Bq/kg}$  on April 13,  $120 \text{ Bq/kg}$  on April 20,  $12 \text{ Bq/kg}$  on April 26, and became undetectable on May 3 (Fig. 4). Among the three foods, the  $^{131}\text{I}$  content was lowest in chicken eggs. It rained on March 20 and 21 in these areas, and the rain accelerated the pollution of water with  $^{131}\text{I}$  (Fig. 5). In Tokyo,  $^{131}\text{I}$  radioactivity in tap water from the Kana-machi water purification plant reached a peak level of  $210 \text{ Bq/kg}$  on March 22. The content of  $^{131}\text{I}$  in the tap water decreased and became undetectable in many cities by mid-April (Fig. 5).

### Contamination of breast milk with $^{131}\text{I}$

Seven of 23 women (30.4%) who were tested in April secreted a detectable level of  $^{131}\text{I}$  in their breast milk (Table 1). The concentrations ranged from 2.2 to  $8.0 \text{ Bq/kg}$  and appeared to be higher than those in tap water available for these seven women at the same time points. As expected from the data on  $^{131}\text{I}$  radioactivity in the fallout, vegetables and water (Figs 3 to 5), the radioactivity of  $^{131}\text{I}$  in the breast milk became undetectable by May 15 in these seven women (Table 1). None of the remaining 96 women tested in May exhibited a detect-



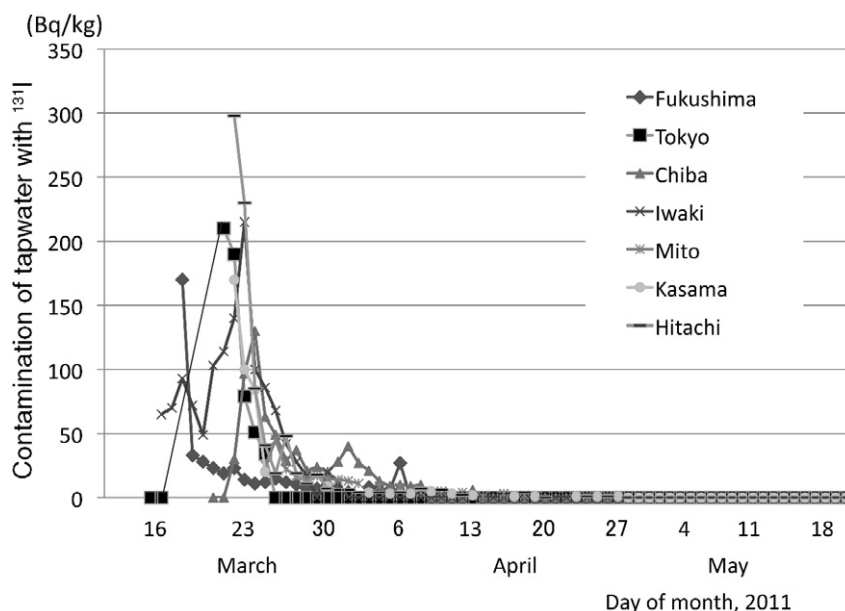
**Figure 4** Chronological changes in  $^{131}\text{I}$  radioactivity levels in spinach, cows milk and chicken eggs in various cities/areas. Kitaibaraki is located 75 km south of the Fukushima nuclear power plant, Tsukuba is 170 km southwest, Izumizaki village is 70 km southwest, Tokorozawa is 228 km southwest, Kasama is 135 km south, Fukaya is 210 km southwest, Date is 60 km northwest, Ishikawa machi is 60 km southwest and Asakawa machi 70 km southwest of the Fukushima nuclear power plant.

able amount of  $^{131}\text{I}$  in their breast milk samples with detection limits of  $1.6 \pm 0.3 \text{ Bq/kg}$  (data not shown).

## Discussion

The present study demonstrated that environmental pollution with  $^{131}\text{I}$  causes the contamination of breast milk with  $^{131}\text{I}$ . According to the data released by a Japanese citizens group represented by Kikuko Murakami, which independently determined the  $^{131}\text{I}$  content in the breast milk of 28 women between March 24 and April 29 (<http://bonyuutyousa.net/>) (Table 2), four out of six women (66.7%) tested in March exhibited a much higher  $^{131}\text{I}$  radioactivity than women tested by our group in April. The amount of  $^{131}\text{I}$  radioactivity decreased over time in two women (cases 26 and 28), as





**Figure 5** Chronological changes in tap water pollution with  $^{131}\text{I}$  in various cities/areas. Chiba is located 220 km south and Hitachi 98 km south of the Fukushima nuclear power plant.

was seen in seven women in our study (Table 1, cases 1 and 6–11). Thus, the contamination of breast milk with  $^{131}\text{I}$  may have reflected the degree of environmental pollution with  $^{131}\text{I}$ . However, if the citizens group had used an assay system similar to the one used by our group, which is able to detect  $^{131}\text{I}$  at a level of around 2.0 Bq/kg, the detection ratio of  $^{131}\text{I}$  among the 28 women may have been higher than the reported rate of 14.3% (4/28).

The most reliable data to date on the relationship between the thyroid radiation dose and the risk for thyroid cancer following the environmental release of  $^{131}\text{I}$  was obtained after the Chernobyl reactor accident in April 1986.<sup>6</sup> Thyroid exposure to radiation after the Chernobyl reactor accident was virtually all internal, from radioiodines.<sup>6</sup> The inhalation of airborne  $^{131}\text{I}$  may occur after its release and prior to the deposition of  $^{131}\text{I}$  on the ground; however, in seven Ukraine cities following the release of radioiodine from the Chernobyl nuclear power plant, the inhalation of  $^{131}\text{I}$  was estimated to contribute to between 2 and 13% of the total absorbed radiation dose, whereas the ingestion pathway contributed between 87 and 98%.<sup>8</sup> Therefore, human breast milk was speculated to contribute to the dose of  $^{131}\text{I}$  received by nursing infants in the vicinity of the Chernobyl reactor accident. Iodine is an essential nutrient required for the production of thyroid hormone, and the diet is the major source of iodine intake. Cows and goats absorb iodine from ingested vegetables and water. The absorbed iodine is then

excreted into their milk.<sup>9</sup> In addition,  $^{131}\text{I}$  administered orally or intravenously for medical purposes also accumulates in the thyroid and breast tissues and is excreted in breast milk.<sup>3–5</sup> These findings have supported the speculation that human breast milk contributed to the development of thyroid cancer in infants after the Chernobyl accident. In some regions, for the first four years after the accident, the incidence of thyroid cancer among children aged 0 to 4 years old at the time of the accident exceeded the expected number of cases by 30- to 60-fold.<sup>6</sup> Before the end of the first decade, the annual incidence of thyroid cancer increased in children under the age of 15 years at the time of accident from a baseline incidence of <1.0 per 100 000 individuals to >100 per 100 000 individuals in the region with the highest contamination levels.<sup>10–13</sup>

A significant correlation is seen between the level of iodine intake and the iodine content of human milk, with a correlation coefficient ( $r$ ) of 0.41 or 0.82.<sup>1,2</sup> The National Research Council (NRC) of the United States recommends a dietary allowance for iodine of 150  $\mu\text{g}/\text{day}$ , with additional allowances of 25 and 50  $\mu\text{g}/\text{day}$  during pregnancy and lactation, respectively.<sup>9</sup> Lactating women in the United States excrete milk containing an iodine level of  $178 \pm 127 \mu\text{g}/\text{L}$  (mean  $\pm$  SD).<sup>1</sup> Korean lactating women reportedly consume  $1295 \pm 946 \mu\text{g}$  of iodine daily and excrete milk containing  $892 \pm 1037 \mu\text{g}/\text{L}$  of iodine.<sup>2</sup> On the assumption that these lactating women produced 600–800 mL of breast milk daily, 40–70% of the iodine consumed by

**Table 1** Radioiodine ( $^{131}\text{I}$ ) concentrations in breast milk from 23 women and in tap water available for these women

| Case No. | City/area (distance, direction from the FNP) | $^{131}\text{I}$ concentration (Bq/kg) |                                  |
|----------|--|--|----------------------------------|
|          |  | Breast milk                            | Tap water†                       |
| 1-1      | Kasama (170 km, S)                           | 2.3 (4/24)                             | 0.6 (4/21), 0.6 (4/27)           |
| 1-2      |  | <1.3 (5/8)                             |                                  |
| 2        | Yuki (160 km, SW)                            | <2.0 (4/24)                            | 5.6 (3/23), ND (4/20)            |
| 3        | Koshigaya (200 km, SW)                       | <1.9 (4/24)                            | 0.6 (4/24), ND (4/25)            |
| 4        | Tokyo (230 km, SW)                           | <1.6 (4/24)                            | 8.0 (4/4), <8 (4/5)              |
| 5        | Tokyo (230 km, SW)                           | <1.8 (4/24)                            | 8.0 (4/4), <8 (4/5)              |
| 6-1      | Iwaki (45 km, S)                             | 3.5 (4/25)                             | 13.6 (4/3), <4.0 (4/5)           |
| 6-2      |  | <1.0 (5/8)                             |                                  |
| 7-1      | Kasama (135 km, S)                           | 2.3 (4/25)                             | 1.2 (4/25), ND (4/28)            |
| 7-2      |  | <1.4 (5/8)                             |                                  |
| 8-1      | Hitachiomiya (110 km, S)                     | 3.0 (4/25)                             | 6.8 (4/14), NA thereafter        |
| 8-2      |  | <1.3 (5/9)                             |                                  |
| 9-1      | Shimozuma (170 km, SW)                       | 2.2 (4/25)                             | 18 (3/29), ND (4/5)              |
| 9-2      |  | <1.1 (5/15)                            |                                  |
| 10-1     | Mito (130 km, S)                             | 8.0 (4/25)                             | 1.4 (4/22), 1.2 (4/26), ND (5/9) |
| 10-2     |  | <1.2 (5/9)                             |                                  |
| 11-1     | Chiba (220 km, S)                            | 2.3 (4/25)                             | 5.8 (4/15), ND (4/16)            |
| 11-2     |  | <1.4 (5/9)                             |                                  |
| 12       | Fukushima (60 km, NW)                        | <1.7 (4/25)                            | 6.0 (4/10), ND (4/11)            |
| 13       | Kooriyama (60 km, SW)                        | <1.8 (4/25)                            | 8.0 (4/16), ND (4/17)            |
| 14       | Soma county (40 km, N)                       | <2.0 (4/25)                            | NA                               |
| 15       | Kasama (135 km, S)                           | <2.1 (4/25)                            | 1.2 (4/25), ND (4/28)            |
| 16       | Hitachinaka (120 km, S)                      | <2.1 (4/25)                            | ND (4/24), 0.7 (4/26)            |
| 17       | Naka (120 km, S)                             | <2.7 (4/25)                            | 6.4 (4/11), ND (4/13)            |
| 18       | Chiba (220 km, S)                            | <3.6 (4/25)                            | 5.8 (4/15), ND (4/16)            |
| 19       | Tokyo (230 km, SW)                           | <2.3 (4/25)                            | 8.0 (4/4), <8 (4/5)              |
| 20       | Tokyo (230 km, SW)                           | <2.8 (4/25)                            | 8.0 (4/4), <8 (4/5)              |
| 21       | Tokyo (230 km, SW)                           | <2.4 (4/25)                            | 8.0 (4/4), <8 (4/5)              |
| 22       | Tokyo (230 km, SW)                           | <6.5 (4/25)                            | 8.0 (4/4), <8 (4/5)              |
| 23       | Tokyo (230 km, SW)                           | <2.6 (4/25)                            | 8.0 (4/4), <8 (4/5)              |

Seven women (cases 1 and 6–11) provided two samples at different time points. The other 96 women who resided within 250 km of the FNP provided breast milk samples between May 20 and May 31 for the determination of  $^{131}\text{I}$  content, but none of the 96 women exhibited a detectable level in their breast milk samples. †Available data released by corresponding prefectural or city Bureau of Waterworks and the Japan Ministry of Education, Culture, Sports, Science and Technology. The date (month/day) of sampling is indicated in parentheses. FNP, Fukushima nuclear power plant; N, north; NA, not available; ND, not detected without information on detection limit; NW, northwest; S, south; SW, southwest; W, west.

the mother enters the breast milk. The  $^{131}\text{I}$  content in the breast milk of cases 25 and 26 (8.7 and 31.8 Bq/kg) determined by a citizens group was approximately one-half of the levels in tap water (16.7 and 80 Bq/kg) available for these women (Table 2). The extent of contamination with  $^{131}\text{I}$  was larger in vegetables than in cows milk or chicken eggs, as shown in Figure 4. Since these two women may have consumed vegetables contaminated to an unknown extent, the major sources of  $^{131}\text{I}$  were considered to be tap water and vegetables. If we assume that cases 25 and 26 consumed 200 g of contaminated vegetables containing 100 Bq/kg  $^{131}\text{I}$  and 1.0 L of tap water and produced 700 mL of milk daily, approximately 17–26% of the  $^{131}\text{I}$  consumed by the mothers would have entered the milk. Because stable iodine (such as potassium iodide) competes with  $^{131}\text{I}$  in being

taken up by the thyroid gland, thus preventing the accumulation of  $^{131}\text{I}$  in the thyroid gland,<sup>14</sup> and is used for the prevention of  $^{131}\text{I}$ -induced thyroid cancer,<sup>15</sup> and because radioiodine is also known to accumulate in the breasts of lactating women,<sup>3</sup> stable iodine may compete with  $^{131}\text{I}$  in being secreted into the breast milk. Because Japanese foods contain high concentrations of iodine<sup>16</sup> it is not surprising that a relatively small fraction of the  $^{131}\text{I}$  consumed by cases 25 and 26 entered their breast milk.

In the presence of a very low level of  $^{131}\text{I}$  in the tap water after mid-April, the  $^{131}\text{I}$  content in the breast milk exceeded that in the tap water in a significant number of women, as shown in cases 1, 7 and 10. This may imply that lactating women had difficulty avoiding contaminated vegetables, because vegetables

**Table 2** Radioiodine ( $^{131}\text{I}$ ) concentrations in breast milk from 28 women and in tap water available to them, as determined by a citizens group

| Case No. | City/area (distance, direction from the FNP) | $^{131}\text{I}$ concentration (Bq/kg) |                                      |
|----------|--|--|--------------------------------------|
|          |  | Breast milk                            | Tap water†                           |
| 24       | Tsukubamirai (210 km, SW)                    | ND (3/23)                              | 36 (3/23), 26 (3/24), <16 (3/28)     |
| 25       | Tsukuba (170 km, SW)                         | 8.7 (3/23)                             | 4.9 (3/20), 16.7 (3/24), 14.9 (3/29) |
| 26-1     | Moriya (210 km, SW)                          | 31.8 (3/23)                            | 80 (3/23), 49 (3/25), 38 (3/26)      |
| 26-2     |  | 8.5 (3/30)                             | 9.0 (3/29), 5.9 (3/31), 5.0 (4/2)    |
| 27       | Tsukuba (170 km, SW)                         | 6.4 (3/29)                             | 14.9 (3/29), 13.4 (3/31), 11.2 (4/2) |
| 28-1     | Kashiwa (200 km, S)                          | 36.3 (3/29)                            | 14 (3/26), ND (3/28)                 |
| 28-2     |  | 14.8 (4/4)                             | 14 (3/26), ND (3/28)                 |
| 29       | Shiroishi (70 km, N)                         | ND (3/30)                              | 10 (3/25), 2.0 (3/30), 1.7 (4/6)     |
| 30       | Tanakura-machi (70 km, SW)                   | <4.7 (4/12)                            | 4.7 (3/27), ND (3/30)                |
| 31       | Fukushima (60 km, NW)                        | <6.8 (4/12)                            | 6.0 (4/10), ND (4/11)                |
| 32       | Sukagawa (60 km, W)                          | <7.1 (4/21)                            | 4.1 (4/4), ND (4/6)                  |
| 33       | Inashiki county (175 km, S)                  | <4.2 (4/21)                            | 1.9 (4/21), 1.3 (4/26), 0.5 (5/6)    |
| 34       | Tuchiura (170 km, S)                         | <5.3 (4/22)                            | 2.1 (4/20), 1.6 (4/25), 1.2 (4/29)   |
| 35       | Iwaki (45 km, S)                             | <5.2 (4/23)                            | 13.6 (4/3), <4.0 (4/5)               |
| 36       | Kooriyama (60 km, SW)                        | <4.0 (4/24)                            | 8.0 (4/16), ND (4/17)                |
| 37       | Fukushima (60 km, NW)                        | <4.7 (4/24)                            | 6.0 (4/10), ND (4/11)                |
| 38       | Iwaki (45 km, S)                             | <5.3 (4/25)                            | 13.6 (4/3), <4.0 (4/5)               |
| 39       | Fukushima (60 km, NW)                        | <4.3 (4/25)                            | 6.0 (4/10), ND (4/11)                |
| 40       | Higashiibaraki county (125 km, S)            | <5.7 (4/26)                            | 1.2 (4/26), 0.8 (4/28), ND (5/9)     |
| 41       | Chousei county (235 km, S)                   | <4.4 (4/26)                            | 5.2 (4/8), ND (4/9)                  |
| 42       | Kisarazu (250 km, S)                         | <4.5 (4/26)                            | 7.6 (3/28), ND (3/29)                |
| 43       | Sukagawa (60 km, S)                          | <5.1 (4/26)                            | 4.1 (4/4), ND (4/6)                  |
| 44       | Iwaki (45 km, S)                             | <5.7 (4/26)                            | 13.6 (4/3), <4.0 (4/5)               |
| 45       | Sukagawa (60 km, S)                          | <5.3 (4/27)                            | 4.1 (4/4), ND (4/6)                  |
| 46       | Date county (45 km, NW)                      | <4.1 (4/27)                            | 23.2 (4/2), ND (4/3)                 |
| 47       | Kooriyama (60 km, SW)                        | <5.5 (4/27)                            | 8.0 (4/16), ND (4/17)                |
| 48       | Kooriyama (60 km, SW)                        | <6.3 (4/27)                            | 8.0 (4/16), ND (4/17)                |
| 49       | Mito (130 km, S)                             | <7.6 (4/28)                            | 0.8 (4/28), 0.5 (5/2), ND (5/9)      |
| 50       | Nishishirakawa county (85 km, W)             | <7.0 (4/28)                            | 5.5 (4/9), ND (4/11)                 |
| 51       | Fukushima (60 km, NW)                        | <6.1 (4/29)                            | 6.0 (4/10), ND (4/11)                |

This table was based on data released by a citizens group represented by Kikuko Murakami on April 21 and May 18. The  $^{131}\text{I}$  content was measured by Tokyo Nuclear Service Tsukuba Development Centre (Tsukuba, Japan) using 120–130 mL samples of breast milk and a germanium detector (GEM20P4; Seiko EG&G, Tokyo, Japan) for 1800 s. The detection limit for  $^{131}\text{I}$  was reportedly 4.0–7.6 Bq/kg, as shown in this table. Two women (cases 26 and 28) provided two samples at different time points. †Available data released by corresponding prefectural or city Bureau of Waterworks and the Japan Ministry of Education, Culture, Sports, Science and Technology. The date (month/day) of sampling is indicated in parentheses. FNP, Fukushima nuclear power plant; N, north; ND, not detected without information on detection limit; NW, northwest; S, south; SW, southwest; W, west.

containing <2000 Bq/kg of  $^{131}\text{I}$  were sold in market-places, according to Japanese regulations.

During the FNP accident, the FNP explosively dispersed a massive radioactive plume on the morning of March 15 (Figs 2 and 3). Although the degrees of food and water contamination with  $^{131}\text{I}$  were monitored in various cities/areas and the data were released promptly through official websites of the Japanese government, the majority of citizens may not have been aware of the danger concerning internal exposure to  $^{131}\text{I}$  ingested from water and vegetables prior to the first announcements made on March 18 and 22 regarding vegetable and tap water contamination, respectively. These announcements were first made after confirming

that the  $^{131}\text{I}$  content exceeded the allowed threshold in Japan: 2000 Bq/kg for vegetables and 100 Bq/kg for drinking water consumed by infants <1 year old (300 Bq/kg for older children and adults). Since the degree of  $^{131}\text{I}$  contamination in the tap water and in vegetables was much higher before March 22 than after March 22 in many cities, as expected from the data shown in Figures 2 to 5, the total amount of  $^{131}\text{I}$  ingested by the mothers before March 22 may have far exceeded that ingested after March 22. If we had conducted this study earlier, around March 20, a much higher  $^{131}\text{I}$  content in the breast milk would likely have been detected. Thus, nursing infants may also have been exposed to large doses before March 22.

The radiation doses received after the Chernobyl accident remain somewhat uncertain.<sup>6</sup> Our findings regarding the extent of breast milk contamination with <sup>131</sup>I in relation to the extent of the pollution of the atmosphere, water and vegetables may be helpful in the future and may enable a relatively accurate estimation of the relationship between breast milk contamination with <sup>131</sup>I and the development of infant thyroid cancer. However, large differences in the level of exposure after the Chernobyl reactor accident were reported to exist between neighboring villages, within families inside the same village, or even within the same family depending on diet, living habits and occupation, and the level of exposure was considered to depend mainly on individual behavior.<sup>17</sup> Therefore, the possibility that the participants in this study may have been more interested in the danger of breast milk contamination with <sup>131</sup>I than lactating women in general should be kept in mind, as the study population may not be representative of lactating women in general.

## Disclosure

All authors declare that they have no financial relationship with a biotechnology manufacturer, a pharmaceutical company or other commercial entity that has an interest in the subject matter or materials discussed in the manuscript.

## Authors' Contributions

Nobuya Unno: study design, data analysis, coordination with the JMHLW and draft preparation; Hisanori Minakami: study design, data analysis and draft preparation; Takahiko Kubo: responsible for privacy protection and illustrations; Keiya Fujimori: data sampling and critical discussion; Isamu Ishiwata: data sampling and critical discussion; Hiroshi Terada: measurement of <sup>131</sup>I in the breast milk; Shigeru Saito: data sampling and critical discussion; Ichiro Yamaguchi: measurement of <sup>131</sup>I in the breast milk; Naoki Kunugita: measurement of <sup>131</sup>I in the breast milk, critical discussion and obtaining approval from the institutional review board of the Ethics Committee; Akihito Nakai: data sampling and critical discussion; Yasunori Yoshimura: supervision. This study was supported by the Japanese Ministry of Health, Labour and Welfare (JMHLW).

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