in all countries. No pattern difference was observed between Japan and Korea, while the lower chlorinated PCB congeners were relatively predominant in human milk from China.

44.4 New POPs

As mentioned above, 12 chlorinated compounds were listed in the Stockholm Convention in 2004. These compounds are a small proportion of the enormous number of chemicals used in consumer and industrial products. Very persistent and very bioaccumulative chemicals are screened based on their degradability and bioaccumulativeness. New POPs and their candidates are not only chlorinated pesticides but also brominated and fluorinated compounds such as flame retardants and surfactants. The application of these chemicals have various formulations due to production processes and patents, which allows for the provision of fingerprints of contamination sources in emission-source countries. We briefly describe the profiles of new POPs and candidates in breast milk in East Asian countries.

44.4.1 Brominated compounds

This class of compounds has been used as a substitute for PCBs in flame retardants. The increase in sales of home appliances led to an increase in fire incidents since the 1970s, which in turn resulted in the development of flame retardants. Flame retardants are produced in a variety that is available today. The brominated flame retardant compounds are often stabilised bromine atoms that dissociate into radical species that compete with chain-propagating steps in the combustion process. There was a concern that PBDEs included an impurity of brominated dibenzo dioxins or may generate brominated dibenzo dioxins by incineration.

Brominated flame retardants are used in housing, electrical products, substrates, and textile products. In 1995, manufacturers of brominated flame retardants began introducing voluntary regulation. Pentabromodiphenylether, relatively toxic congeners of PBDEs and polybrominated biphenyls with a structure similar to PCBs, have been phased out of production. Now they are specified in Annex A of the Stockholm Convention. Decabromodiphenyl ether is considered low risk and is still in use.

PBDEs are comprised of 209 congeners with a different number of bromine atoms and substitution position like PCBs. Their physicochemical natures are very different from the various bromine substitutes. A bioaccumulation test using carp shows a bioconcentration factor of 11,700-1,650 for pentabromodiphenylethers and less than 50 for decabromodiphenyl ether (BDE-209). Although BDE-209 has the high octanol-water partition coefficient (log Kow) of 10.1, it shows no such high bioconcentration of other POPs. An administration assay on SD rats indicated that oral absorption rate of BDE-209 is relatively low – about 10% (Morck *et al.*, 2003). Oral administration of BDE209 in 3 days after birth to day 19 to male mice showed that behavioural abnormalities were observed at 20.1 mg/kg (Viberg *et al.*, 2003). On the other hand, pentabromodiphenylether caused behavioural effects above a dose of 0.8 mg/kg, suggesting its toxicity is higher than a high-brominated PBDEs (Viberg *et al.*, 2004).

In a study of serum of adult women in Japan, geometric mean PBDE concentration is 0.5 ng/g lipid in 1980s, and 1.8 ng/g lipid in the 1990s (Koizumi *et al.*, 2005). Serum of nursing mothers in 2005 in Japan showed geometric mean PBDE concentrations of 2.9 ng/g lipid, which was low compared to the population exposed occupationally but has increased historically (Inoue *et al.*, 2006). Tetrabrominated congeners were the predominant composition in breast milk samples, followed by hexabrominated congeners. The proportion of BDE-209 was about 10%.

Levels of PBDEs in breast milk were compared among mothers from East Asian countries (Haraguchi *et al.*, 2009) (Figure 44.4). The mean concentration of PBDEs was highest in mothers in Korea (3.7 ng/g lipid), followed by China (1.9 ng/g lipid), Japan (1.5 ng/g lipid), and Vietnam (0.42 ng/g lipid). In Korea, the PBDE levels varied widely (range 0.82-24 ng/g lipid), suggesting that subpopulations are still highly exposed to PBDEs. In Japanese mothers, survey results showed no increasing trends in PBDE levels. The potential exposure pathways are thought to be via dietary intake or house dust ingestion/inhalation, which are released from flame retardants in home products. In Chinese breast milk, levels in 2008 (1.9 ng/g lipid) were higher than those in the 2005 survey from Beijing (1.2 ng/g lipid), which may indicate an increasing trend of PBDEs exposure. PBDE levels in breast milk in East Asian countries were low compared with levels in Western countries, especially the USA. Even though there were variations in the measurement method for PBDE congeners, there is a regional difference in exposure to PBDEs.

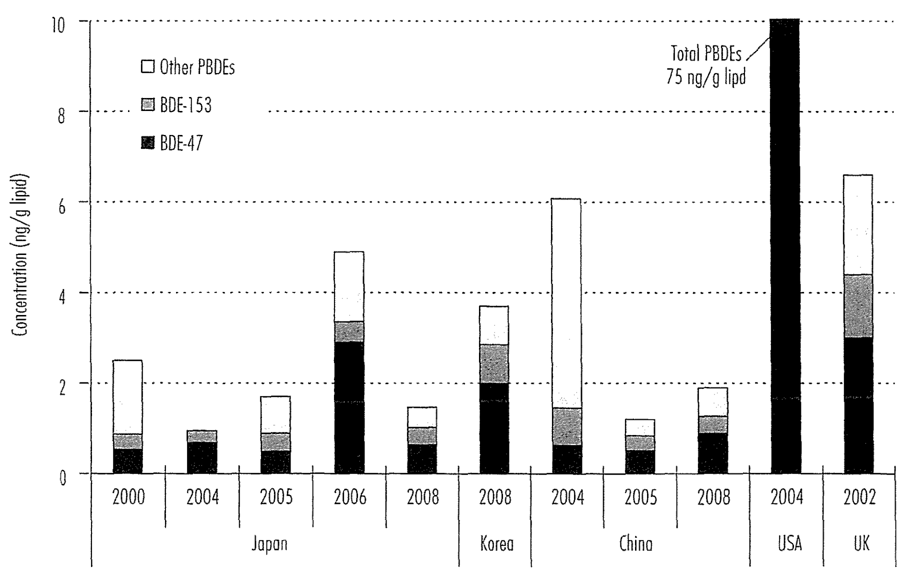

Figure 44.4. Comparison of levels of polybrominated diphenyl ethers (PBDEs) in breast milk in different countries.

44.4.2 Fluorinated compounds

PFCs comprise a large group of man-made fluorinated organic chemicals which consists of a fluorine-substituted alkyl chain. They have been produced for over 60 years and are used for various industrial and consumer-related applications, such as food packaging materials, protective coatings for textiles, carpets, papers, and surfactants for fluoropolymer production. During the last decade, PFCs such as PFOS and PFOA have been found at considerable levels in various biota samples including the liver and tissues, and especially human blood and breast milk, worldwide (Fromme *et al.*, 2009).

Prenatal and postnatal toxic effects of PFCs were observed in rats and mice, which include increased liver weight, growth lags, and delayed development. Several epidemiological studies also suggested the developmental effects of PFOS and PFOA on children (Steenland *et al.*, 2010). PFCAs with longer chains than PFOA are bioaccumulative. The emissions of PFNA and PFUnDA were 25 and 7 metric tons (Prevedouros *et al.*, 2006), respectively, in 2000, and caused mainly in East Asia. These compounds have been detected in wildlife.

Breast milk samples in East Asian countries were investigated for these PFCAs in 2008-2010 (Fujii *et al.*, 2012) (Figure 44.5). PFOA was the predominant compound and was detected in more than 60% of samples in all three Asian countries. The median concentration of PFOA was highest in Japan (89 pg/ml), followed by Korea (62 pg/ml) and China (51 pg/ml). PFNA and PFUnDA were observed at comparable levels to PFOA in these countries. The levels of PFNA and PFUnDA were lower in Korea (median 15 and 19 pg/ml) and China (median 15 and 15 pg/ml) than in Japan (median 31 and 35 pg/ml). In Korea, interestingly, half of the milk samples contained detectable levels of PFTrDA, which was the highest among the three countries (median 10 pg/ml). Regarding the total PFCAs in the milk samples, PFOA accounted for 48%, 54%, and 61% in Japan, Korea, and China, respectively. The infant formulas were also analysed for PFCAs. PFOA, PFNA, perfluorodecanoic acid and PFUnDA were frequently detected in both Japan (21.8, 27.6, 10.1, 10.1 pg/ml) and China (28.1, 22.4, 11.1, <5 pg/ml), but there were no statistical differences between Japan and China (Fujii *et al.*, 2012). No formula samples contained detectable perfluorododecanoic acid and PFTrDA. The PFOA levels in the formula samples were 4-fold and 2-fold lower in Japan and China compared with the breast milk samples, respectively.

There have been limited studies on PFCAs levels in breast milk. An obvious difference in PFOA levels has not yet been observed between East Asian and Western countries. An analysis of PFCAs other than PFOA has not been conducted because of analytical difficulties. Thus, the total amounts of PFCAs and their profiles in breast milk have been unveiled. Future development of analytical methods for PFCAs will envision differences of PFCA exposure profiles in human breast milk in various countries.

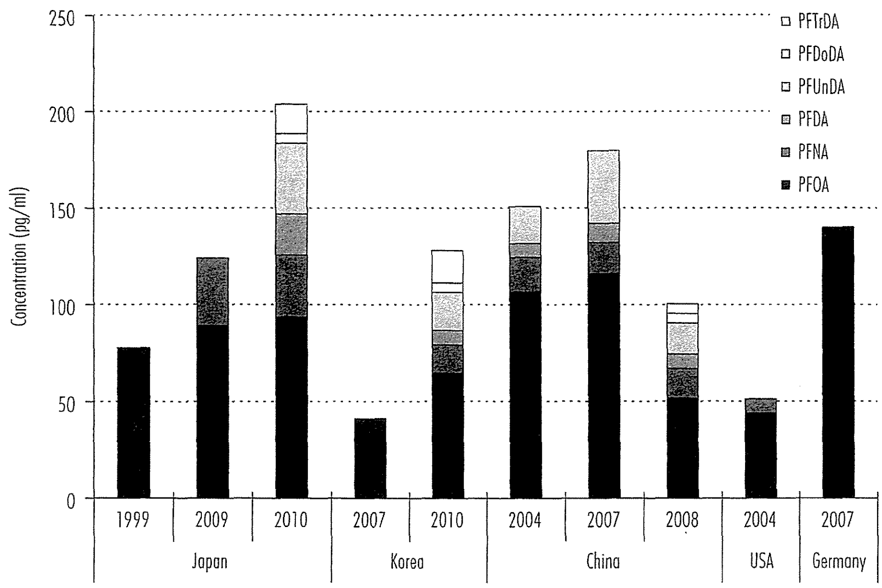

Figure 44.5. Comparison of levels of perfluorinated carboxylic acids in breast milk in different countries. PFOA = perfluorooctanoic acid; PFNA = perfluorononanoic acid; PFDA = perfluorodecanoic acid; PFUnDA = perfluoroundecanoic acid; PFDoDA = perfluorododecanoic acid; PFTrDA = perfluorotridecanoic acid.

44.5 Risk assessment of chemicals in human breast milk for infants

Two parameters, TDI and MOE, are commonly used to express risks through foods. TDI is an estimate with uncertainty of a daily exposure to the human population; the daily exposure is unlikely to be associated with appreciable health risk. MOE is a scale of how much less the current exposure is than the reference dose: MOE = TDI/Exposure. MOE<100 needs further risk analysis. Likewise, these parameters are used to describe risks of chemicals in breast milk for infants.

In Table 44.2, the estimation of daily intake of contaminants was summarised based on standard breast milk consumption by infants (Van Oostdam *et al.*, 1999) (750 g/day; 5 kg body weight). Only limited data have been available for chemicals in human breast milk. The TDI for PFOA was calculated to be 1,500 ng/kg body weight/d by the European Food Safety Authority in 2008. The calculated levels of PFOA intake were less than 1% of TDI in the East Asian population (means: 0.5-0.9% of TDI). For PFCAs other than PFOA, similar TDI levels are used for risk estimation and total PFCA exposure in Japan was at most 1.9% of TDI. Employing the same procedure, PBDE exposures were evaluated. Estimates of total PBDE intake were around 1% of NOAEL of penta-BDEs (NOAEL: 0.4 mg/kg body weight/day) (Viberg *et al.*, 2004), indicating that the health risks of PBDEs from breast milk are low.

Table 44.2. Daily intake estimates (ng/kg body weight/d) and risk assessment for infants.[1,2]

	Japan		Korea		China	
	Mean	% of TDI[3]	Mean	% of TDI[3]	Mean	% of TDI[3]
PFOA	14.0	0.9	9.7	0.6	7.7	0.5
PFNA	4.8	0.3	2.2	0.1	2.3	0.2
PFDA	3.2	0.2	1.1	0.1	1.1	0.1
PFUnDA	5.5	0.4	2.9	0.2	2.4	0.2
PFDoDA	0.8	0.1	0.8	0.1	0.8	0.1
PFTrDA	2.3	0.2	2.5	0.2	0.8	0.1
ΣPFCAs	29.1	1.9	17.7	1.2	13.2	0.9
BDE-28	11	0.03	21	0.05	65	0.16
BDE-47	96	0.24	300	0.75	134	0.33
BDE-99	18	0.05	50	0.12	11	0.03
BDE-100	17	0.04	42	0.11	12	0.03
BDE-153	59	0.15	129	0.32	57	0.14
BDE-154	24	0.06	20	0.05	8	0.02
Σ PBDE	225	0.56	555	1.39	285	0.71

[1] The breast milk consumption rate and body weightwere assumed to be 750 g/d and 5 kg, respectively.
[2] PFCAs = perfluorinated carboxylic acids; PFOA = perfluorooctanoic acid; PFNA = perfluorononanoic acid; PFDA = perfluorodecanoic acid; PFUnDA = perfluoroundecanoic acid; PFDoDA = perfluorododecanoic acid; PFTrDA = perfluorotridecanoic acid; PBDE = polybrominated diphenyl ether; BDE = brominated diphenyl ether.
[3] Tolerable daily intake: 1,500 ng/kg body weight/d for PFCAs and 0.4 mg/kg body weight/d for PBDEs.

44.6 Conclusion

Due to their lipophilicity, persistency and bioaccumulativeness, classic and new POPs are easily transferred from mother to infants via breast milk. POPs profiles in human breast milk in East Asian countries revealed historical aspects of their uses, production processes of POPs and sources of POPs. For example, concentrations of DDT and HCB levels are very high in breast milk of Chinese mothers, while CHL is high in breast milk of Japanese mothers and PBDEs in breast milk of Korean mothers. Those salient differences in profile are associated with the past and current exposure profiles of these chemicals in the three countries. Thus breast milk monitoring is very informative in this sense. In addition, the breast milk is the main energy and nutrient source for infants. Chemicals in human breast milk may cause developmental adverse health effects to infants. Thus continuous monitoring of chemicals in human breast milk is needed for child health. Despite the importance of breast milk monitoring, not much data has been generated. To facilitate the rational monitoring programme, enforcement of three areas is needed: first, rational prediction of the ratio of concentrations of molecules in breast milk to that in plasma; second, advances in analytical methods; and third, a large human breast milk bank.

References

Abadin, H.G., Hibbs, B.F. and Pohl, H.R., 1997. Breast-feeding exposure of infants to cadmium, lead, and mercury: a public health viewpoint. Toxicology and Industrial Health 13, 495-517.

Barber, J., Sweetman, A., van Wijk, D. and Jones, K., 2005. Hexachlorobenzene in the global environment: Emissions, levels, distribution, trends and processes. Science of the Total Environment 349, 1-44.

Dabeka, R.W., Karpinski, K.F., McKenzie, A.D. and Bajdik, C.D., 1986. Survey of lead, cadmium and fluoride in human milk and correlation of levels with environmental and food factors. Food and Chemical Toxicology 24, 913-921.

Fromme, H., Tittlemier, S.A., Volkel, W., Wilhelm, M. and Twardella, D., 2009. Perfluorinated compounds – exposure assessment for the general population in Western countries. International Journal of Hygiene and Environmental Health 212, 239-270.

Fujii, Y., Haraguchi, K., Harada, K.H., Hitomi, T., Inoue, K., Itoh, Y., Watanabe, T., Takenaka, K., Uehara, S., Yang, H.R., Kim, M.Y., Moon, C.S., Kim, H.S., Wang, P., Liu, A., Hung, N.N. and Koizumi, A., 2011. Detection of dicofol and related pesticides in human breast milk from China, Korea and Japan. Chemosphere 82, 25-31.

Fujii, Y., Yan, J., Harada, K.H., Hitomi, T., Yang, H., Wang, P. and Koizumi, A., 2012. Levels and profiles of long-chain perfluorinated carboxylic acids in human breast milk and infant formulas in East Asia. Chemosphere 86, 315-321.

Guidry, A.J., O'Brien, C.N. and Douglass, L.W., 1998. A bovine mammary endothelial/epithelial cell culture model of the blood/milk barrier. Canadian Journal of Veterinary Research 62, 117-121.

Haraguchi, K., Koizumi, A., Inoue, K., Harada, K.H., Hitomi, T., Minata, M., Tanabe, M., Kato, Y., Nishimura, E., Yamamoto, Y., Watanabe, T., Takenaka, K., Uehara, S., Yang, H.R., Kim, M.Y., Moon, C.S., Kim, H.S., Wang, P., Liu, A. and Hung, N.N., 2009. Levels and regional trends of persistent organochlorines and polybrominated diphenyl ethers in Asian breast milk demonstrate POPs signatures unique to individual countries. Environment International 35, 1072-1079.

Inoue, K., Harada, K., Takenaka, K., Uehara, S., Kono, M., Shimizu, T., Takasuga, T., Senthilkumar, K., Yamashita, F. and Koizumi, A., 2006. Levels and concentration ratios of polychlorinated biphenyls and polybrominated diphenyl ethers in serum and breast milk in Japanese mothers. Environmental Health Perspectives 114, 1179-1185.

Koizumi, A., Harada, K.H., Inoue, K., Hitomi, T., Yang, H.R., Moon, C.S., Wang, P., Hung, N.N., Watanabe, T., Shimbo, S. and Ikeda, M., 2009. Past, present, and future of environmental specimen banks. Environmental Health and Preventive Medicine 14, 307-318.

Koizumi, A., Yoshinaga, T., Harada, K., Inoue, K., Morikawa, A., Muroi, J., Inoue, S., Eslami, B., Fujii, S., Fujimine, Y., Hachiya, N., Koda, S., Kusaka, Y., Murata, K., Nakatsuka, H., Omae, K., Saito, N., Shimbo, S., Takenaka, K., Takeshita, T., Todoriki, H., Wada, Y., Watanabe, T. and Ikeda, M., 2005. Assessment of human exposure to polychlorinated biphenyls and polybrominated diphenyl ethers in Japan using archived samples from the early 1980s and mid-1990s. Environmental Research 99, 31-39.

Konishi, Y., Kuwabara, K. and Hori, S., 2001. Continuous surveillance of organochlorine compounds in human breast milk from 1972 to 1998 in Osaka, Japan. Archives of Environmental Contamination and Toxicology 40, 571-578.

Morck, A., Hakk, H., Orn, U. and Klasson Wehler, E., 2003. Decabromodiphenyl ether in the rat: absorption, distribution, metabolism, and excretion. Drug Metabolism and Disposition: The Biological Fate of Chemicals 31, 900-907.

Nakata, H., Nasu, T., Abe, S., Kitano, T., Fan, Q., Li, W. and Ding, X., 2005. Organochlorine contaminants in human adipose tissues from China: mass balance approach for estimating historical Chinese exposure to DDTs. Environmental Science & Technology 39, 4714-4720.

Prevedouros, K., Cousins, I.T., Buck, R.C. and Korzeniowski, S.H., 2006. Sources, fate and transport of perfluorocarboxylates. Environmental Science & Technology 40, 32-44.

Shennan, D.B. and Peaker, M., 2000. Transport of milk constituents by the mammary gland. Physiological Reviews 80, 925-951.

Smit, J.J.M., Schinkel, A.H., Oude Elferink, R.P.J., Groen, A.K., Wagenaar, E., Van Deemter, L., Mol, C.A.A.M., Ottenhoff, R., Van der Lugt, N.M.T., Van Roon, M.A., Van der Valk, M.A., Offerhaus, G.J.A., Berns, A.J.M. and Borst, P., 1993. Homozygous disruption of the murine mdr2 P-glycoprotein gene leads to a complete absence of phospholipid from bile and to liver disease. Cell 75, 451-462.

Steenland, K., Fletcher, T. and Savitz, D.A., 2010. Epidemiologic evidence on the health effects of perfluorooctanoic acid (PFOA). Environmental Health Perspectives 118, 1100-1108.

Taguchi, S. and Yakushiji, T., 1988. Influence of Termite Treatment in the Home on the Chlordane Concentration in Human Milk. Archives of Environmental Contamination and Toxicology 17, 65-71.

Tashiro, S. and Matsumura, F., 1977. Metabolic routes of *cis*- and *trans*-chlordane in rats. Journal of Agricultural and Food Chemistry 25, 872-880.

Tsydenova, O., Sudaryanto, A., Kajiwara, N., Kunisue, T., Batoev, V. and Tanabe, S., 2007. Organohalogen compounds in human breast milk from Republic of Buryatia, Russia. Environmental Pollution 146, 225-232.

UNEP, 2004. Stockholm Convention on POPs to become international law, launching a global campaign to eliminate 12 hazardous chemicals. Available at: http://www.pops.int/documents/press/EIF/pr5-04POPsEIF-E.pdf.

Van Oostdam, J., Gilman, A., Dewailly, E., Usher, P., Wheatley, B., Kuhnlein, H., Neve, S., Walker, J., Tracy, B., Feeley, M., Jerome, V. and Kwavnick, B., 1999. Human health implications of environmental contaminants in Arctic Canada: a review. Science of the Total Environment 230, 1-82.

Viberg, H., Fredriksson, A. and Eriksson, P., 2004. Investigations of strain and/or gender differences in developmental neurotoxic effects of polybrominated diphenyl ethers in mice. Toxicological Sciences 81, 344-353.

Viberg, H., Fredriksson, A., Jakobsson, E., Orn, U. and Eriksson, P., 2003. Neurobehavioral derangements in adult mice receiving decabrominated diphenyl ether (PBDE 209) during a defined period of neonatal brain development. Toxicological Sciences 76, 112-120.

Wong, M., Leung, A., Chan, J. and Choi, M., 2005. A review on the usage of POP pesticides in China, with emphasis on DDT loadings in human milk. Chemosphere 60, 740-752.

Yu, H., Zhao, X., Zhao, J., Zhu, Z. and Zhao, Z., 2006. Continuous surveillance of organochlorine pesticides in human milk from 1983 to 1998 in Beijing, China. International Journal of Environmental Health Research 16, 21-26.

Bull Environ Contam Toxicol
DOI 10.1007/s00128-014-1241-6

Methylmercury Monitoring Study in Karakuwacho Peninsula Area in Japan

Junxia Yan · Kayoko Inoue · Akihiro Asakawa · Kouji H. Harada · Takao Watanabe · Noriyuki Hachiya · Akio Koizumi

Received: 1 October 2013 / Accepted: 24 February 2014

Abstract Methylmercury (MeHg) is a worldwide concern owing to its adverse health effects. To explore MeHg exposure burdens and the potential contributing factors in different subpopulations in a peninsula area (Karakuwacho) in Japan, a cross-sectional survey was performed. This study included 189 individuals from 102 families. The geometric means of total hair mercury (THg) were 5.74, 3.78 and 2.37 μg/g for adult males, females and children, respectively, of which 56.5 %, 30.9 % and 12.9 % had hair THg exceeding 5 μg/g, respectively. Tuna and mackerel were the common fish species that were positively correlated with hair THg levels in different subpopulations (standardized coefficient ranged from 0.20 to 0.58, $p < 0.05$). Frequent consumption of these fish species and a large amount of fish intake are likely major contributors of MeHg exposure in this area. Local-scale risk evaluation and risk communication should be highlighted in future studies.

J. Yan · K. Inoue · K. H. Harada · A. Koizumi (✉)
Department of Health and Environmental Sciences, Kyoto University Graduate School of Medicine, Yoshida-Sakyo, Kyoto 606-8501, Japan
e-mail: koizumi.akio.5v@kyoto-u.ac.jp

A. Asakawa
Department of Social and Behavioral Medicine, Kagoshima University Graduate School of Medical and Dental Sciences, Kagoshima 890-8544, Japan

T. Watanabe
Miyagi University of Education, Sendai 980-0845, Japan

N. Hachiya
Department of International Affairs and Environmental Sciences, National Institute for Minamata Disease, 4058-18 Hama, Minamata 867-0008, Japan

Keywords Hair mercury · Methylmercury · Fish species · Japanese population

Mercury is a recognized toxic pollutant of public health concern. Its global distribution is caused by natural processes and anthropogenic activities. Inorganic mercury can be transformed into organic forms (mainly MeHg), which can be bioaccumulated and biomagnified in aquatic food chains (Matthews 1983; Mergler et al. 2007). Various studies have determined that fish consumption is the main source of human exposure to MeHg (Hightower and Moore 2003; Knobeloch et al. 2005). MeHg exposure can lead to various adverse health effects, especially neurological symptoms (Guallar et al. 2002; Harada 1995; Mahaffey 1998; Mozaffarian and Rimm 2006). These adverse health effects may occur at low levels that were previously thought to be safe (Karagas et al. 2012; Maruyama et al. 2012).

In Japan, people habitually consume more fish products than other countries. Historically, several catastrophes (i.e., Minamata disease and Niigata Minamata disease) have been caused by MeHg-contaminated fish consumption (Harada 1995). Several monitoring studies have indicated higher exposure levels in Japan compared with reference values or other populations (Yasuda et al. 2005; Yasutake et al. 2004). Therefore, identifying the high-risk areas and the major contributors is essential for developing effective reduction strategies. In a previous monitoring study (unpublished data), we identified that peninsula or island areas had higher mercury exposure levels. To further investigate the exposure pattern and the potential contributing factors in different subpopulations in such areas, we performed a family-based study of MeHg exposure in Karakuwacho, a peninsula area in the northeast of Miyagi prefecture in Japan.

Materials and Methods

This study was performed in Karakuwacho and included 189 individuals from 102 families. There were ten families composed of father, mother and children; 28 families composed of mother and children; and the other families we collected one individual or two non-parent–child paired individuals. Generally, there were 104 adults (81 females and 23 males) and 85 children (Table 1). For each individual, hair samples (0.1–1.0 g) were cut from the base of the scalp, behind the ear, and the samples were washed with neutral detergents, rinsed twice with acetone, and then dried at room temperature and stored in a desiccator in Kyoto University Human Specimen Bank (Koizumi et al. 2009) until they were analyzed for total mercury (THg). At the time of hair sampling, body weight was measured by a investigator using a standard scale. A questionnaire was administered to the participants to collect information regarding their age, gender, total fish consumption frequency, and commonly consumed fish species. All participants were fully informed about the purposes of the study and provided written consent. The study was approved by the Institutional Review Board and Ethics Committee of the Kyoto University School of Medicine, Japan.

For hair THg analysis, the preconditioned dry hair sample was cut into small pieces (<2 mm) with scissors. Aliquots of samples (15–20 mg) were dissolved in 0.5 mL 2 N NaOH while being heated at 60°C for 1 h. Ten or twenty microliter of the solution was used to analyze the THg levels by the oxygen combustion–gold amalgamation method and an MD-1 atomic absorption detector (Nippon Instruments, Co., Ltd., Osaka, Japan) (Yasutake et al. 2003). This technique of quantification is based on a pyrolysis process of the sample using a combustion tube heated at 700°C under an oxygen atmosphere. Vaporized mercury was transferred to gold-absorber at a carrier gas flow rate of 0.5 L/min. Gaseous compounds other than mercury was eliminated from the system. Gold-absorber was then heated at 700°C for 2 min to vaporize and transfer concentrated mercury to detector. The concentration of mercury was determined by measuring the absorbance of gas at 253.7 nm emitted from mercury-vapor lamp. The limit of detection was 0.1 μg/g-hair (signal to noise ratio: 3) and the limit of quantitation was 0.3 μg/g-hair. All the individuals had hair THg levels above the limit of quantitation. The external standard was 2.5 nM mercuric chloride (0.5 μgHg/mL) in 0.5 M L-cysteine/2 % bovine serum albumin solution. To ensure precision of instrument, standard solution was analyzed in every ten analyses. The analysis was qualitatively confirmed by analyzing a certified reference material of human hair, NIES CRM No. 13 (hair reference material for MeHg, THg and other trace elements, National Institute for Environmental Studies, Japan) with a certified THg value of 4.42 ± 0.2 μg/g (http://www.nies.go.jp/labo/crm/hair.html). The THg level from our method above was 4.55 ± 0.05 μg/g. No detectable contamination was observed from procedural blank samples in every 20 samples. The hair THg levels were compared with various limit levels (1.0, 2.2, 2.7 and 5.0 μg/g) proposed by Japan or other international authorities (US EPA.2000; WHO/JEFCA 2004; The Food Safety Commission, Japan, 2005).

Statistical analysis was performed using STATiSTiCA64 (Supplied by Statsoft, OK, USA). Normally distributed

Table 1 Hair THg levels (μg/g) in 189 individuals in Karakuwacho

Variables	Ten father–mother–children paired families			28 mother–children paired families		Total		
	Fathers	Mothers	Children	Mothers	Children	Adult female	Adult male	Children
No	10	10	15	28	38	81	23	85
Age (years)								
Mean ± SD	40.7 ± 7.2	39.7 ± 5.9	7.8 ± 4.0	35.0 ± 4.8	5.0 ± 3.2	47.2 ± 14.9	43.3 ± 9.8	5.76 ± 3.3
Range	27–50	31–48	2–14	27–47	1–14	21–82	23–63	1–15
Male/female	–	–	–	–	–	–	–	42/43
Hair THg (μg/g)*								
Min	2.21	1.93	0.64	1.12	0.6	1.08	2.21	0.6
Max	12.24	6.79	10.76	13.78	8.38	15.95	14.98	10.76
GM	6.71	3.75	1.92	3.45	2.57	3.78	5.74	2.37
Correlation analysis	F versus M	M versus C	F versus C	M versus C				
Spearman coefficient	0.079	−0.442	0.267	0.290				
p value	0.828	0.200	0.455	0.134				

F versus M, father versus mother; M versus C, mother versus children; F versus C, father versus children

* Kruskal–Wallis test, $p < 0.001$

variants were described by their means and standard deviations. The minimum, maximum, and geometric mean (GM) were used to describe log-normally distributed variants. Non-parametric Kruskal–Wallis tests were conducted to compare THg differences between different populations. Spearman correlation analysis was conducted to explore the relationship between hair THg and fish consumption frequency. Stepwise multiple linear regression analysis was employed to assess the relationship between log THg levels in hair and covariates, such as age, sex, body weight and commonly consumed fish species. The significance level was set at less than 0.05.

Results and Discussion

Mercury, especially MeHg, is a worldwide concern owing to its adverse health effects. In order to ensure the population health, effective reduction strategies should be developed in the high exposure areas. With the aim to identify the detail exposure pattern and the major contributing factors in such a high exposure area, a cross sectional survey was performed in Karakuwacho, a peninsula area in Japan.

Generally, wide variation in hair THg levels was observed for different members of the same family. Fathers had the highest hair mercury level, follow by mothers, and then children (Kruskal–Wallis test, $p = 0.001$). In the same household, there were no correlations in hair mercury levels between family members (correlation analysis, all $p > 0.05$) (Table 1). Considering the potentially different fish consumption patterns, we divided the participants into three subgroups (adult males, adult females and children) for further analysis. Adult males had higher hair THg levels than adult females and children (GMs of THg were 5.74, 3.78 and 2.37 μg/g, respectively, Kruskal–Wallis test, $p < 0.001$) (Table 1). The hair THg levels in this area were significantly higher than the national average levels in Japan (GM values of 2.42 and 1.37 μg/g for males and females, respectively) (Yasutake et al. 2005) and the United States (GM values of 0.12 and 0.20 μg/g for children and women, respectively) (McDowell et al. 2004). There were 30.9 %, 56.5 % and 12.9 % of adult females, adult males and children with hair THg levels that exceeded the least strict limit (5.0 μg/g), respectively (Fig. 1). When considering the at-risk population (women of childbearing age, n = 50), 76 % of them had hair THg exceeding the corresponding limit in Japan (2.7 μg/g), and 26.0 % had hair THg levels exceeding 5.0 μg/g. These levels are significantly higher than levels in other countries (Mahaffey et al. 2009; Kim and Lee 2010). Because MeHg accounts for more than 80 % of THg in hair, THg in hair is thought to be a reliable indicator of MeHg exposure (Cernichiari et al. 1995). The general population in this area commonly faces high risk of MeHg exposure.

To elucidate the potential contributing risk factors, fish consumption frequency and commonly consumed fish species were investigated. We found that hair THg levels significantly increased with the frequency of fish consumption (Fig. 2). This may explain the hair THg distribution difference between different subpopulations. Adults tended to consume fish more frequently than children; thus, higher hair THg levels were observed (Fig. 2). In present study, we found 25 species of fish were commonly consumed in the survey area (Fig. 3). According to monitoring data released by the government (Online document. MHLW.2010), a majority of these fish species had THg concentrations with less concern (<0.1 ppm), but increasing concern was raised about commonly consumed high-end predatory fish including marlin, tuna, and alfonsino in which the THg concentration was significantly higher than the permitted limit in Japan (>0.4 ppm) (Nakagawa et al. 1997) (Fig. 3).

Multivariate analyses revealed that tuna or canned tuna were the common fish species positively correlated with hair THg levels in different subpopulations (Table 2). Tuna, which is a carnivorous fish with high mercury accumulation, is often consumed in Japan. Yasutake et al. (2004) determined that in the Miyagi area, the average fish consumption was 96 g/person/day, and 69 % of participants in their survey frequently consumed tuna. In the present study, we similarly found that 52.9 % of individuals commonly consumed tuna, and 20.3 % of individuals consumed tuna more than once per week. High tuna consumption tended to be the major contributor to high MeHg exposure in this area.

It is important to note that even fish species contaminated with low levels of mercury, if consumed frequently and eaten in large amounts, may increase the cumulative risk. In the present study, we observed a positive correlation between mackerel consumption and hair THg levels among adult population, even though mackerel has relatively low mercury levels (mean THg level of 0.11 μg/g) (Table 2; Fig. 3). In Japan, the government released a fish advisory in 2003 and further revised it in 2005 (Ser and Watanabe 2012). The advisory mainly focused on ocean fish species. There was no restriction for species with low mercury content. However, in developing a risk reduction strategy, we should consider the contamination level as well as fish consumption amount at the same time.

Although consumption of fish represents a major source of dietary MeHg exposure for the inhabitants of the Karakuwacho peninsula region, this food source also provides high-quality protein and is also rich in unsaturated fatty acids. Consequently, reducing fish consumption as a mean of reducing exposure to MeHg must weigh both the risks and benefits associated with consuming fish as a

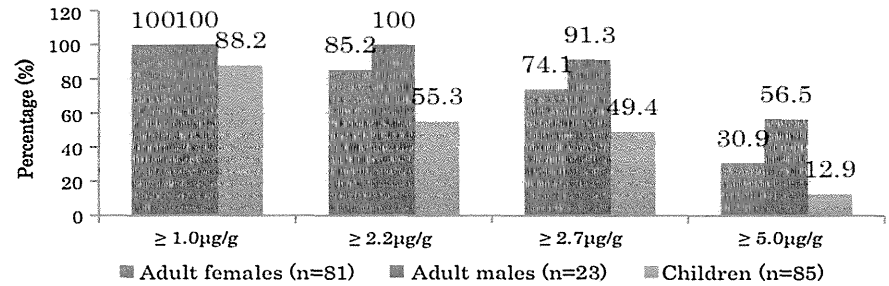

Fig. 1 The percentage of hair THg levels above the limits in different subpopulations of Karakuwacho. Limits: 1.0, 2.2, 2.7 and 5.0 μg/g for hair THg levels correspond to the US EPA, JECFA, and Japanese Food Safety Commission proposed intake limits for pregnant women and the general population (0.7, 1.6, 2.0 and 3.4 μg/kg bw/week), respectively

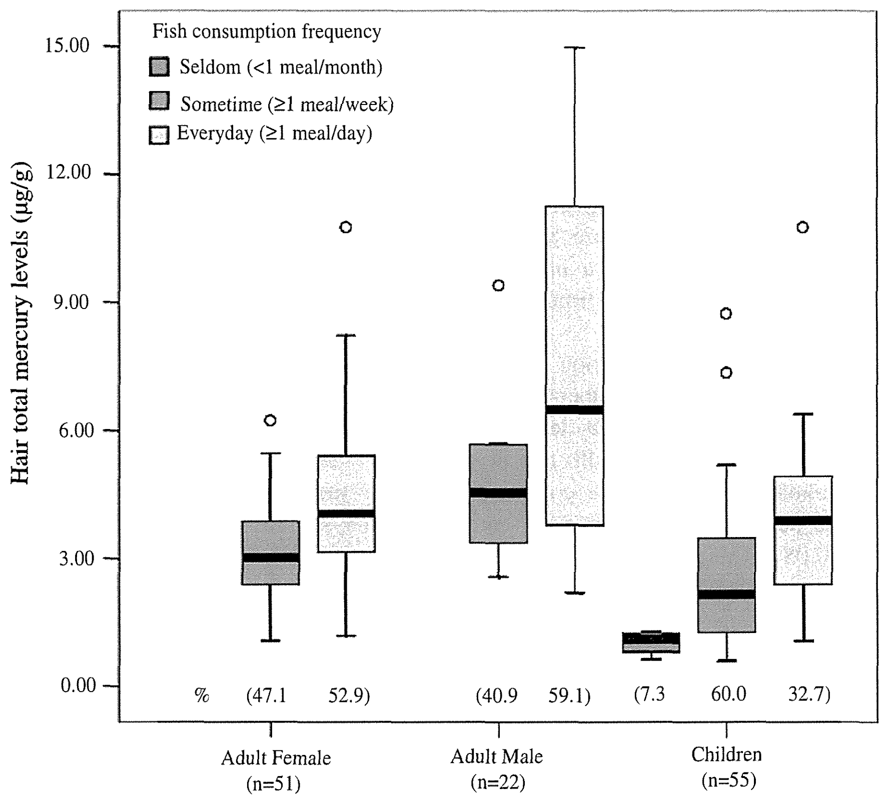

Fig. 2 Correlation between hair THg levels and fish consumption frequency. Hair THg levels significantly increased with fish consumption frequency. *Boxes* depict 25th, 50th and 75th percentiles, and whiskers depict *minimum* and maximum values, excluding outliers. *Circles* depict the outliers. The *number* below the *boxes* indicates the proportion of each fish consumption frequency in the corresponding subpopulation

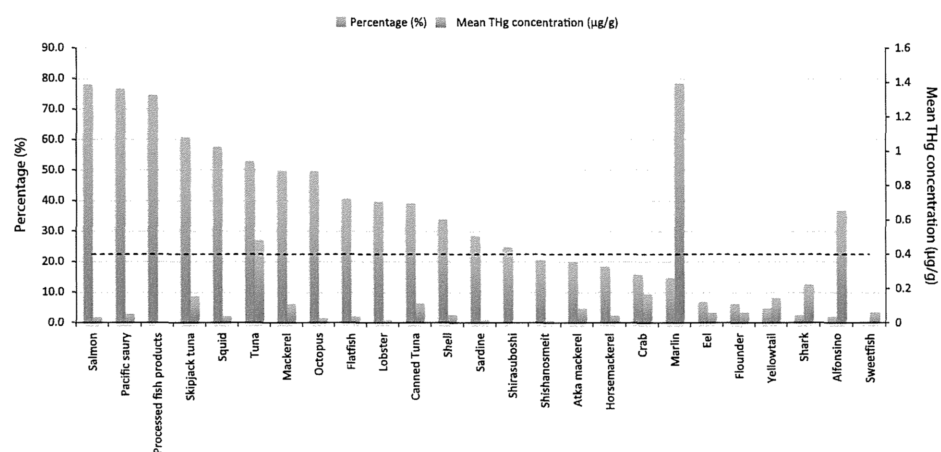

Fig. 3 Fish species commonly consumed by the survey population in Karakuwacho and the means THg concentrations of these fish species, which are cited from the newly released monitoring summary data in Japan (MHLW.2010)

Table 2 Stepwise multiple linear regression analysis between log hair mercury levels and other covariants

Model	Unstandardized coefficients		Standardized coefficients	*t*	*p*	Model summary	
	B	*SE*	*B*			Adjusted R^2	*p*
Adult female (n = 81)							
Constant	0.60	0.06	–	10.16	<0.001	0.254	<0.001
Atka mackerel	−0.13	0.06	−0.21	−2.09	0.041		
Tuna	0.15	0.05	0.31	2.94	0.004		
Octopus	0.17	0.05	0.35	3.18	0.002		
Pacific saury	−0.19	0.07	−0.31	−2.66	0.010		
Mackerel	0.12	0.05	0.24	2.24	0.028		
Squid	−0.12	0.06	−0.23	−2.02	0.047		
Children (n = 85)*							
Constant	0.26	0.05	–	5.70	<0.001	0.101	0.002
Tuna	0.21	0.07	0.34	3.16	0.002		
Adult male (n = 23)							
Constant	0.50	0.09	–	5.28	<0.001	0.337	0.008
Canned tuna	0.30	0.09	0.58	3.16	0.005		
Mackerel	0.23	0.10	0.43	2.35	0.030		

Dependent variable: log hair mercury levels. Independent variables: age, body weight, various commonly consumed fish species

SE standard error

* In this regression model, age, sex, body weight and various commonly consumed fish species were included as independent variables

dietary staple. In this study, we determined that pacific saury was negatively correlated with hair THg levels in the female adults (Table 2). According to mercury exposure monitoring data, Pacific saury had a low THg level of 0.052 ppm. Therefore, according to fish consumption patterns and mercury monitoring levels, populations can be guided to choose fish species having low levels of contamination (i.e., salmon and Pacific saury) and avoid species that may contain high mercury levels (i.e., marlin, alfonsino and tuna). Meanwhile, the total amount of fish consumption should always be considered.

In conclusion, the level of MeHg exposure is high in Karakuwacho peninsula region. Consumption of fish with high mercury contamination (e.g., tuna or canned tuna) and high consumption of fish with low mercury contamination (e.g., mackerel) are likely the major contributors. Therefore, both qualitative and quantitative aspects of fish consumption should be addressed to achieve effective reduction in MeHg exposure.

Acknowledgments This project is supported primarily by a grant in-aid for Health Sciences Research from the Ministry of Health, Labour, and Welfare of Japan (H15-Chemistry-004). We thank all participants for providing samples and those involved in sample collection.

References

Cernichiari E, Brewer R, Myers GJ, Marsh DO, Lapham LW, Cox C et al (1995) Monitoring methylmercury during pregnancy: maternal hair predicts fetal brain exposure. Neurotoxicology 16:705–710

Guallar E, Sanz-Gallardo MI, Van't Veer P, Bode P, Aro A, Gomez-Aracena J et al (2002) Mercury, fish oils, and the risk of myocardial infarction. N Engl J Med 347:1747–1754

Harada M (1995) Minamata disease: methylmercury poisoning in Japan caused by environmental pollution. Crit Rev Toxicol 25:1–24

Hightower JM, Moore D (2003) Mercury levels in high-end consumers of fish. Environ Health Perspect 111:604–608

Karagas MR, Choi AL, Oken E, Horvat M, Schoeny R, Kamai E, Cowell W, Grandjean P, Korrick S (2012) Evidence on the human effects of low-level methylmercury exposure. Environ Health Perspect 120:799–806

Kim NS, Lee BK (2010) Blood total mercury and fish consumption in the Korean 1034 general population in KNHANES III, 2005. Sci Total Environ 408:4841–4847

Knobeloch L, Anderson HA, Imm P, Peters D, Smith A (2005) Fish consumption, advisory awareness, and hair mercury levels among women of childbearing age. Environ Res 97:219–226

Koizumi A, Harada KH, Inoue K, Hitomi T, Yang HR, Moon CS, Wang P, Hung NN, Watanabe T, Shimbo S, Ikeda M (2009) Past, present, and future of environmental specimen banks. Environ Health Prev Med 14:307–318

Mahaffey K (1998) Methylmercury exposure and neurotoxicity. JAMA 280:737–738

Mahaffey KR, Clickner RP, Jeffries RA (2009) Adult women's blood mercury concentrations vary regionally in USA: association with patterns of fish consumption (NHANES 1999–2004). Environ Health Perspect 117:47–53

Maruyama K, Yorifuji T, Tsuda T, Sekikawa T, Nakadaira H, Saito H (2012) Methylmercury exposure at Niigata, Japan: results of neurological examinations of 103 adults. J Biomed Biotechnol 2012:1–7

Matthews AD (1983) Mercury content of commercially important fish of the Seychelles, and hair mercury levels of a selected part of the population. Environ Res 30:305–312

McDowell MA, Dillon CF, Osterloh J, Bolger PM, Pellizzari E, Fernando R, Montes de Oca R, Schober SE, Sinks T, Jones RL, Mhaffey KY (2004) Hair mercury levels in U.S. children and women of childbearing age: reference range data from NHANES 1999–2000. Environ Health Perspect 112:1165–1171

Mergler D, Anderson HA, Chan LH, Mahaffey KR, Murray M, Sakamoto M, Stern AH, Panel on Health Risks and Toxicological Effects of Methylmercury (2007) Methylmercury exposure and health effects in humans: a worldwide concern. AMBIO 36:3–11

MHLW (2013) Summary of investigation of mercury concentrations in fishes (in Japanese) http://www.mhlw.go.jp/shingi/2010/05/dl/s0518-8g.pdf. Accessed on 28 Mar 2013

Mozaffarian D, Rimm EB (2006) Fish intake, contaminants, and human health: evaluating the risks and the benefits. JAMA 296:1885–1899

Nakagawa R, Yumita Y, Hiromoto M (1997) Total mercury intake from fish and shellfish by Japanese people. Chemosphere 35:2909–2913

Ser PH, Watanabe C (2012) Fish advisories in the USA and Japan: risk communication and public awareness of a common idea with different backgrounds. Asia Pac J Clin Nutr 21:487–494

The Food Safety Commission, Japan, the Contaminant Expert Committee (2005) Food safety risk assessment related to methlmercury in seafood http://www.fsc.go.jp/english/topics/methylmercury_risk_assessment.pdf. Accessed on 28 Mar 2013

US EPA (2000) Guidance for assessing chemical contaminant data for use in fish advisories. Volume 2: risk assessment and fish consumption limits. http://www.epa.gov/waterscience/fish/advice/volume2/v2cover.pdf

WHO/JECFA Reports (2004) Evaluation of certain food additives and contaminants (sixty-first report of the joint FAO/WHO expert committee on food additives). WHO Technical Report Series, No. 922

Yasuda H, Yoneshiro T, Yoshida K, Shibazaki T, Ishii T, Tsutsui T (2005) High toxic metal levels in scalp hair of infants and children. Biomed Res Trace Elem 16:39–45

Yasutake A, Matsumoto M, Yamaguchi M, Hachiya N (2003) Current hair mercury levels in Japanese: survey in five districts. Tohoku J Exp Med 199:161–169

Yasutake A, Matsumoto M, Yamaguchi M, Hachiya N (2004) Current hair mercury levels in Japanese for estimation of methylmercury exposure. J Health Sci 50:120–125

Yasutake A, Nagano M, Nakano A (2005) Simple method for methylmercury estimation in biological samples using atomic absorption spectroscopy. J Health Sci 51:220–223

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

Kouji H. Harada[a], Tamon Niisoe[b], Mie Imanaka[c], Tomoyuki Takahashi[d], Katsumi Amako[e], Yukiko Fujii[a], Masatoshi Kanameishi[a], Kenji Ohse[f], Yasumichi Nakai[f], Tamami Nishikawa[f], Yuuichi Saito[f], Hiroko Sakamoto[g], Keiko Ueyama[h], Kumiko Hisaki[i], Eiji Ohara[i], Tokiko Inoue[j], Kanako Yamamoto[k], Yukiyo Matsuoka[l], Hitomi Ohata[e], Kazue Toshima[m], Ayumi Okada[n], Hitomi Sato[o], Toyomi Kuwamori[p], Hiroko Tani[p], Reiko Suzuki[q], Mai Kashikura[q], Michiko Nezu[r], Yoko Miyachi[s], Fusako Arai[t], Masanori Kuwamori[u], Sumiko Harada[v], Akira Ohmori[v], Hirohiko Ishikawa[b], and Akio Koizumi[a,1]

[a]Department of Health and Environmental Sciences and [c]Department of Preventive Medicine, Kyoto University Graduate School of Medicine, Kyoto 6068501, Japan; [b]Research Division of Atmospheric and Hydrospheric Disasters, Disaster Prevention Research Institute, Kyoto University, Uji 6110011, Japan; [d]Department of Nuclear Science and Engineering, Kyoto University Research Reactor Institute, Kumatori 5900494, Japan; [e]Faculty of Nutrition, Kobe Gakuin University, Kobe 6512180, Japan; [f]Fukushima Future Center for Regional Revitalization, Fukushima University, Fukushima 9601296, Japan; [g]Department of Food Nutrition, Kyoto Bunkyo Junior College, Uji 6110041 Japan; [h]Faculty of Human Life Sciences, Senri Kinran University, Suita, 5650873 Japan; [i]Department of Life Design, Osaka International College, Moriguchi 5708555, Japan; [j]Kyoto First Red Cross Hospital, Kyoto 6050981, Japan; [k]Aiseikai Yamashina Hospital, Kyoto 6078086, Japan; [l]National Hospital Organization Kyoto Medical Center, Kyoto 6120861, Japan; [m]Ekiya Rehabilitation Sakura, Fukuyama 7201131, Japan; [n]Best Life Promotion Ltd., Kawasaki 2118588, Japan; [o]Syojyukai Social Welfare Service Corporation Ikoinosato, Hirakata 5731161, Japan; [p]Faculty of Human Life Studies, Jin-ai University, Echizen 9150015, Japan; [q]Department of Food and Nutrition, Fukushima Gakuin Junior College, Fukushima 9600181, Japan; [r]Department of Food and Nutrition, Yamanashi Gakuin Junior College, Kofu 4008575, Japan; [s]Department of Health and Nutrition, Yonezawa Women's Junior College of Yamagata Prefecture, Yonezawa 9920025, Japan; [t]Faculty of Human Life Studies, University of Niigata Prefecture, Niigata 9508680, Japan; [u]Department of Nutrition, Mimasaka Junior College, Tsuyama 7088511, Japan; and [v]Department of Food and Nutrition, Toyama College, Toyama 9300175, Japan

Edited by Kirk R. Smith, University of California, Berkeley, CA, and approved January 22, 2014 (received for review August 21, 2013)

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 first-year 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.

Author contributions: K.H.H. and A.K. designed research; K.H.H., T. Niisoe, M.I., K.A., Y.F., M. Kanameishi, K.O., Y.N., T. Nishikawa, Y.S., H. Sakamoto, K.U., K.H., E.O., T.I., K.Y., Y. Matsuoka, H.O., K.T., A. Okada, H. Sato, T.K., H.T., R.S., M. Kashikura, M.N., Y. Miyachi, F.A., M. Kuwamori, S.H., A. Ohmori, and A.K. performed research; K.H.H., T. Niisoe, Y.F., M. Kanameishi, and A.K. analyzed data; and K.H.H., T. Niisoe, M.I., T.T., K.A., H.I., and A.K. wrote the paper.

The authors declare no conflict of interest.

This article is a PNAS Direct Submission.

Freely available online through the PNAS open access option.

[1]To whom correspondence should be addressed. E-mail: koizumi.akio.5v@kyoto-u.ac.jp.

This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10.1073/pnas.1315684111/-/DCSupplemental.

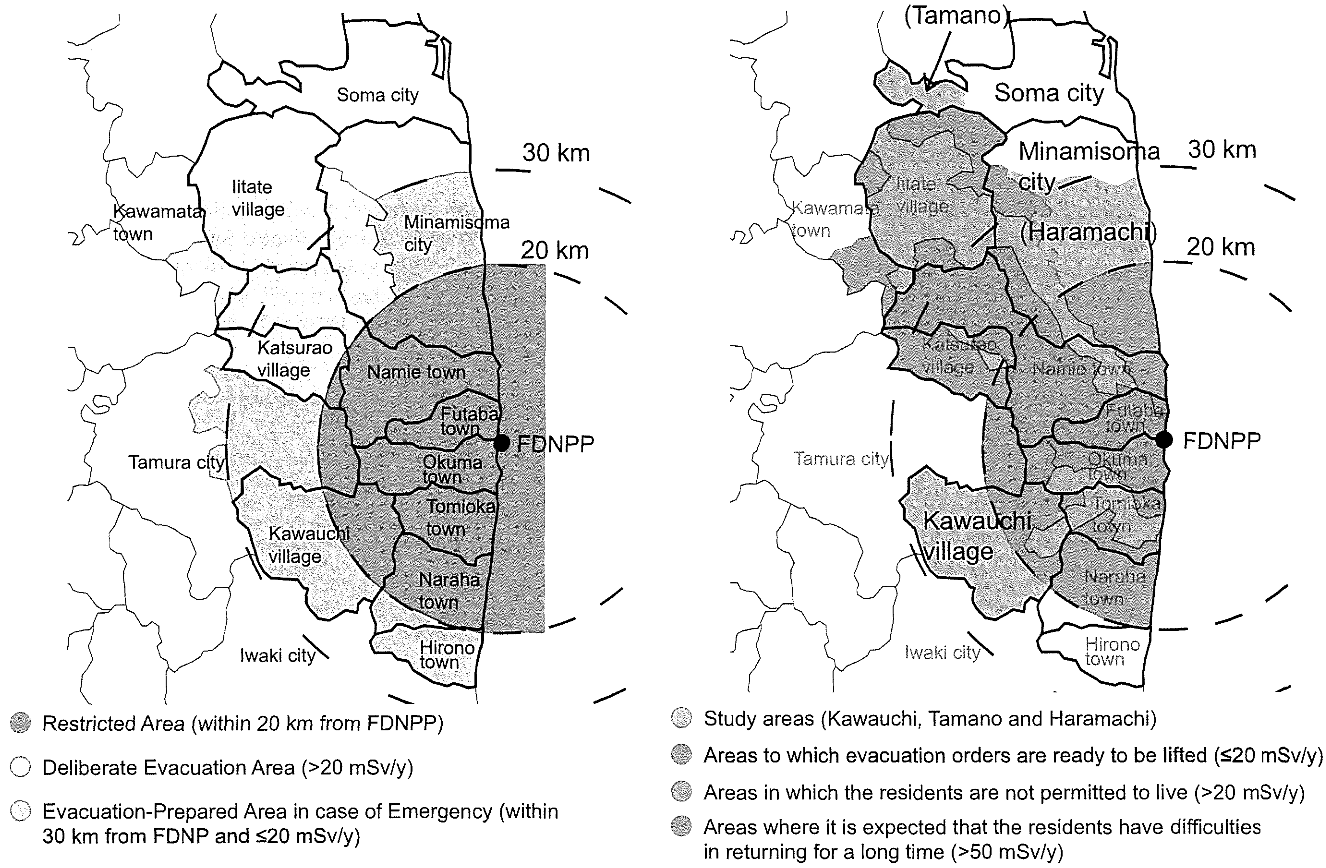

Fig. 1. Former and current statuses of the restricted area and areas to which evacuation orders have been issued around the FDNPP (*Left*, April 22, 2011; *Right*, August 8, 2013). Municipalities belonging to the Sousou region are bordered by thick lines. The three study areas are in blue.

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 ≤20 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 ^{132}Te and ^{133}Xe 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 ^{134}Cs (half-life of 2.07 y) and weathering (11); however, radiation exposure from deposited ^{137}Cs (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/m^3 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). Food-duplicate 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 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. S1*A*). 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. S1*B*). The slope factor was 0.4534 with a small intercept (0.0403 μSv/h), suggesting a shielding effect provided by buildings.

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, *n* = 382	
Range (median)	0.04–1.2 (0.14)
Mean ± 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 ± 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 ± 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).

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 ± SD) was 58.0 ± 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.

^{137}Cs and ^{134}Cs 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 ^{137}Cs and ^{134}Cs did not distribute normally. The median of ^{137}Cs 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 ^{134}Cs had a similar trend to that of ^{137}Cs among the three study groups, whereas the amount was around 60% of ^{137}Cs. The maximum estimates of the committed effective dose rate of radiocesium were 59, 120, and 58 μSv/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, ^{89}Sr, and ^{90}Sr, were analyzed in five food-duplicate samples in Kawauchi; these samples were selected in descending order starting with the sample with the highest ^{137}Cs level (0.8–3.1 Bq/kg). None of the samples contained a detectable amount of radiostrontium (^{89}Sr: <0.26 to <0.44 Bq/kg-wet; ^{90}Sr: <0.039 to <0.070 Bq/kg-wet). Ratios of ^{90}Sr to ^{137}Cs in the samples were <1.7% to <8.6%.

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 ^{137}Cs 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/m^3, fractionated dust samples contained discernible levels of radiocesium (Tables S5 and S6). Radiocesium contained in the respirable fraction (<4.9 μm) accounted for 43–77% of total activities (<100 μm). Total dust samples were also taken using

Table 2. Dietary intake of ^{134}Cs and ^{137}Cs

Area	Dietary intake, Bq/d		Estimates of committed effective dose, μSv/y*
	^{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 ± 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 ± 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 ± 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 ^{134}Cs and 0.013 μSv/Bq for ^{137}Cs). 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 ^{134}Cs and ^{137}Cs) were 0.61 ± 0.43, 0.60 ± 0.26, and 0.92 ± 0.26 mBq/m^3 in Kawauchi, the Tamano area, and the Haramachi area, respectively. The highest dose rates were estimated as 0.23 μSv/y for adults and 0.27 μSv/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 ^{137}Cs 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

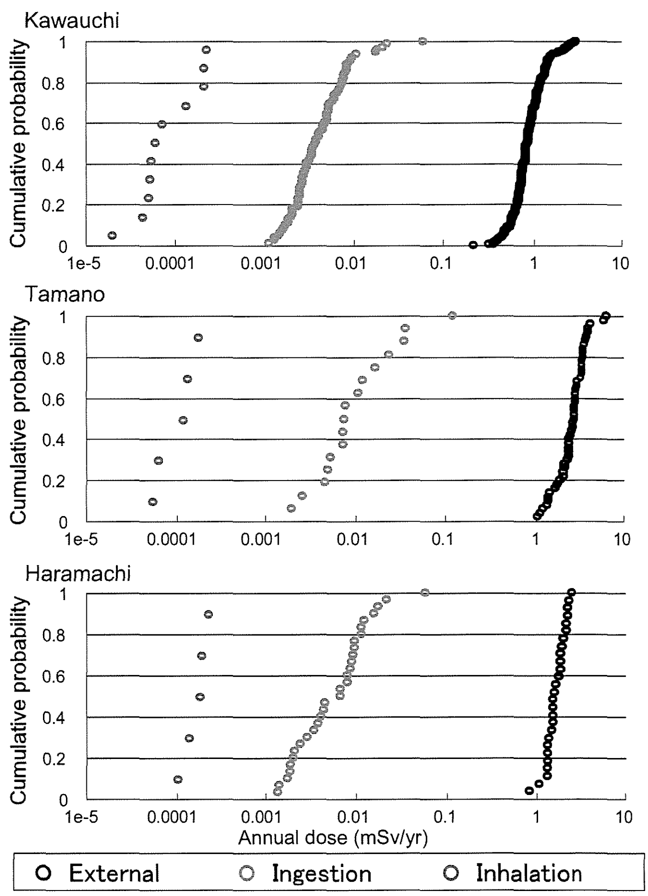

Fig. 2. Cumulative probability plot of external, ingestion, and inhalation radiation dose rates in 2012.

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 ^{210}Po and ^{210}Pb 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 10^4 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.

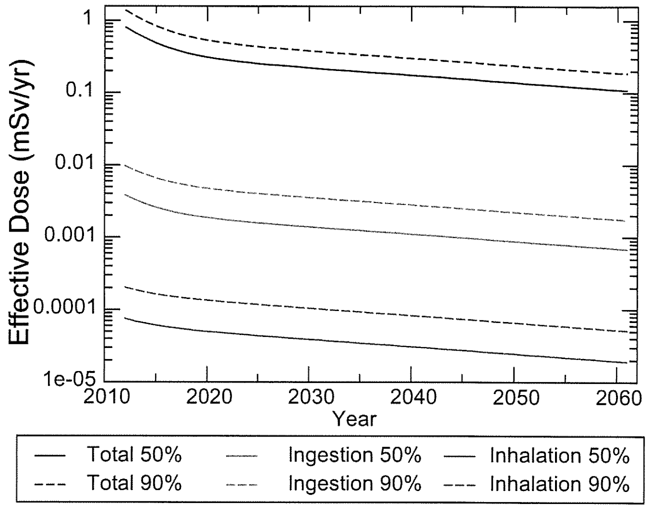

Fig. 3. Time course of total, ingestion, and inhalation radiation dose rates in the village of Kawauchi based on the physical decay of radiocesium. The 50th (solid line) and 90th percentile (dotted line) values were plotted. The total dose rate was calculated by Monte Carlo simulation. Ingestion and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

Attributable risks over a 15-y period (AR_{15}) (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. AR_{15} values for solid cancer incidence were 0.001–0.015%, and highest for 20-y-old adult females in the Tamano area. Leukemia AR_{15} was 0.001–0.009%, and highest for 1-y-old infant males in the Tamano area. AR_{15} 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 (AR_{15}/BR_{15}) 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 the migration of radiocesium

Age in 2012, y	Area	All solid cancers: Lifetime exposure, mSv	All solid cancers: LAR, % Male	All solid cancers: LAR, % Female	Leukemia: Lifetime exposure, mSv	Leukemia: LAR, % Male	Leukemia: LAR, % Female	Breast cancer: Lifetime exposure, mSv	Breast cancer: LAR, % 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.

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 ^{90}Sr and other less volatile radionuclides from the FDNPP meltdown was considered to be limited compared with that of ^{137}Cs (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 μSv/h, respectively), followed by the Kennan region (0.18 μSv/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 μSv/h to less than 0.116 μSv/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 μSv/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 ^{137}Cs 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 ^{137}Cs 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 ^{137}Cs 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 ^{137}Cs from the fallout of a nuclear test is estimated using the ^{134}Cs:^{137}Cs ratio. Ratios of ^{134}Cs:^{137}Cs 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 m^3/d) was used, whereas the International Commission on Radiological Protection reference manual gives a larger rate (22.9 m^3/d for males and 18.5 m^3/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 Iitate.

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 μSv/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 μSv/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