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Estimation of thyroid doses and health risks resulting from the intake of radioactive iodine in foods and drinking water by the citizens of Tokyo after the Fukushima nuclear accident

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ABSTRACT

The release of radioactive materials from the Fukushima nuclear power plant after the Great East Japan Earthquake on 11 March 2011 poses health risks. In this study, the intake of iodine 131 (I-131) in drinking water and foods (milk, dairy products, and vegetables) by citizens of Tokyo was estimated. The effects of countermeasures (restrictions on the distribution of foods and the distribution of bottled water for infants) on reducing intake were also evaluated. The average thyroid equivalent doses without countermeasures from 21 March 2011 were 0.42 mSv in adults, 1.49 mSv in children, and 2.08 mSv in infants. Those with countermeasures were 0.28, 0.97, and 1.14 mSv respectively, reductions of 33%, 35%, and 45%. Drinking water contributed more to intake by adults and children than foods. The intake of I-131 within the first 2 weeks was more than 80% of the estimated intake, owing to its short half-life, indicating that rapid countermeasures are important in reducing intake. The average risks of cancer incidence and mortality due to I-131 for infants were estimated to be 3×10^{-5} and 0.2×10^{-5}, respectively, lower than the annual risks of traffic accidents, naturally occurring radioactive material (potassium 40), and environmental pollutants such as diesel exhaust particles.

1. Introduction

Radioactive materials were released from the Tokyo Electric Power Company's Fukushima nuclear power plant after the Great East Japan Earthquake on 11 March 2011. They were diffused into the atmosphere, deposited with rainfall, and incorporated into surface waters and agricultural crops. Consequently, radioactive iodine 131 (I-131) and caesium 134 and 137 (Cs-134 and Cs-137) have been detected in drinking water and foods in several prefectures in Japan (Ministry of Health, Labour and Welfare, 2011b; Tokyo Metropolitan Institute of Public Health, 2011).

Since I-131 is a human carcinogen, causing thyroid cancer in particular, the increased health risk caused by consumption of drinking water and foods is a matter of concern. Children and adolescents exposed to radioiodines from fallout from the Chernobyl accident of 1986 had a sizeable dose-related increase in thyroid cancer (Cardis and Hatch, 2011). Health effects of the Chernobyl accident for most individuals were attributed mainly to the intake of I-131 in fresh cow's milk (World Health Organization, 2006).

To protect the health of the populace, the Japanese government announced "indices relating to limits on food and drink ingestion" as provisional regulation values on 17 March 2011, and revised them on 5 April 2011 (Table S1) (Ministry of Health, Labour and Welfare, 2011b). These values were basically established according to control index levels set by the Nuclear Safety Commission of Japan (Nuclear Safety Commission of Japan, 1998; Suga and Ichikawa, 2000) after the Chernobyl accident. The index levels for I-131 were determined as follows.

- The total annual thyroid equivalent dose of total radioiodines due to the ingestion of foods and water must not exceed an intervention level of 50 mSv y^{-1} (=2 mSv y^{-1} of effective dose calculated by using a tissue weighting factor of 0.04 (ICRP, 2007)); this was determined by considering the balance between the health risk and the cost of countermeasures. The dose of I-131 accounts for approximately 90% of total radioiodines.

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- Two-thirds of the total thyroid equivalent dose was distributed equally among drinking water, milk and dairy products, and vegetables except root vegetables and tubers (11.1 mSv y^{-1} each). The other one-third (16.7 mSv y^{-1}) was reserved for other foods.
- The concentration of I-131 in water and foods was assumed to decrease according to the half-life (8.04 d (ICRP, 1983)).
- The minimum concentration was established as the index level by considering the intake by adults, children (5 y old), and infants (<1 y old).

Since I-131 concentrations in some foods such as milk and spinach exceeded the provisional regulation values (Ministry of Health, Labour and Welfare, 2011b), the government restricted the distribution of foods collected in the some municipalities from 21 March 2011. The Tokyo Metropolitan Government distributed 1650 mL of bottled water for each infant on the morning of 24 March 2011, because of the detection of 210 Bq kg^{-1} of I-131 in drinking water from the Kanamachi purification plant in Tokyo on 22 March, which exceeded the provisional regulation value for infants (Tokyo Metropolitan Institute of Public Health, 2011).

Internal exposure by the consumption of water and foods has not been unravelled, whereas external exposure can be determined through concentrative monitoring of radioactive materials. Although a working group of the Ministry of Health, Labour and Welfare (2011a) has started to assess the nationwide-average internal exposure to radioactive materials by consumption of foods, internal exposure must be assessed at the regional scale.

Therefore, this study estimated the average and maximum thyroid doses resulting from intake of I-131 by citizens of Tokyo. The effects of countermeasures (i.e., restrictions on the distribution of foods and the distribution of bottled water for infants) on reducing intake were also evaluated. The total and fatal risks of cancer (i.e., risks of cancer incidence and mortality) from the intake of I-131 were estimated for comparison with the risks from other causes including naturally occurring radioactive material (potassium 40 (K-40)), environmental pollutants, diseases, and accidents. This is the first study to reveal the thyroid doses and the risks posed by intake of I-131 in drinking water and foods after the Fukushima nuclear accident.

2. Materials and methods

2.1. Intake pathway and scenarios

The thyroid equivalent doses of I-131 in adults, children (5 y old), and infants (<1 y old) were estimated to assess intake via the ingestion of drinking water, milk and dairy products, and vegetables (except root vegetables and tubers), based on the classification of control index levels by the Nuclear Safety Commission of Japan (1998). Although I-131 was detected in *Ammodytes personatus* (sand lance; one species of Perciformes) (Ministry of Health, Labour and Welfare, 2011b), intake by consumption of these fish was not considered here owing to the low catch and low consumption in comparison to drinking water, milk and dairy products, and vegetables. The thyroid dose coefficients for ingestion of I-131 were 4.3×10^{-4} mSv Bq^{-1} in adults, 2.1×10^{-3} mSv Bq^{-1} in children, and 3.7×10^{-3} mSv Bq^{-1} in infants (ICRP, 1993). These values were used so as not to underestimate the dose, with the assumption that the fractional thyroid uptake from blood is 0.3, as used in the determination of control index levels by the Nuclear Safety Commission of Japan (1998).

The intake of I-131 via the consumption of milk and dairy products and of vegetables collected in Fukushima Prefecture and the Kanto region (Ibaraki, Tochigi, Gunma, Saitama, Chiba, and Kanagawa prefectures plus Tokyo) was considered, but that in other prefectures was regarded to be negligible, because I-131 concentrations in these products in other prefectures were much lower (Figs. S1 and S2). Data released from March to July 2011 (Ministry of Health, Labour and Welfare, 2011b; Tokyo Metropolitan Institute of Public Health, 2011) were used.

On the assumption that the current situation will continue without further incident, the annual intake was estimated from 21 March 2011, when the Japanese government restricted the distribution of foods. Although intake before then might not have been negligible, sufficient monitoring data were not available. It is likely that rainfall on the night of 15/16 March (Japan Meteorological Agency, 2011) caused heavy radionuclide contamination in surface waters and agricultural crops. Milk and dairy products and vegetables collected on 16 March might therefore have been contaminated. Therefore, the intake from 18 to 20 March was separately estimated, on the assumption that collection for delivery to market in Tokyo takes 2 days; the results are shown separately in the text and the Supplementary Materials.

The effects of countermeasures (restrictions on the distribution of foods and the distribution of bottled water for infants) on reducing intake were evaluated. When the countermeasures were considered, the consumption of foods collected in the areas where the distribution was restricted was assumed to be nil (that is, residents ate alternative foods containing negligible I-131). The effects of the distribution of bottled water for infants were evaluated by assuming that the intake via the consumption of drinking water on 24 and 25 March was negligible.

2.2. Drinking water

I-131 concentrations in tap water in Shinjuku, Tokyo (Tokyo Metropolitan Institute of Public Health, 2011), were used to estimate intake via the consumption of drinking water in Tokyo (Fig. S3). The concentration of I-131 in the tap water peaked on 26 March. As I-131 has been not detected in tap water since 4 May, intake after then was regarded as negligible. The average daily consumption rates of drinking water were set at 1650 g d^{-1} per adult, 1000 g d^{-1} per child, and 710 g d^{-1} per infant (Nuclear Safety Commission of Japan, 1998). The consumption of water in soup and rice was added. The average daily consumption rate of soup was 160 g d^{-1} per adult (Kuroda et al., 2011). The consumption rates of 84 g d^{-1} per child and 42 g d^{-1} per infant were calculated from the consumption by adults and the ratio of miso (soybean paste) consumption between adults and children or infants (Izumo, 2001). The consumption rates of water included in rice were calculated from those of rice (170.2 g d^{-1} per adult, 97.7 g d^{-1} per child, and 48.9 g d^{-1} per infant; (Izumo, 2001)) and the ratio of cooking water to rice (=1.3 g g^{-1} (Nakamura et al., 1996)): 220 g d^{-1} per adult, 130 g d^{-1} per child, and 64 g d^{-1} per infant. In total, the average daily consumption rates of drinking water were 2030 g d^{-1} per adult, 1210 g d^{-1} per child, and 820 g d^{-1} per infant (Table S2). Since these values include bottled soft drinks (nationwide average of 400 g d^{-1} per person (Japan Soft Drink Association, 2010)), the intake in drinking water could be overestimated.

2.3. Milk and dairy products

The concentrations of I-131 in milk and dairy products (Ministry of Health, Labour and Welfare, 2011b) were classified according to prefecture and date. The prefectures where the distribution was restricted were further classified into individual groups (Table S3). The maximum I-131 concentration in milk and dairy products was 5300 Bq kg^{-1} (Fig. S1), which was much lower than that during the Chernobyl accident (~40 000 Bq kg^{-1}, time-corrected to 8 May 1986 (Balonov et al., 2000)). The arithmetic mean concentration in each area on each day was calculated. The post-incident concentrations before monitoring began were projected from the initial measured concentrations and the half-life of I-131 (Fig. 1). The consumption dates and concentrations in milk and dairy products were assumed to be the same as those released by the Ministry of Health, Labour and Welfare (2011b).

The daily consumption rates of milk and dairy products were 200 g d^{-1} per adult, 500 g d^{-1} per child, and 600 g d^{-1} per infant (Nuclear Safety Commission of Japan, 1998) (Table S2). To estimate the average intake, arrival shares in individual areas (i.e., proportions of raw milk produced in each area for the market in Tokyo) were based on the production and transport of raw milk in each prefecture and the amount of milk transported from each prefecture to Tokyo (Ministry of Agriculture Forestry and Fisheries, 2011b) (Table S4). The number of cows in each municipality of Fukushima, where distribution was restricted (Ministry of Agriculture Forestry and Fisheries, 2007), was used to estimate the ratios of raw milk production among the groups. The average intake was estimated from the following equation:

$$\text{Average intake of I-131(Bq)} = A \times \sum_t \sum_i (B_i/100 \times C_{it}) \qquad (1)$$

where A is the average daily consumption rate (g d^{-1} per person), B is the arrival share in an area (%), C is the arithmetic mean concentration of I-131 in milk and dairy products in each area (Bq g^{-1}), t is the consumption date, and i is the individual area (classified group).

The maximum intake was estimated by assuming that all milk and dairy products ingested by a person were contaminated at the maximum values, as

$$\text{Maximum intake of I-131(Bq)} = A \times \sum_t D_t \qquad (2)$$

where D is the maximum concentration of I-131 in the whole target area (Bq g^{-1}).

2.4. Vegetables

Nine vegetables were considered: *Spinacia oleracea* (spinach), *Brassica oleracea* var. *capitata* (cabbage), *B. rapa* var. *glabra* (Chinese cabbage), *Lactuca sativa* var. *capitata* (head lettuce), *L. sativa* var. *crispa* (non-head lettuce (loose-leaf lettuce)), *B. rapa* var. *chinensis* (ging-geng-cai), *B. rapa* var. *perviridis* (mustard spinach), *Glebionis coronaria* (garland chrysanthemum), and *Allium fistulosum* (naganegi onion (Welsh onion)) (Fig. S4). Temporal trends of I-131 concentrations in spinach in Fukushima (Group RM3) are shown in Fig. 2. The average and maximum intakes of I-131 were estimated as with the milk and dairy products. The arrival shares were based on the amounts arriving from each prefecture at the Tokyo Metropolitan Central Wholesale Market in 2010 (Tokyo Metropolitan Central Wholesale Market, 2011) (Table S4). For prefectures where the distribution was restricted, the shares were estimated from the cultivated areas in each municipality (Ministry of Agriculture Forestry and Fisheries, 2011a).

I-131 concentrations in vegetables were not available in some areas. Since the arrival shares were very low in most cases except garland chrysanthemum (e.g., cabbage in Tochigi Pref., 0.03%), the intake from these areas was regarded as negligible. I-131 concentrations in garland chrysanthemum in Gunma were used for Ibaraki and Chiba (Group G2), where there were no monitoring data but the arrival shares were 10% and 32%, respectively, because there was a similarity in concentrations in spinach or ging-geng-cai among the three prefectures.

The daily consumption rates of spinach, cabbage, and Chinese cabbage followed the values in the report by Izumo (2001). The daily consumption rates of other vegetables were estimated from the total consumption rate of vegetables and the ratio of the arrival amounts of the targeted vegetable to the total arrival amounts of vegetables in the Tokyo Metropolitan Central Wholesale Market in 2010 (Tokyo Metropolitan Central Wholesale Market, 2011) (Table S2).

2.5. Estimation of cancer risk

The total and fatal cancer risks for infants, children, and adults from the ingestion of I-131 were estimated by using the average thyroid equivalent dose with countermeasures from 21 March 2011. The risks of fatal cancer at a given thyroid equivalent dose were calculated by Oka (2011) using Japanese life tables and the age-declining excess relative risk model based on the mortality of the atomic bomb survivors (Preston et al., 2003). The relationship between cancer risk and dose is not well understood and still uncertain (Mullenders et al., 2009). ICRP (2007) adopted dose and dose-rate effectiveness factor (DDREF) of 2, while the BEIR VII committee used a value of 1.5 (NAS/NRC, 2006). In this study, a linear relationship between cancer risk and dose (i.e. DDREF = 1) was assumed, in order not to underestimate the risks. The risks of fatal cancer at a thyroid equivalent dose of 1000 mSv were regarded as 1.78×10^{-3} for infants (0 y old), 1.47×10^{-3} for children (0–9 y old), and 6.4×10^{-4} for adults (20–34 y old). The total risks of cancer were calculated from the fatal risks and a lethality fraction of 0.07 (ICRP, 2007). The risks of I-131 were compared with those of environmental pollutants, K-40, diseases, and accidents. The risks

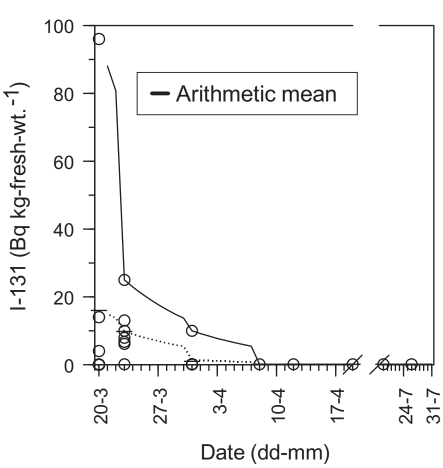

Fig. 1. Temporal trends of I-131 concentrations in milk and dairy products collected in Fukushima Pref. (Group RM1: Table S3) (Ministry of Health, Labour and Welfare, 2011b). Lines represent decline curves calculated from the half-life.

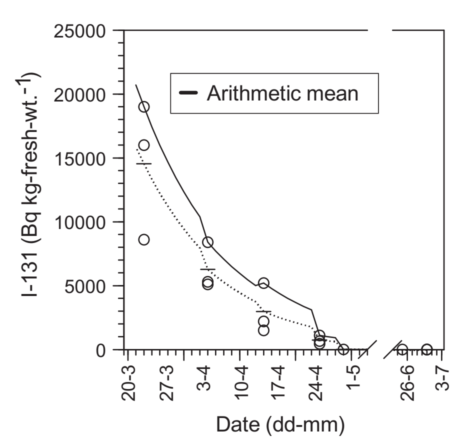

Fig. 2. Temporal trends of I-131 concentrations in spinach collected in Fukushima Pref. (Group RM3: Table S3) (Ministry of Health, Labour and Welfare, 2011b). Lines represent decline curves calculated from the half-life.

of cancer here represent the lifetime risks from annual intake of I-131, K-40, and environmental pollutants. The cancer risks from intake of environmental pollutants in Japan were estimated by following a study of risk ranking (Gamo et al., 2003). Diesel exhaust particles (Iwai and Utiyama, 2000), formaldehyde (USEPA, 1991a; Ministry of Health and Welfare, 1997), arsenic (Mashiko, 1989; USEPA, 1998a), dioxins (USEPA, 1997; Toyoda et al., 1999), benzene (Ministry of Health and Welfare, 1999; USEPA, 2000), DDTs (USEPA, 1988a,b, 1991b; Toyoda et al., 1998) and chlordane (Gamo et al., 1995; Tani et al., 1996; USEPA, 1998b) were selected. In addition, the risk of K-40, which is a dominant nuclide in foods irrespective of nuclear accidents, was also estimated. The annual effective dose of K-40 in the total diet was 0.17 mSv y^{-1}, which is the mid-value of the range of 0.13–0.217 mSv y^{-1} (Sugiyama et al., 2007). The fatal cancer risk of K-40 at an effective dose of 1000 mSv was regarded as 8.86×10^{-2} (for all ages) (Preston et al., 2003; Oka, 2011), and the lethality fraction was 0.241 (ICRP, 2007). The annual risk of cancer was calculated by dividing the lifetime risk by a life expectancy of 70 y.

3. Results and discussion

3.1. Intake of I-131 by consumption of drinking water and foods, and effects of countermeasures

The average thyroid equivalent doses without countermeasures from 21 March 2011 to 20 March 2012 were estimated to be 0.42 mSv in adults, 1.49 mSv in children, and 2.08 mSv in infants (Fig. 3, Table S5). Those from 18 March to 20 March 2011 were estimated to be 0.13 mSv in adults, 0.48 mSv in children, and 0.71 mSv in infants, although these values were uncertain, as mentioned above. Those with countermeasures from 21 March 2011 to 20 March 2012 were 0.28, 0.97, and 1.14 mSv respectively, reductions of 33%, 35%, and 45%. The reduction for infants was largest because of the distribution of bottled water. The average doses with countermeasures from 21 March 2011 to 20 March 2012 were 1–2 orders of magnitude lower than the intervention level of 50 mSv y^{-1}. The dose was largest for infants, followed by children and adults.

The contribution from drinking water was larger than that from foods for adults and children, while that from milk and dairy products was also large for children and infants. The intake of I-131 by consumption of drinking water was smallest for adults, although the consumption of drinking water is the largest. This discrepancy is attributed to the thyroid ingestion dose coefficient, which depends on age.

The maximum thyroid equivalent doses without countermeasures from 21 March 2011 to 20 March 2012 were estimated to be 5.0 mSv in adults, 35 mSv in children, and 67 mSv in infants (Fig. 3, Table S6). Those from 18 March to 20 March 2011 were estimated to be 2.5 mSv in adults, 20 mSv in children, and 40 mSv in infants. Those with countermeasures from 21 March 2011 to 20 March 2012 were reduced to 1.3, 6.9, and 12 mSv respectively, reductions of 74%, 80%, and 83%. In particular, the reduction of intake by consumption of milk and dairy products was noticeable. The contribution from milk and dairy products was dominant among sources for infants and children. Although I-131 concentrations in milk and dairy products in Ibaraki were high (Fig. S1), restrictions on the distribution of raw milk in Ibaraki started from 23 March (Table S3). The dominant contribution of intake from milk and dairy products is attributable to this delay.

The cumulative thyroid equivalent dose for adults without countermeasures from 21 March 2011 was 63% of the total in week 1 and 86% in week 2 (Fig. 4). With countermeasures it was 70% and 87%, respectively. Similar trends were observed for children and infants (Fig. S5). This result highlights that the intake of I-131 was dominant within the first 2 weeks owing to the short half-life, and that rapid countermeasures are therefore important in reducing intake.

3.2. Evaluation of cancer risk of I-131 by consumption of drinking water and foods

The cancer risks of I-131 by consumption of drinking water and foods from 21 March 2011 to 20 March 2012 were 3×10^{-5} for infants, 2×10^{-5} for children, and 0.3×10^{-5} for adults, which were lower than those from the annual intake of ubiquitous K-40 and diesel exhaust particles (Table 1). The estimated fatal cancer risks

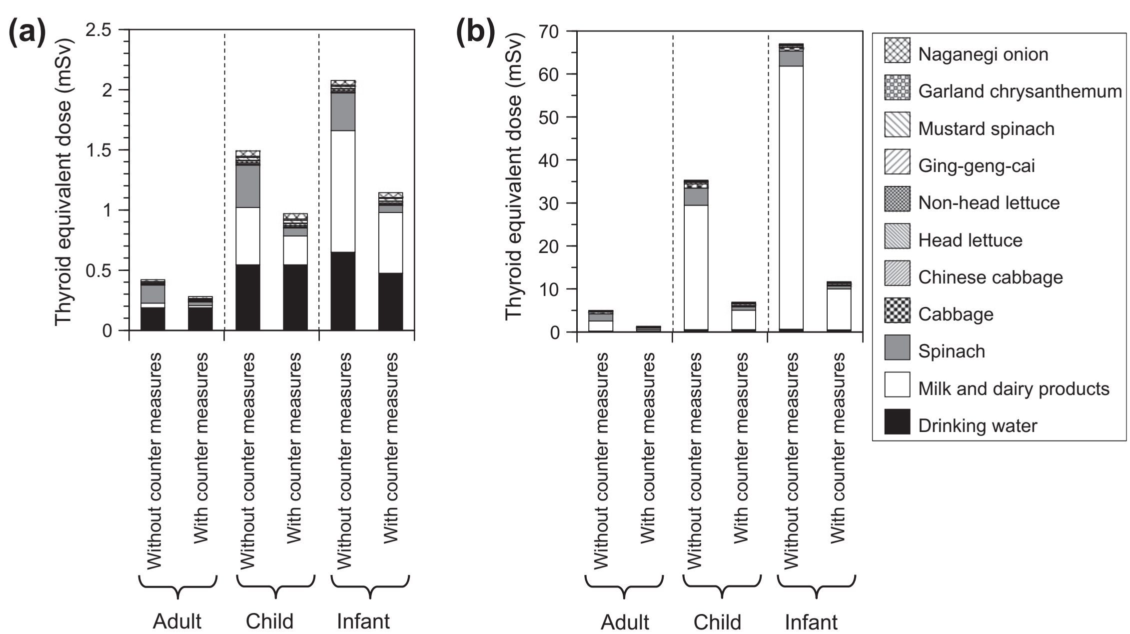

Fig. 3. Thyroid equivalent dose with and without countermeasures: (a) average intake; (b) maximum intake. Thyroid equivalent doses from 18 March to 20 March 2011 are shown in Tables S5 and S6.

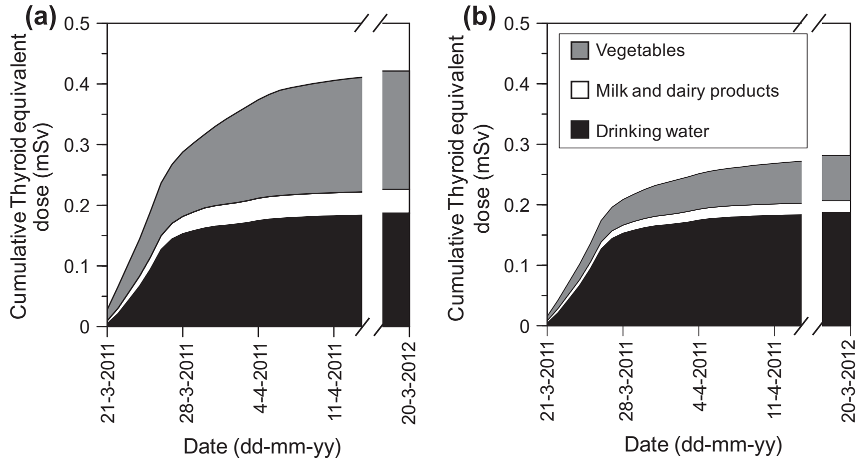

Fig. 4. Cumulative thyroid equivalent dose for adults (average intake): (a) without countermeasures; (b) with countermeasures.

Table 1
Estimated risks of cancer from I-131, K-40, and environmental pollutants in Japan.

Radioactive materials and environmental pollutants	Estimated cancer risks (10^5 y^{-1})	Reference
K-40	6.2	Preston et al. (2003), ICRP (2007), Sugiyama et al. (2007) and Oka (2011)
Diesel exhaust particles	4.5	Iwai and Utiyama (2000)
I-131 (infant)	**3**	**This study**
I-131 (child)	**2**	**This study**
Formaldehyde	1.3	USEPA (1991a) and Ministry of Health and Welfare (1997)
Arsenic	1.3	Mashiko (1989) and USEPA (1998a)
Dioxins	0.4	USEPA (1997) and Toyoda et al. (1999)
I-131 (adult)	**0.3**	**This study**
Benzene	0.05	Ministry of Health and Welfare (1999) and USEPA (2000)
DDTs	0.005	USEPA (1988a,b, 1991b) and Toyoda et al. (1998)
Chlordane	0.003	Gamo et al. (1995), Tani et al. (1996) and USEPA (1998b)

Table 2
Numbers of deaths per year in Japan and estimated fatal cancer risks.

Causes of deaths and radioactive materials	Number of deaths or estimated fatal cancer risks ((10^5 persons)$^{-1}$ y^{-1})	Reference
Number of deaths by diseases and accidents		
Cancer	250	Nakayachi (2006)
Smoking	80	Nakayachi (2006)
Passive smoking	5.3	Japan Preventive Association of Life-style related Disease (2010)
Traffic accident	4.5	National Police Agency (2011)
Drowning in a bath	2.6	Nakayachi (2006)
Murder	0.52	Nakayachi (2006)
Railroad crossing accident	0.09	Nakayachi (2006)
Estimated fatal cancer risks		
K-40	1.5	Preston et al. (2003), Sugiyama et al. (2007) and Oka (2011)
I-131 (infant)	**0.2**	**This study**
I-131 (child)	**0.1**	**This study**
I-131 (adult)	**0.02**	**This study**

were also compared with the number of annual deaths by diseases and accidents (Nakayachi, 2006; Japan Preventive Association of Life-style related Disease, 2010; National Police Agency, 2011) (Table 2). The fatal risks of I-131 were 0.2×10^{-5} for infants, 0.1×10^{-5} for children, and 0.02×10^{-5} for adults, approximately 1–2 orders of magnitude lower than deaths by traffic accident.

4. Conclusions

The intake of I-131 in drinking water and foods by citizens of Tokyo was estimated.

(1) The average thyroid equivalent doses without countermeasures (restrictions on the distribution of foods and the distribution of bottled water for infants) from 21 March 2011 to 20 March 2012 were estimated to be 0.42 mSv in adults, 1.49 mSv in children, and 2.08 mSv in infants. Those with countermeasures were 0.28, 0.97, and 1.14 mSv respectively. The countermeasures reduced the intake by 33% in adults, 35% in children, and 45% in infants.
(2) Drinking water contributed the most to intake of I-131 by adults and children. Milk and dairy products also contributed substantially in children and infants.

(3) The intake of I-131 within the first 2 weeks was more than 80% of annual intake, owing to the short half-life. Rapid countermeasures are important in reducing intake.
(4) The estimated average risks of cancer by ingestion of I-131 were 3×10^{-5} for infants, 2×10^{-5} for children, and 0.3×10^{-5} for adults, which were lower than the cancer risks from annual intakes of K-40 and diesel exhaust particles. The estimated average risks of fatality due to I-131 were 0.2×10^{-5} for infants, 0.1×10^{-5} for children, and 0.02×10^{-5} for adults, approximately 1–2 orders of magnitude lower than death by traffic accident.

Further studies on other areas, especially Fukushima Pref., are necessary. The intake of Cs-134 and Cs-137 will be investigated in a future study.

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Appendix A. Supplementary material

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.chemosphere.2012.02.028.

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