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Radiation dose rates now and in the future for residents neighboring restricted areas of the Fukushima Daiichi Nuclear Power Plant

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Radiation dose rates were evaluated in three areas neighboring a restricted area within a 20- to 50-km radius of the Fukushima Daiichi Nuclear Power Plant in August-September 2012 and projected to 2022 and 2062. Study participants wore personal dosimeters measuring external dose equivalents, almost entirely from deposited radionuclides (groundshine). External dose rate equivalents owing to the accident averaged 1.03, 2.75, and 1.66 mSv/y in the village of Kawauchi, the Tamano area of Soma, and the Haramachi area of Minamisoma, respectively. Internal dose rates estimated from dietary intake of radiocesium averaged 0.0058, 0.019, and 0.0088 mSv/y in Kawauchi, Tamano, and Haramachi, respectively. Dose rates from inhalation of resuspended radiocesium were lower than 0.001 mSv/y. In 2012, the average annual doses from radiocesium were close to the average background radiation exposure (2 mSv/y) in Japan. Accounting only for the physical decay of radiocesium, mean annual dose rates in 2022 were estimated as 0.31, 0.87, and 0.53 mSv/y in Kawauchi, Tamano, and Haramachi, respectively. The simple and conservative estimates are comparable with variations in the background dose, and unlikely to exceed the ordinary permissible dose rate (1 mSv/y) for the majority of the Fukushima population. Health risk assessment indicates that post-2012 doses will increase lifetime solid cancer, leukemia, and breast cancer incidences by 1.06%, 0.03% and 0.28% respectively, in Tamano. This assessment was derived from short-term observation with uncertainties and did not evaluate the firstyear dose and radioiodine exposure. Nevertheless, this estimate provides perspective on the long-term radiation exposure levels in the three regions.

Fukushima nuclear disaster | exposure assessment | Strontium-90 | forest contamination | food duplicate

The Fukushima Daiichi Nuclear Power Plant (FDNPP) underwent a series of hydrogen explosions in the days after a gigantic tsunami struck the northeast coast of Japan associated with the magnitude-9 Tohoku earthquake on March 11, 2011 (1). The March 12–15 explosions discharged radionuclides including rare gases, iodine, tellurium, and cesium into the atmosphere in the northern regions of Japan and also into the ocean by direct release and deposition from the atmosphere (2–5). A radioactive plume flowed away from the FDNPP in a west-to-northwest direction and was washed out by rain (6, 7). On April 22, 2011, the Japanese government designated a restricted area within 20 km of the FDNPP; even outside the restricted area, areas where exposure was predicted to exceed 20 mSv/y were designated as deliberate evacuation areas, and areas within 30 km of the FDNPP other than the restricted and deliberate evacuation areas were designated as evacuation-prepared areas in case of emergency (Fig. 1, *Left*).

Residents of nine municipalities in the Sousou region of the Fukushima prefecture (*ca.* 92,000 people) were evacuated following the FDNPP disaster (Table S1). The designation of evacuation-prepared areas in case of emergency was lifted on

Significance

There is a potential risk of human exposure to radiation owing to the March 2011 Fukushima Daiichi Nuclear Power Plant accident. In this study, we evaluated radiation dose rates from deposited radiocesium in three areas neighboring the restricted and evacuation areas in Fukushima. The mean annual radiation dose rate in 2012 associated with the accident was 0.89– 2.51 mSv/y. The mean dose rate estimates in 2022 are comparable with variations of the average 2 mSv/y background radiation exposure from natural radionuclides in Japan. Furthermore, the extra lifetime integrated dose after 2012 is estimated to elevate lifetime risk of cancer incidence by a factor of 1.03 to 1.05 at most, which is unlikely to be epidemiologically detectable.

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Fig. 1. Former and current statuses of the restricted area and areas to which evacuation orders have been issued around the FDNPP (*Left*, April 22, 2011; *Right*, August 8, 2013). Municipalities belonging to the Sousou region are bordered by thick lines. The three study areas are in blue.

September 30, 2011, because the power plant's reactors were by then considered to be stabilized. Thereafter, residents of several municipalities in the former evacuation-prepared area, including the village of Kawauchi and the town of Hirono, where radiation exposure was estimated to be $\leq 20 \text{ mSv/y}$, had subsequently made plans to return home in former evacuation-prepared areas in case of emergency (Table S1 and pink areas in Fig. 1, *Left*).

In April 2012, the Japanese government reexamined the evacuation areas in Fukushima and categorized them into three areas according to estimates of residents' annual radiation dose (8) (Fig. 1, *Right*). Evacuation orders are scheduled to be lifted in several areas in which estimated radiation exposure does not exceed 20 mSv/y (green areas in Fig. 1, *Right*).

Dietary, inhalation, and external exposures are considered major radiation exposure routes. In the early stages of radiation exposure following the FDNPP accident, short-lived radionuclides such as ¹³²Te and ¹³³Xe were observed (9, 10) and were noted as a major source of external exposure. The outdoor external dose rate within 80 km of the FDNPP decreased by 11% from June to November 2011 (i.e., less than 1 y after the accident) owing to the physical decay of ¹³⁴Cs (half-life of 2.07 y) and weathering (11); however, radiation exposure from deposited ¹³⁷Cs (half-life of 30.2 y) (i.e., groundshine) will persist long term. Regarding dietary exposure, agricultural products and seafood were contaminated with released radiocesium (12, 13). The Ministry of Health, Labor, and Welfare of Japan began food monitoring on March 16, 2011 and set provisional regulation values for contaminated food and water on March 17, 2011 (14). Except for the early stage of the accident, such food sources have been screened, and those containing more than 100 Bq/kg of radiocesium (current regulation value) have been

eliminated from circulation (14). Recent research, including our own previous study, has shown that doses from the ingestion of radiocesium 5–10 mo after the nuclear accident are considered to be less than those from natural radionuclides such as 40 K and 210 Po (15, 16). The inhalation of airborne dust comprising resuspended radioactive deposits represents an additional route of internal exposure (17, 18). For example, a survey in July 2011 showed that the radiocesium concentration in the air was on the order of 1 mBq/m³ outside restricted areas (19).

In this study, individual radiation dose rates were evaluated in three areas—the village of Kawauchi, the area of Tamano in the city of Soma, and the area of Haramachi in the city of Minamisoma—in the Sousou region in the Fukushima prefecture, over the 2-mo period, August–September 2012 (blue areas in Fig. 1, *Right*). The Kawauchi and Haramachi areas are located in former evacuation-prepared areas in case of emergency and the Tamano area has never been designated in any evacuation category. Whereas these areas neighbor the restricted areas and the deliberate-evacuation area (red and yellow areas in Fig. 1, *Left*), residents generally live as they did before the nuclear accident.

The personal dose equivalent (H_p) (10) was measured using optically stimulated luminescent dosimeters worn around the neck. The contribution of additional radioactivity from the FDNPP was estimated by subtracting a preaccident background value from the measured value. An evaluation of personal external doses is needed because individual behavior varies largely among residents. Concurrently, food-duplicate samples consisting of all meals consumed within a 24-h period were collected from residents and assayed for radiocesium, as in our previous study (15). Foodduplicate samples that contained relatively high levels of radiocesium were further subjected to radiostrontium analysis. In parallel, air sampling was conducted at three sites to evaluate radioactivity in whole dust and in the respirable fraction of dust in study areas. From the radiocesium concentration, the inhalation dose rate and outdoor external dose rate from radiocesium in aerosols (i.e., cloudshine) were estimated. From these observations, estimates of annual radiation doses after 10 and 50 y beyond 2012 were calculated using the Monte Carlo method and compared with background radiation doses in Japan. Finally, health risk assessment based on a linear nonthreshold (LNT) model was conducted for cancer risk from radiation exposure after 2012.

Results

Characteristics of the Study Participants and Results of Personal External Dose Monitoring. A total of 483 people living within a 20- to 50-km radius of the FDNPP were recruited for personal external dose monitoring in three study areas. More than half of the Kawauchi residents who had initially returned to their village in former evacuation-prepared areas in case of emergency (388 of 666, 58%) were enrolled. In Soma's Tamano area, the participants were recruited from 63 of 151 households (42%). In Minamisoma's Haramachi area, 30 people (30 of 13,755, 0.2%) were recruited through a local women's society. The ages and sexes of study participants are summarized in Table S2. Of the initial 483 study participants, 24 people (5.0%) dropped out of the survey because of complications from disease or medical procedures, dosimeter loss, or evacuation from the area. One male in the Tamano area was excluded from the study analysis because of a history of medical X-ray irradiation confirmed during interview.

The personal external dose equivalent H_p (10) from deposited radionuclides of the FDNPP accident in August–September 2012 is given in Table 1. The median, mean, and geometric mean (GM) of doses of participants within each of the three groups were similar. The maximum cumulative doses of H_p (10) were 1.2, 1.1, and 0.40 mSv/2 mo in Kawauchi, the Tamano area, and the Haramachi area, respectively.

In Kawauchi, residents' cumulative doses ranged from 0.04 to 1.2 mSv/2 mo, with a median of 0.15, showing a skewed distribution. Nineteen Kawauchi residents with a relatively high radiation dose were interviewed. Of these individuals, the person with the highest dose (1.2 mSv/2 mo) had transiently visited his former home, which is located in an area in which residents are not permitted to live. The person with the second-highest dose (0.81 mSv/2 mo) worked in a forest located in a restricted area, the town of Tomioka. Six individuals had engaged in decontamination efforts in the village or had worked in restricted areas, and four were other outdoor workers. Four individuals lived

Table 1. Personal dose equivalent for residents over a 2-mo period (August-September 2012)

| Area | Deep dose equivalent; 10 mm depth, mSv/2 mo* |
|---|---|
| Kawauchi village, <i>n</i> = 382 | |
| Range (median) | 0.04-1.2 (0.14) |
| Mean \pm SD | 0.17 ± 0.10 |
| GM (GSD) | 0.15 (1.6) |
| Tamano area of Soma city, $n = 50$ | |
| Range (median) | 0.17–1.1 (0.44) |
| Mean \pm SD | 0.46 ± 0.18 |
| GM (GSD) | 0.42 (1.5) |
| Haramachi area of Minamisoma city, $n = 27$ | |
| Range (median) | 0.14–0.40 (0.26) |
| Mean \pm SD | 0.28 ± 0.07 |
| GM (GSD) | 0.27 (1.3) |

*External dose rate from natural radiation was subtracted using literature data before the FDNPP accident (0.63 mSv/y in Kawauchi and Tamano; 0.61 mSv/y in Haramachi) (25, 26).

in a relatively highly contaminated area where the outdoor external dose rate was high. For the remaining three individuals with a relatively high dose, specific reasons or routes for this exposure were not identified.

Personal external doses were further compared within husband-and-wife pairs in Kawauchi (Fig. S14). The slope factor of linear regression was 0.987 (SE of 0.189), indicating that doses were in good accord for each couple. Three husbands deviated from the 95% upper limit of the predicted dose, and each of these men worked in areas with a high outdoor external dose rate, as described above.

Personal external dose rates were plotted against outdoor external dose rates monitored by the Japanese government around the residence of each participant (Fig. S1B). The slope factor was 0.4534 with a small intercept (0.0403 μ Sv/h), suggesting a shielding effect provided by buildings.

Characteristics of the Study Participants and Results of a Food-Duplicate Survey. The age, sex, physical size, and occupation of individuals participating in the food-duplicate survey are given in Table S2. A total of 131 people were recruited from the three study areas, of which 6 people dropped out because they missed part of the survey or because they were in poor physical condition following the Tohoku earthquake and their subsequent evacuation. The age of study participants (mean \pm SD) was 58.0 \pm 11.1 y. Participants' primary occupations at the time of the survey included office workers (35%), homemakers (21%), retirees (20%), and farmers (12%). The majority of survey participants residing in Kawauchi and the Tamano area consumed homegrown vegetables (56 of 79 participants and 12 of 16 participants, respectively).

The food materials in composite food-duplicate samples are listed in Tables S3 and S4. Food items were similar across each of the three groups, and there was no large deviation from the averages by weight in the age group (50–59 y old) published in the National Health and Nutrition Survey (20). Participants consumed various food products of the Fukushima prefecture origin, particularly rice and vegetables.

and vegetables. ¹³⁷Cs and ¹³⁴Cs were detected in more than half of the samples consumed in the three participant groups (Table 2). As the SDs of the dietary intake of radiocesium were larger than the corresponding means, levels of ¹³⁷Cs and ¹³⁴Cs did not distribute normally. The median of ¹³⁷Cs intake was 0.52, 0.86, and 0.63 Bq/d for participants in Kawauchi, the Tamano area, and the Haramachi area, respectively. Intake of ¹³⁴Cs had a similar trend to that of ¹³⁷Cs among the three study groups, whereas the amount was around 60% of ¹³⁷Cs. The maximum estimates of the committed effective dose rate of radiocesium were 59, 120, and 58 μ Sv/y for participants in Kawauchi, the Tamano area, and the Haramachi area, respectively, which did not exceed the standard limit for the dietary dose of radionuclides (1 mSv/y) (21).

Radiostrontium, ⁸⁹Sr, and ⁹⁰Sr, were analyzed in five foodduplicate samples in Kawauchi; these samples were selected in descending order starting with the sample with the highest ¹³⁷Cs level (0.8–3.1 Bq/kg). None of the samples contained a detectable amount of radiostrontium (⁸⁹Sr: <0.26 to <0.44 Bq/kg-wet; ⁹⁰Sr: <0.039 to <0.070 Bq/kg-wet). Ratios of ⁹⁰Sr to ¹³⁷Cs in the samples were <1.7% to <8.6%. A total of 77 participants in Kawauchi (n = 50) and the

A total of 77 participants in Kawauchi (n = 50) and the Haramachi area (n = 27) were evaluated with both the personal external dose survey and the food-duplicate survey. There was no association between external radiation doses and dietary intake of ¹³⁷Cs for residents of Kawauchi and the Haramachi area (Pearson's correlation, r = 0.234 and r = 0.152; and P = 0.10 and P = 0.45, respectively).

Radiocesium in Airborne Dust. Although the levels were sub-mBq/m³, fractionated dust samples contained discernible levels of radiocesium (Tables S5 and S6). Radiocesium contained in the respirable fraction (<4.9 μ m) accounted for 43–77% of total activities (<100 μ m). Total dust samples were also taken using

Table 2. Dietary intake of ¹³⁴Cs and ¹³⁷Cs

Estimates of committed

| | Dietary int | effective dose, µSv/y* | |
|-----------------------------------|----------------------|------------------------|---------------------------------------|
| Area | ¹³⁴ Cs | ¹³⁷ Cs | ¹³⁴ Cs + ¹³⁷ Cs |
| Kawauchi village | | | |
| n > LOD (%) | 28 (35) | 48 (61) | |
| Range (median) | <0.40 to 4.0 (<0.40) | <0.21 to 6.6 (0.52) | <2.3 to 59 (3.8) |
| Mean \pm SD | 0.38 ± 0.50 | 0.67 ± 0.86 | 5.8 ± 7.4 |
| GM (GSD) | 0.27 (2.0) | 0.45 (2.3) | 4.2 (2.1) |
| Tamano area of Soma city | | | |
| n > LOD (%) | 14 (88) | 14 (88) | |
| Range (median) | <0.31 to 8.7 (0.54) | <0.38 to 13 (0.86) | <3.8 to 120 (7.7) |
| Mean \pm SD | 1.3 ± 2.1 | 2.2 ± 3.3 | 19 ± 30 |
| GM (GSD) | 0.73 (2.7) | 1.11 (3.1) | 10.4 (2.9) |
| Haramachi area of Minamisoma city | | | |
| n > LOD (%) | 19 (63) | 21 (70) | |
| Range (median) | <0.21 to 3.9 (0.40) | <0.26 to 6.6 (0.63) | <2.7 to 58 (6.8) |
| Mean \pm SD | 0.60 ± 0.71 | 0.98 ± 1.26 | 8.8 ± 10.7 |
| GM (GSD) | 0.40 (2.4) | 0.58 (2.8) | 5.7 (2.5) |

*Committed effective doses by ingestion of radiocesium were calculated using effective dose coefficients (0.019 µSv/Bq for ¹³⁴Cs and 0.013 µSv/Bq for ¹³⁷Cs). It is assumed that the daily intake of radiocesium was constant over the course of a year.

a high-volume sampler at the same sites. The average radiocesium concentrations (sum of ¹³⁴Cs and ¹³⁷Cs) were 0.61 ± 0.43 , 0.60 ± 0.26 , and 0.92 ± 0.26 mBq/m³ in Kawauchi, the Tamano area, and the Haramachi area, respectively. The highest dose rates were estimated as $0.23 \ \mu$ Sv/y for adults and $0.27 \ \mu$ Sv/y for children (3–7 y old), but these values were 1,000-fold less than the ordinary permissible annual dose of 1 mSv/y (22). The outdoor external dose rate did not simply associate with the ¹³⁷Cs concentration in airborne dust (Pearson's r = -0.11; P = 0.622). Cloudshine from resuspended radiocesium had a magnitude of 100 pSv/h, which was negligible compared with groundshine (Table S6).

Total Radiation Dose Rates Estimated by Monte Carlo Simulation After 2012 and Comparison with the Background Radiation Dose. We excluded from the total radiation dose rate estimation by Monte Carlo simulation a total of eight external dose data for subjects in Kawauchi who had specific reasons for high exposure levels. The dose for a specific subgroup that enters an evacuated area or works in a contaminated area, which are not normal behaviors of residents, needs to be evaluated differently from the dose for the public. Nonetheless, analysis using all of the data gave an approximate mean dose rate as discussed below.

Cumulative probability was plotted against each radiation dose rate (Fig. 2). The external radiation dose rate dominated the other two measured exposure dose rates. The external dose rate and dose rate from the ingestion of radiocesium had a distribution with an S-like shape on a logarithmic scale.

We tested distributions for three variables: the personal external radiation dose rate, dose rate from the ingestion of radiocesium, and inhalation dose rate of radiocesium (Fig. S2). Variables were found to be log-normally distributed. Correlations among observed external radiation doses, radiocesium levels in food, and radiocesium levels in the air were not observed; we thus considered these levels to be independent, and thus generated values independently in a Monte Carlo simulation.

The mean annual radiation doses for the three exposure routes in 2012 were 0.89, 2.51, and 1.51 mSv/y (Table 3). These dose rates were similar to the mean personal external dose rate. When the physical decay of radiocesium was considered, a significant decrease in each dose rate was predicted until 2020 owing to physical decay of 134 Cs (Fig. 3). The annual dose in 2022 was one-third of the dose rate in 2012 and gradually decreased until 2062 (Table 3). The 99.9th percentile values of annual radiation doses in 2012, 2022, and 2062 were 3.16, 1.11, and 0.41 mSv/y; 7.82, 2.70, and 1.01 mSv/y; and 3.26, 1.14, and 0.42 mSv/y in Kawauchi, the Tamano area, and the Haramachi area, respectively (Table S7). When the eight outlying data of the personal external dose rate in the village of Kawauchi were included and fitted to the log-normal distribution, the geometric SD (GSD) increased but the mean dose was only slightly increased (Table 3 and Table S7). When



Fig. 2. Cumulative probability plot of external, ingestion, and inhalation radiation dose rates in 2012.

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a reduction factor for the long-term migration of radiocesium into the soil was incorporated into our calculations (23), external radiation dose rates after 50 y at 99.9% decreased to 0.16, 0.39, and 0.16 mSv/y in Kawauchi, the Tamano area, and the Haramachi area, respectively (Table S8).

Japanese people are generally exposed to a background radiation of 2.09 mSv/y (Table S9) (24). Nationwide surveys on terrestrial and cosmic radiation and radon concentration have shown an annual dose ranging from 0.72 to 1.24 mSv among 47 prefectures (25–28). Internal exposure to ²¹⁰Po and ²¹⁰Pb has been investigated in several cities in Japan and found to range from 0.15 to 0.81 mSv/y and from 0.03 to 0.16 mSv/y, respectively (29, 30). Dose rates for 210 Po and 210 Pb were bootstrapped into 47 prefectures with 1,000 replicates. The 5th and 95th percentiles of the total dose rate from these background radiations were calculated as 1.38 (1.32-1.46) mSv/y and 2.21 (1.87-2.32) mSv/y (mean [95% confidence interval (CI)]). The difference between 5th and 95th percentile dose rates from background radiation in Japan was also calculated as 0.85 (0.47-0.95) mSv/y, which is compared with the Monte Carlo distribution of the annual dose due to the FDNPP accident. In 2012, 45.7% (90.0-35.6%), 99.6% (100-99.1%), and 98.7% (100-96.1%) of the population in Kawauchi, the Tamano area, and the Haramachi area, respectively, were exposed to an annual dose of radiocesium larger than the variation in the background radiation dose rate. These proportions in Kawauchi, the Tamano area, and the Haramachi area are estimated to decrease to 0.4% (10.7–0.2%), 44.6% (91.0–34.3%), and 2.1% (60.9–0.7%) in 2022 and 0% (0–0.03%), 0.4% (12.7-0.2%), and 0% (0-0.01%) in 2062, respectively.

Health Risk Assessment for Cancer Incidence. Excess cancer incidence attributable to the lifetime radiation dose after 2012 without the reduction factor of radiocesium migration is presented in Table 4. Lifetime attributable risk (LAR) values for solid cancer incidence were 0.145-0.375%, 0.410-1.061%, and 0.246–0.638% in Kawauchi, the Tamano area, and the Haramachi area, respectively. In each study area, the LAR was higher for females and infants than for males and 20-y-old adults, respectively. In terms of leukemia incidence, LAR values were 0.004-0.012%, 0.012-0.033%, and 0.007-0.020% in Kawauchi, the Tamano area, and the Haramachi area, respectively, and values were higher for males and infants than for females and 20-y-old adults, respectively. LARs for breast cancer incidence in females were 0.038-0.100%, 0.109-0.284%, and 0.065-0.171% in Kawauchi, the Tamano area, and the Haramachi area, respectively, and infants had the highest LAR among age groups. Compared with the lifetime baseline risk (LBR) of solid cancer, leukemia, and breast cancer, the highest relative increased risks (LAR/LBR) were 3.3% in female infants, 4.6% in male infants, and 4.5% in female infants, respectively, in the Tamano area. When a reduction factor for the long-term migration of radiocesium was considered, the lifetime dose decreased by ca. 40% (Table S10). As a result, LARs of solid cancer, leukemia, and breast cancer decreased by 25-32%.

Table 3. Annual radiation doses 10 and 50 y after 2012 obtained using the Monte Carlo method

| | Year | | | | |
|------------------------|-------------------|------|-------------------|--|--|
| Mean annual dose, mSv* | 2012 | 2022 | 2062 | | |
| Kawauchi village | 0.89 | 0.31 | 0.12 | | |
| | 1.02 ⁺ | 0.35 | 0.13 [†] | | |
| Tamano area | 2.51 | 0.87 | 0.33 | | |
| Haramachi area | 1.51 | 0.53 | 0.2 | | |

*Total radiation doses were generated 10⁴ times for each study area from three exposure distributions. Only physical decay of radiocesium was considered to project the dose rates in 2022 and 2062. Variation in time of cesium in soil, food, and aerosols other than physical decay was not considered. [†]Eight sets of outlying data of the personal external dose rate in the village of Kawauchi were included and fitted to a log-normal distribution.



Fig. 3. Time course of total, ingestion, and inhalation radiation dose rates in the village of Kawauchi based on the physical decay of radiocesium. The 50th (solid line) and 90th percentile (dotted line) values were plotted. The total dose rate was calculated by Monte Carlo simulation. Ingestion and inhalation dose rates were derived from parameter estimates for a log-normal distribution.

Attributable risks over a 15-y period (AR₁₅) (2012–2026), were calculated for the radiation dose in the period without a reduction factor for radiocesium migration (Table S11). The estimates avoid uncertainties from extrapolation of current survival and cancer incidence in the long period ahead. AR₁₅ values for solid cancer incidence were 0.001–0.015%, and highest for 20-y-old adult females in the Tamano area. Leukemia AR₁₅ was 0.001–0.009%, and highest for 1-y-old infant males in the Tamano area. AR₁₅ for breast cancer incidence was highest (0.004%) for 20-y-old adult females in the Tamano area. Compared with the 15-y baseline risks of solid cancer, leukemia, and breast cancer, the highest relative increased risks (AR₁₅/BR₁₅) were 6.5% for female infants, 13.6% for male infants, and 27.5% for female children, respectively, in the Tamano area.

Discussion

Radiation doses for external, dietary, and inhalation exposures 1 y after the FDNPP accident were evaluated in residents living in three of the areas neighboring restricted and evacuation areas. External radiation doses constituted the major exposure across all three study areas. The mean of the dose rates in 2012 was greater than the ordinary permissible dose level of 1 mSv/y (31), particularly in the Tamano area, but was less than the permissible annual dose of 20 mSv/y during radiation emergencies (22). Simple and conservative estimates of dose rates 10 y after 2012 were less than the dose limit for the public (1 mSv/y) in most of the simulated percentiles given in Table S7. From the dose rate estimation after 2012, increased cancer incidence was calculated using the LNT model as in Table 4. The lifetime excess risk is small compared with the baseline risk of the Japanese population.

As discovered following nuclear weapon tests, dose rates decrease through the migration of radiocesium into the soil (32). Decontamination also reduced the external dose rate by 10–20% in the Chernobyl accident (33). Migration of radiocesium remains an ongoing process, and its reduction effect requires evaluation. To achieve the goal of reducing residents' annual dose to less than 1.0 mSv/y, decontamination around homes is critical and must be evaluated by continuous monitoring.

Personal external dose monitoring did not indicate considerable variation in Kawauchi residents' external exposure in usual daily behavior, except for workers engaged in decontamination. The correlation of external doses within husband-and-wife pairs also

| Table 4. | LAR of cancer incidence to an age of 89 y from 2012 in current study areas without consideration of a reduction factor for | or the |
|----------|--|--------|
| migratio | n of radiocesium | |

| | | All solid cancers | | | Leukemia | | | Breast cancer | |
|-------------------|-----------|---------------------------|-------|--------|---------------------------|-------|--------|---------------------------|-----------|
| Age in 2012, y | | | LAI | R, % | | L۵ | .R, % | | LAR, % |
| | Area | Lifetime exposure, mSv | Male | Female | Lifetime exposure, mSv | Male | Female | Lifetime exposure, mSv | Female |
| 1 | Kawauchi | 15.1 | 0.251 | 0.375 | 15.5 | 0.012 | 0.008 | 17.2 | 0.100 |
| | Tamano | 42.8 | 0.712 | 1.061 | 43.8 | 0.033 | 0.022 | 48.5 | 0.284 |
| | Haramachi | 25.7 | 0.428 | 0.638 | 26.3 | 0.020 | 0.013 | 29.2 | 0.171 |
| | LBR | | 43.92 | 31.76 | | 0.71 | 0.51 | | 6.29 |
| 10 | Kawauchi | 14.6 | 0.197 | 0.289 | 14.9 | 0.008 | 0.005 | 16.8 | 0.066 |
| | Tamano | 41.4 | 0.556 | 0.819 | 42.1 | 0.024 | 0.015 | 47.4 | 0.186 |
| | Haramachi | 24.9 | 0.335 | 0.492 | 25.3 | 0.014 | 0.009 | 28.5 | 0.112 |
| | LBR | | 43.96 | 31.76 | | 0.67 | 0.48 | | 6.28 |
| 20 | Kawauchi | 13.9 | 0.145 | 0.209 | 13.9 | 0.006 | 0.004 | 15.9 | 0.038 |
| | Tamano | 39.3 | 0.410 | 0.590 | 39.2 | 0.018 | 0.012 | 45.0 | 0.109 |
| | Haramachi | 23.6 | 0.246 | 0.355 | 23.6 | 0.011 | 0.007 | 27.1 | 0.065 |
| | LBR | | 44.02 | 31.75 | | 0.63 | 0.45 | | 6.29 |

Lifetime exposure doses were calculated using means for corresponding areas in 2012 with only physical decay of radiocesium (Table 3).

suggested that exposure to radiation in a home setting, as opposed to radiation exposure occurring through specific behaviors outside the home, constitutes the majority of daily doses. In Soma's Tamano area and Minamisoma's Haramachi area, annual doses ranged from 1.1 to 6.6 mSv/y and from 0.84 to 2.4 mSv/y, respectively. Decontamination is ongoing in these two areas, and its effects on personal doses need to be evaluated further.

its effects on personal doses need to be evaluated further. Dietary exposure to 134 Cs and 137 Cs was observed in food-duplicate samples in the three areas (median: <0.40 to 0.54 Bq/d for 134 Cs and 0.52 to 0.86 Bq/d for 137 Cs), whereas the estimated annual committed effective doses did not exceed the standard limit of Japan (1 mSv/y) (21). The maximum of estimated annual doses was 120 µSv/y in the Tamano area. Nevertheless, these radiation levels were much lower than external radiation dose rates. Participants' residences neighbor the restricted and evacuation areas and participants consume homegrown vegetables; however, the doses were comparable to those in our previous research, which examined food-duplicate samples from relatively urban areas in Fukushima (15, 19). Local food supply in farming areas did not necessarily increase exposure to radiocesium because local municipalities routinely screen food products for personal consumption. Release of ⁹⁰Sr and other less volatile radionuclides from the FDNPP meltdown was considered to be limited compared with that of ¹³⁷Cs (34). A survey showed that the concentration of 90 Sr in soil samples was quite low (ca. 1 Bq/g at most) even near the power plant (35), and the contribution of radiostrontium to internal exposure is likely to be small, as observed in this study.

In areas within a 20- to 50-km radius of the FDNPP where outdoor external dose rates were relatively low, as in the village of Kawauchi, people have been gradually returning to their homes. In our study areas, the average outdoor external dose rate in 2012 ranged from 0.24 to 0.55 µSv/h (Table S1). The Japanese government has indicated that evacuation orders are ready to be lifted in restricted and evacuation areas where the outdoor external dose rate is less than 3.8 µSv/h. Assuming that a major component of radiation exposure is the external radiation dose, the radiation dose rate in such areas is likely to increase proportionally to the outdoor external dose rate according to the estimates made for the current study areas. In the Fukushima prefecture, the outdoor external dose rates in current habitable areas are relatively high in the Sousou, Kenpoku, and Kenchu regions (mean values of 0.22, 0.29 and 0.24 μ Sv/h, respectively), followed by the Kennan region $(0.18 \ \mu \text{Sv/h})$ (Table S1). In the western part of Fukushima (Aizu and Minamiaizu regions), the outdoor external dose rates are around 0.1 µSv/h. Using the relationship between the personal

external dose rate and outdoor external dose rate for this study population (Fig. S1*B*), the personal dose rates from groundshine at other municipalities in the Fukushima prefecture were estimated (Table S1). The median estimated dose rate ranged from 0.177 μ Sv/h to less than 0.116 μ Sv/h. The values were less than those measured in the Tamano area. Therefore, the majority of the residents in Fukushima are considered to have less radiation exposure than the participants in the current study. Outside Fukushima, the US Navy's external dosimetry program revealed that the effect of the accident on control dosimetry at Yokosuka (300 km to the south of the FDNPP) was less than the detection limit (0.375 μ Sv/d) (36). Even though the magnitude of contamination from radionuclides was not regionally uniform (37), released radiocesium was deposited mainly in the eastern part of Japan around the FDNPP and the Pacific Ocean (2).

The United Nations Scientific Committee on the Effects of Atomic Radiation reported that public exposure to natural radiation averages 2.4 mSv/y, with a typical range of 1.0–13 mSv/y due to large variations in radon exposure (38). As stated above, the general population in Japan is annually exposed to a background radiation of 2.09 mSv/y (Table S9). Internal exposure to fallout from nuclear tests is currently very low (0.0025 mSv/y). In our study areas, the mean additional dose rate after the FDNPP accident was estimated as 0.89–2.51 mSv/y in 2012 (Table 3), which is comparable with, or in places slightly higher than, the background radiation in Japan. However, 10 y later, in 2022, the additional dose rate will have decreased to 0.31–0.87 mSv/y, which is comparable with geographical variations in background radiation exposures (1.38–2.21 mSv/y, Table S9).

Estimated dose rates in this study have several uncertainties. First, dietary exposure surveys over the course of a single day cannot evaluate day-to-day variation in individuals. It is possible that a relatively high intake of ¹³⁷Cs in this study period may be occasional or, on the other hand, the single-day intake of heavily contaminated products in another period may surpass usual dietary intakes. However, recent research into whole-body counting in Fukushima showed that the ¹³⁷Cs body burden was below 300 Bq per body for 99% of subjects after March 2012, which corresponds to a dietary intake of 2.4 Bq/d of ¹³⁷Cs for an adult (39). Although those study areas were different from those examined in our study, the magnitude of exposure appears comparable. The contribution of ¹³⁷Cs fratio. Ratios of ¹³⁴Cs:¹³⁷Cs activities in food products and airborne radionuclides immediately following the FDNPP meltdown were reported to be 0.98 and 1

on March 11, 2011, respectively (13, 40). Because the corrected ratio of 134 Cs: 137 Cs in food-duplicate samples was 1.1 on March 11, 2011, detected radiocesium was mainly derived from the FDNPP accident.

Second, the estimation of inhalation exposure to radiocesium is dependent on subjects' daily inhalation volume, the chemical form of radiocesium, and the aerodynamic diameter of dust samples. In this study, the breathing rate recommended by the US Environmental Protection Agency (EPA) (15.7 m³/d) was used, whereas the International Commission on Radiological Protection reference manual gives a larger rate (22.9 m³/d for males and 18.5 m³/d for females), and Japanese adults are typically smaller physically than the individuals described by the EPA standards (41). The effective dose coefficient for inhalation of radiocesium was conservatively assumed as type slow (S); i.e., radiocesium absorption from dust is slow and persists in the lungs (42). However, radiocesium was distributed in the aerosol fraction larger than 1.1 µm, where the effective dose coefficient is lower. Among these uncertainties, the absorption type is critical, varying from 0.0046 µSv/Bq (type Fast) to 0.039 µSv/Bq (type S) for 137 Cs (42). Considering the above, inhalation dose rates of radiocesium are not underestimated in this study and are far less than the dose rates for the other two exposure routes.

Third, regarding external exposure, this survey was conducted only in the summer season. In the estimation of the annual dose, we assumed that the summer values are valid throughout the year, but practically, the seasonal variation should be considered. Snow accumulation is usually observed in the Fukushima prefecture in winter and it shields against the external radiation dose from terrestrial radiation by 5-20% at a water-equivalent thickness of 30 cm (43). Monitoring data of outdoor external dose rates in February 2012 in Fukushima were reported to be 8.3% less than those in May 2012 (44). People also likely stayed indoors more during this period than in summer. Among the three areas selected for this study, participants living in Minamisoma's Haramachi area were recruited from a limited population, and the external radiation doses of these recruits are not appropriately representative of the doses of adults in that area.

In addition to the uncertainties above, the external dose from background radiation was subtracted using literature data from before the FDNPP accident (25, 26). In the Sousou region in the Fukushima prefecture, there are several areas where granite contents are high, although the current study areas are not included therein (45) and, therefore, the external dose rate from deposited radiocesium in 2012 is likely to be largely unaffected. On the other hand, the external dose rate from natural radiation is often higher in indoor environments than outdoor, owing to building materials (46). Hence, personal external dose rates from deposited radiocesium might be slightly overestimated.

Finally, residents were thought to live as they did before the accidents without restriction. However, they were found to be strictly restricting activities in the neighboring forest, in which contaminations of trees with radiocesium have been reported (47). In fact, we found several residents with high levels of external radiation doses, who worked in the forest. Thus, current estimations underestimate the external radiation dose rates for residents when they work in the forest routinely. Because the neighboring forest is a major economic resource, rigorous external radiation monitoring is needed for residents who routinely work in the forest.

Health risk assessment demonstrated that solid cancer incidence increased by 1.061% at most in the Tamano area after 2012 owing to the radiation dose. Apparently, this assessment did not assess the first-year dose immediately after the accident and especially the radioiodine dose for thyroid tissue. In addition, the dose rates for infants and children were assumed to be the same as those observed for adults. In addition, input data of mortality and cancer incidence in Japan may change over decades. In particular, breast cancer incidence in Japan has been increasing significantly over the past few decades (48). In this assessment, breast cancer risk was calculated using an excess absolute risk (EAR) model of 1958–1998 (49), which may underestimate the risk in future settings.

In terms of dose allocations in a recent risk assessment conducted by the World Health Organization (WHO) (50), the lifetime doses were assumed to be twice as large as the doses in 2011. In this study, however, lifetime dose rates were integrated based on the dose rates in 2012. In addition, the current lifetime risks do not include the exposure in 2011. These two differences in exposure dose allocation may lower cancer risks per unit lifetime dose; e.g., for solid cancer incidence in the 1-y-old female infant, they were 0.025% per mSv in this study (Table 4) and 0.035–0.041% per mSv in the WHO report (50).

In our study population, the average radiation dose rate is predicted to be comparable to the variation in background radiation in a decade. The baseline risk of solid cancer in Japanese is 31.7–44.0% (Table 4), resulting from carcinogens other than radiation or a stochastic process. These factors need a large population to obtain robust statistical power. A previous epidemiological study with a large population and long follow-up period could not detect a statistically significant increase in cancer risk at a dose less than 100 mSv (51). Indeed, the largest increase of 1.061% in solid cancer incidence corresponds to a relative risk of 1.03 for females exposed from the age of 1 y, which is likely difficult to detect from other risk factors unless a large population cohort is followed up.

In conclusion, food supply and associated regulations are considered effective in the study areas in Fukushima thus far, and external exposure is a major component of the radiation dose rate. However, these levels can be easily elevated when residents preferentially take contaminated mushrooms and wild boar meats from the field, as in the case of the Chernobyl accident (33). Our study found that in most of the populations in the study areas, annual radiation doses 10 y after 2012 onward are unlikely to exceed 1 mSv. Doses could also reduce through the migration of radiocesium into the soil and/or decontamination. According to the dose rate after 2012, a detectable increase in cancer risk is unlikely. This estimate provides perspective on the long-term radiation exposure levels in these areas.

Materials and Methods

Study Population and Dose Evaluation. Participant recruitment was conducted in June–July 2012. The Ethics Committee of the Kyoto University Graduate School of Medicine approved this study protocol. All of the research participants and the parents of minor children participating in the survey submitted written informed consent.

Participants were recruited from three areas in the Sousou region of the Fukushima prefecture: the village of Kawauchi, the area of Tamano in the city of Soma, and the area of Haramachi in the city of Minamisoma (Fig. 1, *Right*). Among the three areas neighboring restricted and evacuation areas, outdoor external dose rates were apparently different (Table S1). Outdoor external dose rates were usually monitored in residential regions in each area. In the Kawauchi and Haramachi areas, populous regions are apart from contaminated areas whereas the Tamano area is located close to the relatively contaminated areas of the village of litate.

Kawauchi residents evacuated after the FDNPP accident began to return home in former areas that were evacuation prepared in the case of emergency in April 2012. The majority of the restricted areas in this region have been reexamined by government authorities to determine areas for which evacuation orders are ready to be lifted. A total of 666 residents initially indicated to the village office that they intended to return. They were invited to participate in this study via postal mail.

The Tamano area is a somewhat rural area to the north of the evacuated areas, and the outdoor external dose rate was relatively high in this city (mean of 0.55 μ Sv/h, Table S1). A representative of the residents' association in the Tamano area invited all participants to join the study, as he has routinely done for other local activities.

The Haramachi area is an urban area, and the outdoor external dose rate in residential areas was low within the city itself (mean of $0.26 \,\mu$ Sv/h, Table S1), because the western part of the city is forested and thinly populated. Participants were invited to enroll with the help of members of a local women's society in the Haramachi area.

All study participants were asked to wear a small, optically stimulated luminescent dosimeter on a neck strap for 2 mo (August–September 2012). In

Kawauchi, those individuals who lived in the village less than 5 d/wk were not eligible to participate in our study's personal external dose monitoring.

One participant from each household was invited to participate in the 24-h food-duplicate survey at the same time as the personal external dose monitoring survey, except in Kawauchi. There, public health nurses invited residents to participate in the survey, as part of their standard health service activities (health guidance, health consultation, and so on). The 24-h food-duplicate samples were analyzed for radiocesium and, in part, radiostrontium.

Evaluation of Personal External Doses. The cumulative doses of H_p (10) in participants from August 1 to September 30, 2012, were evaluated. Optically stimulated luminescence personal dosimeters of aluminum oxide were used (Quixel badge system type S; Nagase Landauer, Ltd.). Dosimeters were calibrated following Japanese Industrial Standards JIS Z 4339 (52). The dosimeters were worn on a neck strap. A radiation dose was detected at above 0.01 mSv. Participants recorded their representative daily behavior, and participants whose radiation dose was relatively high were interviewed. The annual radiation dose was calculated by multiplying the dose measured over the course of 2 mo by the number 6.

The external radiation dose from natural radiation was subtracted using literature data for background radiation in Japan (25, 26). Before the FDNPP accident, terrestrial radiation from the ground was comparable between the Kyoto and Fukushima prefectures (0.34 and 0.35 mSv/y, respectively) (25). Ionizing cosmic rays had levels of 0.28 mSv/y in Kawauchi, 0.26 mSv/y in Haramachi, and 0.25 mSv/y in Kyoto (no data for Tamano) (26). Kawauchi and Tamano are located in a mountain-ringed region ~400 m above sea level. The control measurement in Kyoto was 0.10 mSv/2 mo and was in fair agreement with the sum of terrestrial and cosmic radiation. Therefore, background radiation estimates (0.63 mSv/y in Kawauchi and Tamano; 0.61 mSv/y in Haramachi) were subtracted from the values obtained by the dosimeter.

Food Sampling and Preparation. The food-duplicate samples were collected as previously described (15, 53). Participants prepared an additional portion of all meals and beverages consumed in the 24 h of a single day and provided our research team with a list of what they consumed. Those people who avoided eating any food products from the Fukushima region were excluded from this study. From August 7 to 11, 2012, food-duplicate samples were collected from 79 people living in the village of Kawauchi. In the Tamano and Haramachi areas, a food-duplicate survey was conducted between August 27 and 31, 2012, for 16 and 30 individuals, respectively. The homogenates of food-duplicate samples, including drinks and sweets, were lyophilized.

Registered dietitians recorded the weight of food items in the samples and categorized these items into 18 categories on a food composition table. The amount of food of Fukushima origin was measured using the menu records of places of production. A questionnaire regarding eating habits and preferences was also administered.

Airborne Dust Sampling. Airborne dust samples were collected at three sites in the study areas between August 6 and October 30, 2012. Air sampling was conducted using a high-volume sampler (HV-1000F; Sibata). Whole airborne dust in the air was collected on a quartz membrane filter at a height of 1.5 m from the ground. In the period from August 6 to 12, 2012, more than 80 m³ of air were inspired at a rate of 1,000 L/min within a day. To collect a sufficient volume of dust, air samplers continued running for a week after the end of sample collection on August 12. An Andersen cascade impactor sampler (AN-200; Tokyo Dylec Co.) connected with a low-volume sampler (flow rate of 28.2 L/min) (SL-30; Sibata) was used to collect dust samples with different aerodynamic diameters. Airborne dust was fractionated onto nine filters, ranging from 100 to 11.4 μ m to <0.46 μ m. Filters corresponding to fractions <100, <4.9 (considered the respirable fraction), and <1.1 μ m were combined and subjected to radiometry.

Determination of ¹³⁷Cs and ¹³⁴Cs. Lyophilized food-duplicate samples (200–300 g) were placed in cylindrical polyethylene containers. Quartz fiber filters from dust sampling were pressed into thin polyethylene containers. A high-resolution Ge semiconductor detector was used for determination of radiocesium. Characteristic photon energies of radiocesium were monitored (¹³⁴Cs: 604.7 and 795.9 keV; ¹³⁷Cs: 661.7 keV). The intensity ratios of 604.7 keV:661.7 keV were plotted against distances between the detector and sample. A summing effect was observed and corrected for ¹³⁴Cs. Food-duplicate samples and dust samples were measured for >20,000 and >10,000 s, respectively. The limits of detection (LODs) were determined using Kaiser's method (54). Radioactivities were corrected to the sampling date. Every 20 measurements, procedural blanks were prepared to check contamination by equipment and handling. No detectable contamination was observed in the procedural blanks (*n* = 6).

Determination of ⁸⁹Sr and ⁹⁰Sr in Food-Duplicate Samples. ⁸⁹Sr and ⁹⁰Sr measurements were conducted by radiochemical analysis (55). The freezedried food samples (75–126 g) were ashed at 450 °C for 24 h. Ashed samples were decomposed with aqua regia and an additional amount of nitric acid, and then dissolved in hydrochloric acid. Strontium was precipitated and separated by fuming nitric acid. The activity of ⁸⁹Sr was immediately measured with a Cherenkov light and liquid scintillation counter (LSC-6101B; Hitachi-Aloka Medical). After attaining a radioactive equilibrium of ⁹⁰Sr and ⁹⁰Y for 2 wk, the activity of ⁸⁹Sr. The LODs were 0.25 and 0.04 Bq for ⁸⁹Sr and ⁹⁰Sr, respectively. Activities were corrected to the sampling date, using the physical half-lives of radiostrontium (⁸⁹Sr: 50.5 d; ⁹⁰Sr: 28.9 y).

Estimation of Radiation Dose Levels and Monte Carlo Simulation over the Next 10-50 v. Committed effective doses for ingestion of radiocesium were calculated using effective dose coefficients (0.019 μ Sv/Bq for ¹³⁴Cs and 0.013 μ Sv/Bq for ¹³⁷Cs) (42). For inhalation exposure, it was assumed that adult (51- to 60-y-old males and females) and child (3- to 5-y-old males and females) residents inhaled 15.7 and 10.1 m³ air per d, respectively (56). The effective dose coefficients of 0.020 $\mu\text{Sv/Bq}$ for ^{134}Cs and 0.039 $\mu\text{Sv/Bq}$ for ^{137}Cs in the case of inhalation (absorption type S and aerosol size of 1 μ m) by adults and 0.041 μ Sv/Bq for ¹³⁴Cs and 0.070 μ Sv/Bq for ¹³⁷Cs in the case of inhalation by children (3–7 y) were used to calculate committed effective doses (42). To estimate the inhalation exposure doses, it was assumed that all ¹³⁴Cs and ¹³⁷Cs in airborne dust samples collected using the high-volume air sampler were respirable; we considered this to be an appropriate hypothesis because 43-77% of radiocesium was collected in the respirable fraction in this study. The estimates of the annual committed effective dose were made under the assumption that the daily intake and inhalation of radiocesium was approximately constant over the course of a year. Cloudshine was estimated using the dose rate coefficient for a semiinfinite volume source in air to obtain effective doses for adults [0.26 nSv/(Bq/m³)/h for $^{134}\mbox{Cs}$ and 0.093 nSv/(Bq/m³)/h for ¹³⁷Cs] (57). We then estimated the probability distribution of the observed personal external dose rates, observed radiation dose rates from food, and observed radiation dose rates from the air, as obtained in the three study areas. We further tested for associations among the external radiation dose rates, radiation dose rates from food, and radiation dose rates from the air. When those values did not correlate to one another (P > 0.05), we considered them to be independent. If we could discard the null hypothesis of uncorrelatedness at P = 0.05, we considered the data to be correlated.

Next, those data were tested to determine whether their distribution can be assumed to be log-normal using the Kolmogorov–Smirnov–Lilliefors test. If the test showed P > 0.05, we assumed that values were log-normally distributed and estimated GMs and GSDs. The external dose rates, radiation dose rates from food, and radiation dose rates from the air were simulated using the Monte Carlo method using estimated GMs and GSDs. If dose rates were correlated, we generated correlated values by taking into consideration their associations. The total radiation dose rate for an adult subject was calculated by adding the generated values of external dose rates, radiation dose rates from food, and radiation dose rates from the air. Total radiation dose rates were generated 10^4 times for each study area. Annual doses 10 and 50 y after August 2012 were estimated by assuming the physical decay of radiocesium, with or without reduction factor r(t) for long-term migration of radiocesium into the soil (23):

$$r(t) = 0.34 \times e^{-0.693 \times \frac{t}{1.5}} + 0.66 \times e^{-0.693 \times \frac{t}{50}}.$$

The variation in time of radiocesium in food and aerosols other than physical decay was not considered.

The external dose rate was divided into components attributable to ^{134}Cs and ^{137}Cs , assuming that the external dose coefficients per unit deposit are 2.7×10^{-8} and 1.2×10^{-8} (Sv/y)/(Bq/m²) for ^{134}Cs and ^{137}Cs , respectively (23), and the ratio of ^{134}Cs is 1 on March 11, 2011 (13, 40).

To compare dose rate with background radiation in Japan, reference values were cited from the literatures (24–30). Regional dose rates of external exposure and radon inhalation were estimated from means among 47 prefectures and 7 regions in Japan (25–28). Variation in internal exposure to ²¹⁰Po and ²¹⁰Pb was calculated by bootstrapping the dose rate estimates among several cities in Japan into 47 prefectures (1,000 iterations) (29, 30). In each bootstrap dataset, the 5th and 95th percentile dose rates were calculated and their means were presented as a range of the total dose rate from background radiation. Other exposures were assumed to be constant across Japan.

Statistical Analysis. If analytical values were less than the LODs, they were converted to one-half of the LODs. Summary statistics (mean, range, and GM) were calculated for the radiation doses. Pearson's *r* coefficient was used to evaluate

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correlation. *P* values less than 0.05 were considered as statistically significant differences. JMP (Version 4; SAS Institute Inc.) was used for these analyses.

Health Risk Assessment of Cancer Incidence from Radiation Dose Associated with the FDNPP Accident. A health risk assessment was conducted according to a framework used in a recent report by the WHO (50).

Mean dose rate estimated by Monte Carlo simulation was used to calculate LAR with or without a reduction factor for the long-term migration of radiocesium. The effective dose rates from external exposure and the dietary intake of radiocesium of a 1-y-old infant and 10-y-old child were assumed to be the same as those for an adult. The organ (colon, breast, and bone marrow) dose rate was calculated using organ dose-toeffective dose ratios for a 1-y-old infant, 10-y-old child, and 20-y-old adult in the three study areas (57). Whereas the ratios were calculated for an 8wk old and 7-y old, we used the values for a 1-y-old infant and 10-y-old child, respectively. For the child and infant, the skin dose was regarded as the dose for breast tissue. The lifetime organ dose was estimated from the dose rate from August 2012 to an age of 89 for the three age groups, assuming that the dose rate in 2012 decreases only by physical decay unless otherwise indicated. Infants and children were defined as having age ranges of 1-3 y and 4-15 y, respectively. The cancer-free survival rates of males and females were calculated from age- and sex-stratified allcause mortality in Japan in 2010, age- and sex-stratified all-cancer mortality (International Classification of Diseases 10 codes: C00-C96) in Japan in 2010 (58), and age- and sex-stratified all-cancer incidence (C00-C96) in Japan in 2008 (48). LBR of solid cancer (C00-C89), leukemia (C91-95), and breast cancer (C50) was calculated by integrating the product of cancer incidence probability and the cancer-free survival rate at each age from the age in 2012 to an age of 89 y. LAR from the annual dose (D[Sv]) at each age of exposure (e) for each sex (g) was calculated as

$$\begin{aligned} \mathsf{LAR}(D,e,g) &= \int_{e+L}^{89} \left[w \cdot \mathsf{EAR}(D,e,a,g) + (1-w) \cdot \mathsf{ERR}(D,e,a,g) \cdot m(a,g) \right] \frac{\mathsf{S}(a,g)}{\mathsf{S}(e,g)} da, \end{aligned}$$

where m(a,g) is the cancer incidence probability in the unexposed population and S(a,g) is the cancer-free survival rate. The minimum latency period, L, was set at 2 y for leukemia and 5 y for breast cancer and solid cancer.

The weights of an EAR model and an excess relative risk (ERR) model for solid cancer and leukemia were equal (w = 0.5) and EAR was considered for breast cancer (w = 1). The dose and dose rate effectiveness factor was taken as 1.

- 1. Tanaka S (2012) Accident at the Fukushima Dai-ichi nuclear power stations of TEPCO outline & lessons learned. Proc Jpn Acad, Ser B, Phys Biol Sci 88(9):471-484.
- 2. Buesseler K, Aoyama M, Fukasawa M (2011) Impacts of the Fukushima nuclear power plants on marine radioactivity. *Environ Sci Technol* 45(23):9931–9935.
- Yasunari TJ, et al. (2011) Cesium-137 deposition and contamination of Japanese soils due to the Fukushima nuclear accident. Proc Natl Acad Sci USA 108(49):19530–19534.
- 4. Povinec PP, et al. (2013) Dispersion of Fukushima radionuclides in the global atmosphere and the ocean. *Appl Radiat Isot* 81:383–392.
- Stohl A, et al. (2012) Xenon-133 and caesium-137 releases into the atmosphere from the Fukushima Dai-ichi nuclear power plant: Determination of the source term, atmospheric dispersion, and deposition. Atmos Chem Phys J 12(5):2313–2343.
- Chino M, et al. (2011) Preliminary estimation of release amounts of ¹³¹I and ¹³⁷Cs accidentally discharged from the Fukushima Daiichi nuclear power plant into the atmosphere. J Nucl Sci Technol 48(7):1129–1134.
- 7. Sugiyama G, et al. (2012) Atmospheric dispersion modeling: Challenges of the Fukushima Daiichi response. *Health Phys* 102(5):493–508.
- Nuclear Emergency Response Headquarters (2011) [The basic concepts and agenda for reviewing restricted area and evacuation area following the completion of Step 2] (Cabinet Office, Government of Japan, Tokyo). Available at www.meti.go.jp/earthquake/ nuclear/pdf/111226_01a.pdf. Accessed February 5, 2014. Japanese.
- 9. Hosoda M, et al. (2011) The time variation of dose rate artificially increased by the Fukushima nuclear crisis. *Sci Rep* 1:87.
- Petit G, et al. (2012) Analysis of Radionuclide Releases from the Fukushima Dai-Ichi Nuclear Power Plant Accident Part I. Pure Appl Geophys, 10.1007/s00024-012-0581-6.
- Ministry of Education Culture, Sports, Science, and Technology, Japan (December 16, 2011) Results of the Fourth Airborne Monitoring Survey by MEXT. Available at http://radioactivity. nsr.go.jp/en/contents/4000/3179/24/1270_1216.pdf.
- Merz S, Steinhauser G, Hamada N (2013) Anthropogenic radionuclides in Japanese food: Environmental and legal implications. *Environ Sci Technol* 47(3):1248–1256.
- Buesseler KO, et al. (2012) Fukushima-derived radionuclides in the ocean and biota off Japan. Proc Natl Acad Sci USA 109(16):5984–5988.
- Hamada N, Ogino H, Fujimichi Y (2012) Safety regulations of food and water implemented in the first year following the Fukushima nuclear accident. J Radiat Res (Tokyo) 53(5):641–671.

Risk models for solid cancer (49) are

$$\mathsf{ERR}(D,e,a,g) = \beta_1 \cdot D \cdot \exp[\tau \cdot (e-30) + \upsilon \cdot \ln(a/70)](1+\sigma g),$$

where β_1 = 0.4666, τ = –0.01849, υ = –1.621, σ = 0.2465, and g = –1 for male and +1 for female;

$$\mathsf{EAR}(D, e, a, g) = \beta_1 \cdot D \cdot \exp[\tau \cdot (e - 30) + \upsilon \cdot \ln(a/70)](1 + \sigma g)$$

where β_1 = 0.005163, τ = –0.02805, υ = 2.406, σ = 0.1622, and g = –1 for male and +1 for female.

Risk models for leukemia (59) are

$$\mathsf{ERR}(D, e, a, g) = (\alpha \cdot D + \beta \cdot D^2) \exp[\kappa_1 \cdot \ln(a)],$$

where $\alpha = 864.552$, $\beta/\alpha = 1.18092$, $\kappa_1 = -1.647$;

$$\mathsf{EAR}(D, e, a, g) = (\alpha \cdot D + \beta \cdot D^2) \exp[\kappa_1 \cdot \mathsf{I}_{s=\mathsf{female}} + \kappa_2 \cdot \mathsf{In}(a - e)],$$

where $\alpha = 7.51650 \ 10^{-4}$, $\beta/\alpha = 1.03455$, $\kappa_1 = -0.52526$, $\kappa_2 = -0.6141$, and $I_{s = female} = 0$ for males and +1 for females. Although the model was developed for leukemia mortality, it was considered to be approximate to incidence in the WHO report owing to poor survival in the original population of the model.

The risk model for breast cancer (49) is

$$\mathsf{EAR}(D, e, a, g) = \beta_1 \cdot D \cdot \exp[\tau \cdot (e - 30) + \upsilon \cdot \ln(a/70)],$$

where $\beta_1 = 0.0009257$, $\tau = -0.04543$, and $\upsilon = 1.725$.

 $AR_{15}\ from\ 2012\ was\ calculated\ as$

$$AR_{15}(D,e,g) = \int_{e+L}^{e+15} \left[w \cdot EAR(D,e,a,g) + (1-w) \cdot ERR(D,e,a,g) \cdot m(a,g) \right] \frac{S(a,g)}{S(e,g)} dx$$

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- Harada KH, et al. (2013) 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(6): 2520–2526.
- Fisher NS, et al. (2013) Evaluation of radiation doses and associated risk from the Fukushima nuclear accident to marine biota and human consumers of seafood. Proc Natl Acad Sci USA 110(26):10670–10675.
- Garland JA, Pomeroy IR (1994) Resuspension of fall-out material following the Chernobyl accident. J Aerosol Sci 25(5):793–806.
- Ishikawa H (1995) Evaluation of the effect of horizontal diffusion on the long-range atmospheric transport simulation with Chernobyl data. J Appl Meteorol 34(7):1653–1665.
- Koizumi A, et al. (2012) Preliminary assessment of ecological exposure of adult residents in Fukushima Prefecture to radioactive cesium through ingestion and inhalation. *Environ Health Prev Med* 17(4):292–298.
- Ministry of Health Labour, and Welfare, Japan (2012) The National Health and Nutrition Survey in Japan, 2010. Available at www.mhlw.go.jp/bunya/kenkou/eiyou/dl/ h22-houkoku-02.pdf. Accessed February 5, 2014. Japanese.
- Hamada N, Ogino H (2012) Food safety regulations: What we learned from the Fukushima nuclear accident. J Environ Radioact 111:83–99.
- International Commission on Radiological Protection (2007) The 2007 Recommendations of the International Commission on Radiological Protection. *ICRP publication 103. Ann ICRP* 37(2-4):1–332.
- World Health Organization (2013) Preliminary Dose Estimation from the Nuclear Accident after the 2011 Great East Japan Earthquake and Tsunami (World Health Organization, Geneva). Available at www.who.int/iris/bitstream/10665/44877/1/9789241503662_eng.pdf.
- Nuclear Safety Research Association Japan (2011) Seikatsu Kankyo Hosyasen (Environmental Radiation) (Nuclear Safety Research Association, Tokyo), 2nd Ed. Japanese.
- Abe S, Fujitaka K, Abe M, Fujimoto K (1981) Extensive Field Survey of Natural Radiation in Japan. J Nucl Sci Technol 18(1):21–45.
- Fujimoto K (2004) Mapping of Cosmic Ray Doses in Japan (National Institute of Radiological Sciences, Chiba, Japan), NIRS-M-170. Available at http://repo.nirs.go.jp/dspace/ bitstream/918273645/233/1/nirs_m_170.pdf. Accessed February 5, 2014.
- 27. Oikawa S, et al. (2003) A nationwide survey of outdoor radon concentration in Japan. *J Environ Radioact* 65(2):203–213.

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- Sanada T, et al. (1999) Measurement of nationwide indoor Rn concentration in Japan. J Environ Radioact 45(2):129–137.
- Sugiyama H, et al. (2009) Internal exposure to ²¹⁰Po and ⁴⁰K from ingestion of cooked daily foodstuffs for adults in Japanese cities. J Toxicol Sci 34(4):417–425.
- Yamamoto M, Sakaguchi A, Tomita J, Imanaka T, Shiraishi K (2009) Measurements of ²¹⁰Po and ²¹⁰Pb in total diet samples: Estimate of dietary intakes of ²¹⁰Po and ²¹⁰Pb for Japanese. J Radioanal Nucl Chem 279(1):93–103.
- International Atomic Energy Agency (2011) Radiation protection and safety of radiation sources: International Basic Safety Standards (International Atomic Energy Agency, Vienna), interim Ed. Available at www-pub.iaea.org/MTCD/publications/PDF/ p1531interim_web.pdf. Accessed February 5, 2014.
- 32. United Nations Scientific Committee on the Effects of Atomic Radiation (1982) Ionizing Radiation: Sources and Biological Effects. United Nations Scientific Committee on the Effects of Atomic Radiation 1982 Report to the General Assembly, with Annexes (United Nations Scientific Committee on the Effects of Atomic Radiation, Vienna).
- 33. United Nations Scientific Committee on the Effects of Atomic Radiation (2011) Annex D: Health effects due to radiation from the Chernobyl accident. UNSCEAR 2008 Report to the General Assembly, with Scientific Annexes (United Nations Scientific Committee on the Effects of Atomic Radiation, Vienna), Vol 2.
- 34. Schwantes JM, Orton CR, Clark RA (2012) Analysis of a nuclear accident: Fission and activation product releases from the Fukushima Daiichi nuclear facility as remote indicators of source identification, extent of release, and state of damaged spent nuclear fuel. *Environ Sci Technol* 46(16):8621–8627.
- Steinhauser G, Schauer V, Shozugawa K (2013) Concentration of strontium-90 at selected hot spots in Japan. PLoS ONE 8(3):e57760.
- Romanyukha A, King DL, Kennemur LK (2012) Impact of the Fukushima nuclear accident on background radiation doses measured by control dosimeters in Japan. *Health Phys* 102(5):535–541.
- 37. limoto T, et al. (2012) Measures against increased environmental radiation dose by the TEPCO Fukushima Dai-ichi NPP accident in some local governments in the Tokyo metropolitan area: Focusing on examples of both Kashiwa and Nagareyama cities in Chiba prefecture. *Radiat Prot Dosimetry* 152(1-3):210–214.
- United Nations Scientific Committee on the Effects of Atomic Radiation (2008) Annex B: Exposures of the public and workers from various sources of radiation. UNSCEAR 2008 Report: Sources of Ionizing Radiation (United Nations Scientific Committee on the Effects of Atomic Radiation, Vienna), Vol 1.
- Hayano RS, et al. (2013) 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 B, Phys Biol Sci 89(4):157–163.
- Masson O, et al. (2011) Tracking of airborne radionuclides from the damaged Fukushima Dai-ichi nuclear reactors by European networks. *Environ Sci Technol* 45(18): 7670–7677.
- 41. International Commission on Radiological Protection (2002) Basic anatomical and physiological data for use in radiological protection reference values. ICRP Publication 89. *Ann ICRP* 32(3-4). Available at www.icrp.org/publication.asp?id=ICRP%20Publication% 2089. Accessed February 5, 2014.
- 42. International Commission on Radiological Protection (1996) Age-dependent doses to the members of the public from intake of radionuclides—Part 5 compilation of ingestion and inhalation coefficients. ICRP Publication 72. Ann ICRP 26(1). Available at www.icrp. org/publication.asp?id=ICRP%20Publication%2072. Accessed February 5, 2014.

- Saito K (1991) External Doses to Terrestrial Gamma Rays on the Snow Cover. Radiat Prot Dosimetry 35(1):31–39.
- 44. Fukushima Prefecture (2012) Readings of the Fourth Fukushima Prefecture Environmental Radiation Monitoring in Mesh Survey. Available at www.cms.pref. fukushima.jp/download/1/monitaring.mesyu20121001.pdf. Accessed February 5, 2014. Japanese.
- 45. Geological Survey of Japan (2004) *Geochemical Map of Japan* (National Institute of Advanced Industrial Science and Technology, Tsukuba, Japan).
- Saito K, Sakamoto R, Nagaoka T, Tsutsumi M, Moriuchi S (1997) Measurements of Gamma Dose Rates in Dwellings in the Tokyo Metropolitan Area. *Radiat Prot Do*simetry 69(1):61–67.
- Koizumi A, et al. (2013) ¹³⁷Cs trapped by biomass within 20 km of the Fukushima Daiichi Nuclear Power Plant. *Environ Sci Technol* 47(17):9612–9618.
- Matsuda A, et al. (2013) Cancer incidence and incidence rates in Japan in 2007: A study of 21 population-based cancer registries for the Monitoring of Cancer Incidence in Japan (MCIJ) project. Jpn J Clin Oncol 43(3):328–36.
- Preston DL, et al. (2007) Solid cancer incidence in atomic bomb survivors: 1958-1998. Radiat Res 168(1):1–64.
- 50. World Health Organization (2013) Health Risk Assessment from the Nuclear Accident After the 2011 Great East Japan Earthquake and Tsunami, Based on a Preliminary Dose Estimation (World Health Organization, Geneva). Available at www.who.int/iris/ bitstream/10665/78218/1/9789241505130_eng.pdf. Accessed February 5, 2014.
- Committee to Assess Health Risks from Exposure to Low Levels of Ionizing Radiation, National Research Council of the National Academies (2006) Health Risks from Exposure to Low Levels of Ionizing Radiation: BEIR VII-Phase 2 (National Academies, Washington, DC).
- Japan Industrial Standard (2004) Z 4339, Optically stimulated luminescence dosimetry systems (Japanese Standards Association, Tokyo).
- 53. Koizumi A, et al. (2009) Past, present, and future of environmental specimen banks. Environ Health Prev Med 14(6):307–318.
- Kaiser H, Specker H (1956) Bewertung und Vergleich von Analysenverfahren. Fresenius Z Anal Chem 149(1-2):46–66.
- Ministry of Education, Culture, Sports, Science, and Technology (2003) Analytical method of radioactive strontium. *Radioactivity Measurement Series No. 2* (Japan Chemical Analysis Center, Chiba, Japan). Japanese.
- National Center for Environmental Assessment (2011) Exposure Factors Handbook: 2011 Edition (US Environmental Protection Agency, Washington, DC).
- 57. Jacob P, Rosenbaum H, Petoussi N, Zankl M (1990) Calculation of organ doses from environmental gamma rays using human phantoms and Monte Carlo methods. Part II: Radionuclides distributed in air or deposited on the ground. *GSF-Report* 12/90 (GSF National Research Center on Environment and Health, Neuherberg, Germany).
- 58. Statistics Bureau of Japan (2011) Table 5.16: Death rates (per 100,000 population) by sex, age and causes (the condensed list of causes of death for Japan), 2010. Vital Statistics of Japan: General Mortality, Vol 1. Available at www.e-stat.go.jp/SG1/estat/ ListE.do?lid=000001101825. Accessed February 5, 2014.
- 59. United Nations Scientific Committee on the Effects of Atomic Radiation (2008) Annex A: Epidemiological studies of radiation and cancer. UNSCEAR 2006 Report to the General Assembly: Effects of Ionizing Radiation (United Nations Scientific Committee on the Effects of Atomic Radiation, Vienna), Vol 1.