研究成果の刊行に関する一覧表

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Letter

Internal exposure to ^{210}Po and ^{40}K from ingestion of cooked daily foodstuffs for adults in Japanese cities

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ABSTRACT — The isotope ^{210}Po was suspected of being involved in the death of a former Russian intelligence agent in 2006 in the UK. Although human exposure to this natural radionuclide in foods is estimated to be high, few studies are available. UNSCEAR Report 2000 does not contain data on ^{210}Po concentrations of foodstuffs in Japan. We analyzed samples of the everyday Japanese diet cooked with foodstuffs purchased at supermarkets in 7 major domestic cities in 2007-2008. ^{210}Po was quantified by alpha spectrometry and natural radionuclides such as ^{40}K by gamma spectrometry. The daily intake and committed effective dose of ^{210}Po, ^{40}K, and other natural radionuclides for Japanese adults were calculated. Daily intake was 0.34-1.84 (mean ± σ: 0.66 ± 0.53) and 68.5-94.2 (81.5 ± 8.5) Bq/d and the committed effective dose was 0.15-0.81 (0.29 ± 0.24) and 0.16-0.21 (0.18 ± 0.02) mSv for ^{210}Po and ^{40}K, respectively, comprising a high percentage of the total exposure. The total of the mean committed effective dose for the two nuclides (0.47 mSv) was higher than the annual effective dose from ingestion of foods reported by UNSCEAR 2000 (0.29 mSv). The mean committed effective dose of ^{40}K in the 7 major Japanese cities was comparable to the global average (0.17 mSv). The dietary exposure of Japanese adults can be characterized by a higher ^{210}Po contribution than in other countries. Of the total daily dietary ^{210}Po exposure (13 food categories excluding water) for adults in Yokohama, about 70% was from fish/shellfish and 20% from vegetables/mushrooms/seaweeds, reflecting preferences of Japanese to eat a considerable amount of fish/shellfish containing high ^{210}Po concentrations.

Key words: ^{210}Po, ^{40}K, Alpha spectrometry, Foodstuff, Intake, Dose

INTRODUCTION

Polonium belongs to Group 16 of the periodic table, and is considered to resemble Se and Te of the same group in terms of chemical properties. ^{210}Po is produced during the disintegration of ^{238}U, and is found widely in nature. ^{210}Po has a long half-life (138.4 d) among natural isotopes of Po. The biological half-life of Po is about 50 days. The specific radioactivity of ^{210}Po is 1.66 x 10^{14} Bq/g, 10 billion times higher than that of ^{238}U. ^{210}Po emitted only alpha particles and causes high levels of exposure inside the human body if ingested. The unexplained death of a former officer of the Federal Security Service of the Russian Federation, which took place in November 2006 in the United Kingdom, has been pointed out to be possibly an outcome of assassination, because ^{210}Po was highly detected in the dead officer's urine (Stather, 2007). Regarding previous experiments conducted on the toxic-

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ity of ^{210}Po, there is a published report on the subacute toxicity of ^{210}Po, demonstrating that all rats intravenously treated with ^{210}Po (1.45 MBq/kg body weight) died 14-44 days after administration (Rencová *et al.*, 1997). If this dose (1.45 MBq/kg body weight) is converted into mass, based on ^{210}Po's specific radioactivity (1.66 x 10^{14} Bq/g), the lethal dose of ^{210}Po is 8.7 ng/kg body weight (amount of ^{210}Po injected intravenously). This means that the ingestion of 609 ng of Po by an adult male weighing 70 kg can result in subacute death. ^{210}Po is known to show high-level toxicity. This lethal dose of ^{210}Po is much smaller than the lethal dose (0.15 g) of potassium cyanide, one of the chemical compounds known to have the highest level of toxicity. The review of health risk evaluations for ingestion exposure of humans to ^{210}Po was reported (Scott, 2007). ^{210}Po found in nature can affect human exposure to radiation if it is ingested from food, inspired on cigarette smoking, or is inhaled together with radon released into residential areas. The ingestion of food and beverages is considered the most important route of human exposure to ^{210}Po. Also, K is usually contained in food. The percentage made up by the natural radionuclide ^{40}K, one of the isotopes of K, is 0.0117%. According to a report from the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000) on the sources of ionizing radiation and its effects, the exposure of humans to radiation through food ingestion is primarily attributable to natural radionuclides represented by ^{40}K and those of uranium and thorium series, with their worldwide average effective dose being 0.29 mSv/y (range: 0.2-0.8 mSv/y). That report provides information on the radioactivity level of natural nuclides of the uranium and thorium series (including ^{210}Po) contained in food and drinking water as well as their annual level of intake. However, the report does not refer to the ^{210}Po concentrations in food or drinking water consumed in Japan. Studies have been conducted in Japan on the level of ^{226}Ra and ^{210}Pb in food and the intake of ^{232}Th and ^{238}U by Japanese people (Kametani *et al.*, 1981; Shiraishi *et al.*, 1995, 2000), but very few such studies pertaining to ^{210}Po have been published. It is known that marine products usually contain ^{210}Po at relatively high concentrations. A representative study conducted in Japan was designed to evaluate the amount of ^{210}Po ingested through analysis of the data on ^{210}Po in fish and shellfish collected primarily in the north-east region of Japan (Yamamoto *et al.*, 1994). In that study, analysis and evaluation were confined to fish and shellfish caught in limited areas around Japan. After that report, no comprehensive evaluation of ^{210}Po radioactivity and dietary intake in Japan has been conducted. Very recently, however, a report was published concerning the effective dose in adults evaluated on the basis of data on 8 nuclides including ^{210}Po contained in foodstuffs purchased in 11 prefectures of Japan (Ota *et al.*, 2009). Following such previous studies, the present study was undertaken, classifying foodstuffs into 14 categories and analyzing ordinary foods cooked from materials in each category purchased (except for drinking water) at markets in 7 major cities of Japan. These food samples were subjected to the analysis of ^{210}Po by alpha spectrometry and determination of gamma-emitting nuclides (^{40}K and others) by gamma spectrometry. On the basis of the data thus collected, the dose conversion coefficients of International Commission on Radiological Protection (ICRP, 1996) and food consumption data in each city, we evaluated the daily intake and committed effective dose of radionuclides (^{210}Po, ^{40}K and others) for adults in Japan.

MATERIALS AND METHODS

Sample

The foods studied were divided into 14 categories including drinking water, referring to the results of the national health and nutrition surveys in 2002-2004 (Ministry of Health, Labor and Welfare, 2003; 2004; 2005): (1) Rice; (2) Grains, potatoes, seeds and nuts; (3) Sugar and preserves, sweets; (4) Fats and oils; (5) Legumes; (6) Fruits; (7) Green, yellow vegetables; (8) Other vegetables, mushrooms, and seaweeds; (9) Beverages; (10) Fish and shellfish; (11) Meat and poultry, eggs; (12) Milk and dairy products; (13) Seasonings and spices; and (14) Drinking water. The first 13 food categories were purchased in 2007-2008 at supermarkets in 7 major cities of Japan (Sapporo, Sendai, Niigata, Yokohama, Osaka, Kochi, and Fukuoka). The number of foodstuffs purchased in each city as samples for this study ranged from 153 to 174. Drinking waters were sampled from tap water supplied to households in each city. Each category of food purchased was cooked by boiling, frying, baking, etc., referring to the data on amounts consumed for each food category (Ministry of Health, Labor and Welfare, 2003; 2004; 2005) to prepare total dietary samples, reproducing the typical daily dietary menu of Japanese people. When foods were cooked, we avoided using additives or mixing one food category with another. For the evaluation of the total exposure level to ^{210}Po, samples of cooked foods from all 13 categories were combined at a ratio corresponding to the ratio of the consumed amount for each category in each city, referring to the data on amounts consumed for each category, to yield mixed food samples. In addition, to evaluate the contribution of each of the 13 food categories (excluding drinking water) to

the ^{210}Po exposure level, separate samples of each of the 13 food categories in Yokohama were prepared. All of these samples were tested without further processing after they were initially cooked. For the evaluation of exposure to gamma-emitting nuclides, those individually cooked samples of each of the 13 food categories were freeze- or heat-dried, followed by 24-hr ashing at 450°C, to yield test samples. Drinking water (100 l) was evaporated to dryness for test samples.

Radioanalytical procedure of ^{210}Po

Mixed samples were analyzed to evaluate the amount of ^{210}Po ingested in all foodstuffs. The ^{210}Po were analyzed by using Sr-Spec™ (Eichrom Technologies, Lisle, IL, USA) resin column with an alpha spectrometry (Miura *et al.*, 2003). From the sample for analysis, 30 g (wet weight) was taken and spiked with ^{209}Po tracer for the correction of the chemical yield. The mixture was digested with conc. HNO_3, concentrated followed by degradation. After degradation, the solution was concentrated and filtrated. The filtrate was concentrated by heating, digested with 4 M HCl, and then heated. After cooling, the residue was filtrated, and the filtrate was passed through Sr-Spec™ was followed by eluting with 6 M HNO_3, and the eluate was concentrated by evaporation. The concentrated eluate was digested with 0.5 M HCl and dissolved by heating. The thus prepared test solution was combined with ascorbic acid, and Po was electrodeposited on a stainless steel disk. The stainless steel disk was washed with distilled water and ethanol and dried to yield samples for measurement. The samples were subjected to measurement for 80,000-160,000 s with an alpha spectrometry system equipped with the Si detector (Canberra, Meriden, CT, USA). Fig. 1 shows the alpha spectrum isolated from a mixed food sample. On the basis of the net reading from each sample, the counts of tracer ^{209}Po for the yield correction and the amount of sample subjected to analysis, the ^{210}Po radioactivity level was calculated. The results of analysis were corrected for decay time according to the date of sample preparation. The average chemical recovery of ^{209}Po in this method was found to be 74%. When this analytical method was applied to the reference sample-sediment (IAEA-368), the ^{210}Po analytical value of 24 ± 1.6 Bq/kg was identical to the reference value (23.2 Bq/kg).

Individual food category samples were analyzed to evaluate the amount of ^{210}Po intake derived from each food category. The radioanalytical procedure was without an electrodepositing technique (Tolmachyov, 2001). From each food category, 30 g of cooked sample was taken (freeze-dried samples equivalent to 30 g of cooked samples for fish/shellfish, meats and eggs) and spiked with ^{208}Po tracer for the yield correction. The samples were digested with conc. HNO_3 and H_2O_2. The sample solutions were concentrated to a level close to drying, and then decomposed with 10 ml of conc. HCl. It was then concentrated to a level close to drying. The sample solutions were subsequently decomposed with 50 ml of 0.5 M HCl and heated. If a residue was formed, the solutions were filtrated. A silver disk with a Teflon tape attached to one side and ascorbic acid were added to the filtrate in a plastic bottle, followed by Po sedimentation on the silver disk. The silver disks were washed with distilled water and ethanol, followed by drying to yield samples for measurement. The samples were subjected to measurement for 200,000 s with an alpha spectrometry system equipped with the Si detector (EG&G Ortec, Oak Ridge, TN, USA). On the basis of the net reading from each sample, the counts of tracer ^{208}Po for yield correction, and the amount of sample subjected to the analysis, the level of ^{210}Po radioactivity in the prepared sample was calculated. The results of analysis were corrected for decay time according to the date of sample preparation. The average chemical recovery of ^{208}Po in this method was found to be 72%.

Measurement of gamma-emitting nuclides

Individual food category samples were subjected to measurement to evaluate the amount of ^{40}K and other gamma-emitting nuclides ingested in each food category. Ash samples from each food category were placed individually into cylindrical plastic containers (100-ml capacity) and sealed with a silicone sealing. The containers filled with samples were left to stand for about 2 weeks, to yield samples for measurement. Because fats and oils are difficult to ash, they were placed into Marinelli containers (1-L capacity) to yield samples for measurement. Each sample was subjected to determination for 80,000-300,000 s with a gamma spectrometry system equipped with the high-purity Ge detector (Canberra, Eurysis, or Oxford). The nuclides measured were natural radionuclides (^{40}K, ^{214}Pb, ^{214}Bi, ^{212}Pb, ^{228}Ac and ^{208}Tl) and artificial radionuclides (^{137}Cs). The reliability of data yielded from this gamma spectrometry was certified in a previous report (Sugiyama *et al.*, 2007).

RESULTS

The amount of daily intake of ^{210}Po from ingestion of the foods for adults in 7 major cities of Japan was investigated. Daily ^{210}Po intake was calculated from the analytical data on each mixed sample of 13 food categories

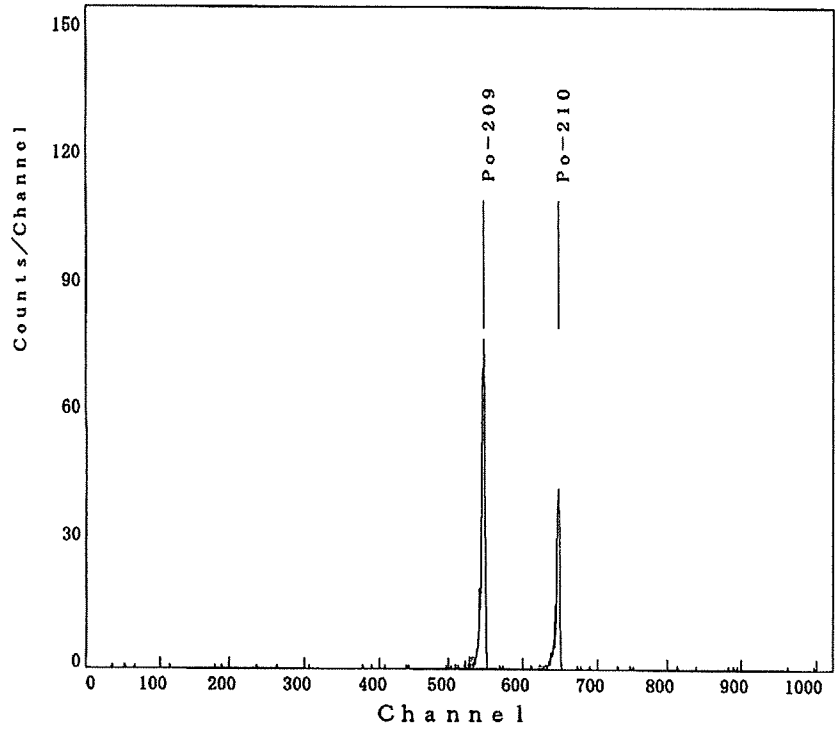

Fig. 1. Alpha spectrum of polonium separated from a mixed food sample.
5.305 MeV for ^{210}Po (half life; 138.4d), 4.877 MeV for ^{209}Po (half life: 102y).

excluding drinking water and data on food consumption in each city. Table 1 shows the results in the present study and data reported in the literature. The amount of daily ^{210}Po intake by adults in the 7 cities ranged from 0.34 to 1.84 (mean ± σ: 0.66 ± 0.53) Bq/d. Levels showed an ascending order of 0.34 in Fukuoka, 0.37 in Yokohama, 0.38 in Sapporo, 0.45 in Osaka, 0.58 in Sendai, 0.69 in Niigata, and 1.84 in Kochi. It was thus largest in Kochi.

The amount of daily intake of each of the 13 food categories excluding drinking water for ^{210}Po was investigated in Yokohama. The amount of ^{210}Po intake in each of the 13 food categories was calculated on the basis of analytical data and data on food consumption in Yokohama. ^{210}Po was determined for 5 food categories (other vegetables, mushrooms, and seaweeds; beverages; fish and shellfish; meat and poultry, eggs; and seasoning and spices). The detection limit of ^{210}Po for each food categories was 0.05, 0.04, 0.04, 0.03, 0.04, 0.03, 0.04, 0.07, 0.006, 1.4, 0.04, 0.02, 0.06, and 0.002 mBq/g in order according to the category. The intake of ^{210}Po in these five food categories was 37.5, 3.8, 130.0, 2.2 and 18.7 mBq/d, respectively. When the percentage of each food category among the total daily ^{210}Po intake was evaluated, as illustrated in Fig. 2, it was shown that about 70% of the total ^{210}Po was derived from fish and shellfish and 20% from other vegetables, mushrooms, and seaweeds.

The amount of daily intake of gamma-emitting radionuclides was calculated on the basis of data from the measurement of individual samples of 14 food categories as well as data on food consumption. K is generally abundant in food. The percentage occupied by the natural radionuclide ^{40}K, one of the isotopes of K, is 0.0117%. It is known that foods contain high levels of ^{40}K radioactivity. In the present study, ^{40}K in all food categories excluding some fats and oils was determined. Table 1 shows the total daily intake of ^{40}K in 14 food categories for adults in 7 cities. The total intake of ^{40}K in 14 food categories for adults in 7 major cities of Japan (Bq/d) was 68.5 in Fukuoka, 78.2 in Sendai, 78.7 in Kochi, 79.4 in Osaka, 81.4 in Sapporo, 90.5 in Yokohama, and 94.2 in Niigata. It averaged 81.5 Bq/d. When analyzed for each food category, the amount of ^{40}K intake in each of the 7 cities was particularly large from other vegetables, mushrooms and seaweeds (Category VIII), at 8.8-29.1 Bq/d. After

Table 1. Comparison of daily intake and effective dose of ^{210}Po and ^{40}K from foods with literature values

Sample	^{210}Po inatake (Bq/d)	^{210}Po effective dose (mSv)	^{40}K intake (Bq/d)	^{40}K effective dose (mSv)	Region	Reference
Daily diet	0.38	0.17	81.4	0.18	Sapporo, Japan	Present study
	0.58	0.25	78.2	0.18	Sendai, Japan	Present study
	0.69	0.30	94.2	0.21	Niigata, Japan	Present study
	0.37	0.16	90.5	0.20	Yokohama, Japan	Present study
	0.45	0.19	79.4	0.18	Osaka, Japan	Present study
	1.84	0.81	78.7	0.18	Kochi, Japan	Present study
	0.34	0.15	68.5	0.16	Fukuoka, Japan	Present study
	0.11, 0.22	0.06, 0.11	-	-	India	Avadhani *et al.* (2001)
	0.08, 0.19	-	-	-	India	Narayana *et al.* (1995)
	0.05-0.06	-	-	-	USA	Holtzman *et al.* (1980)
Foodstuff	1.67 (610 Bq/y)	0.73	-	-	Japan	Ota *et al.* (2009)
	-	-	39.4	0.10	Korea	Choi *et al.* (2008)
Seafood	0.48-0.69	-	-	-	Japan	Yamamoto *et al.* (1994)
	1.20	-	-	-	Portugal	Carvalho *et al.* (1995)
	0.14 (52 Bq/y)	-	-	-	International	Aarkrog *et al.* (1997)
Diet	0.05-0.36 (18-130 Bq/y)	0.022-0.16	-	0.17	International	UNSCEAR (2000)
	0.60 (220 Bq/y)	0.27			Japan	UNSCEAR (2000)

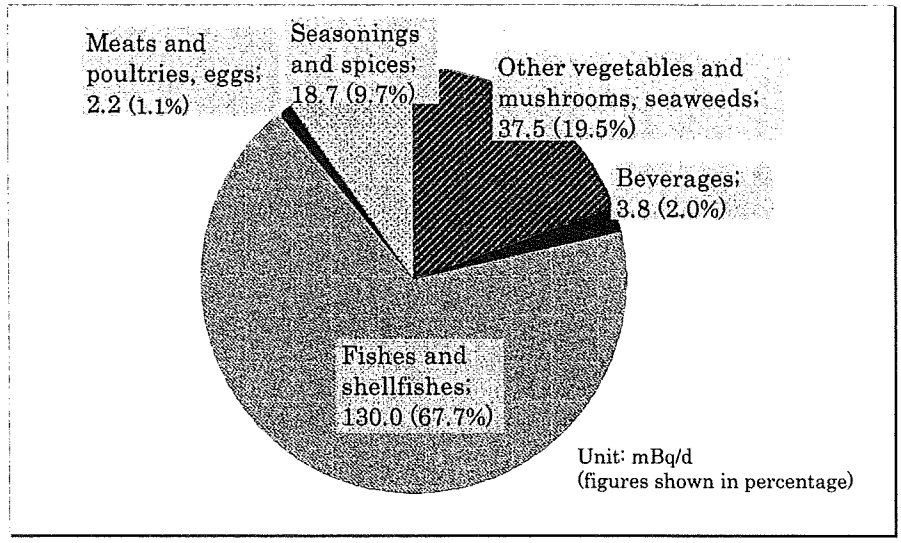

Fig. 2. Daily intakes of ^{210}Po from fourteen food categories for adults in Yokohama city, Japan.

this category, the amount was large from grains/potatoes/seeds, green/yellow vegetables, meat/poultry/eggs and fish/shellfish. The levels of nuclides belonging to the uranium and thorium series (^{214}Pb, ^{214}Bi, ^{212}Pb, ^{228}Ac, and ^{208}Tl) were less than the detectable limit or very low. The amount of these nuclides ingested daily was very small, with the maximum being: ^{214}Pb < 0.14 Bq/d, ^{214}Bi < 0.16 Bq/d, ^{228}Ac < 0.32 Bq/d and ^{212}Pb < 0.13 Bq/d. ^{208}Tl was not detected in any food tested. ^{137}Cs (an artificial radionuclide originating from past atmospheric nuclear weapon tests and the accident at the Chernobyl nuclear power plant) was found to have been ingested with fish/shellfish, meat/poultry/eggs, and other vegetables/mushrooms/seaweeds, with the maximum amount daily intake being 0.03 Bq/d (Osaka).

The committed effective dose of ^{210}Po and ^{40}K from ingestion of foods for adults was calculated from the amounts of daily ^{210}Po and ^{40}K intake. In the present study, a relatively simple method, usually used for environmental radioactivity monitoring around atomic power facilities, was employed for dose evaluation. The method used for calculation was described elsewhere (Sugiyama *et al.*, 2007). The dose conversion coefficients for adult persons (^{210}Po: 1.2×10^{-6} Sv/Bq, ^{40}K: 6.2×10^{-9} Sv/Bq) were obtained form ICRP Publication 72 (ICRP, 1996). The results are shown in Table 1. The committed effective dose (mSv) of ^{210}Po for adults in the 7 cities was as follows (in ascending order): 0.15 in Fukuoka, 0.16 in Yokohama, 0.17 in Sapporo, 0.19 in Osaka, 0.25 in Sendai, 0.30 in Niigata, and 0.81 in Kochi. The mean ± σ of the 7 cities was 0.29 ± 0.24 mSv. Thus, the committed effective dose in adults was higher in Kochi than the other 6 cities. When the committed effective dose from individual food categories in Yokohama was calculated, it was high in the descending order of fish/shellfish, other vegetables/mushrooms/seaweeds, seasoning/spices, beverages and meat/poultry/eggs, similar to the order of the amount of daily ^{210}Po intake in this city. The committed effective dose (mSv) of ^{40}K was as follows (in ascending order): 0.16 in Fukuoka, 0.18 in Sapporo, 0.18 in Sendai, 0.18 in Osaka, 0.18 in Kochi, 0.20 in Yokohama, and 0.21 in Niigata. The mean ± σ for the 7 cities was 0.18 ± 0.02 mSv. Thus, this amount differed little among the 7 cities. Fig. 3 shows the contribution of 14 food categories for the committed effective dose to the daily intake of ^{40}K for adults of 7 cities in Japan. As shown in Fig. 3, although, the values of the committed effective dose excluding those of other vegetables/mushrooms/seaweeds (Category VIII) among 7 cities were relatively similar levels (0.13-0.16 mSv), but it was found that the contributions of the ^{40}K dose of Category VIII to those doses showed a very high rate (14.7-44.6%). As described in the evaluation of the amount of daily intake, the level of radioactivity of other natural radionuclides (^{214}Pb, ^{214}Bi, ^{212}Pb, ^{228}Ac, and ^{208}Tl) excluding ^{40}K was low in each food category, and the committed effective dose was also low, with the maximum effective dose (μSv) being: ^{214}Pb < 0.002, ^{214}Bi < 0.007, ^{212}Pb < 0.279, and ^{228}Ac < 0.051. Dose calculation

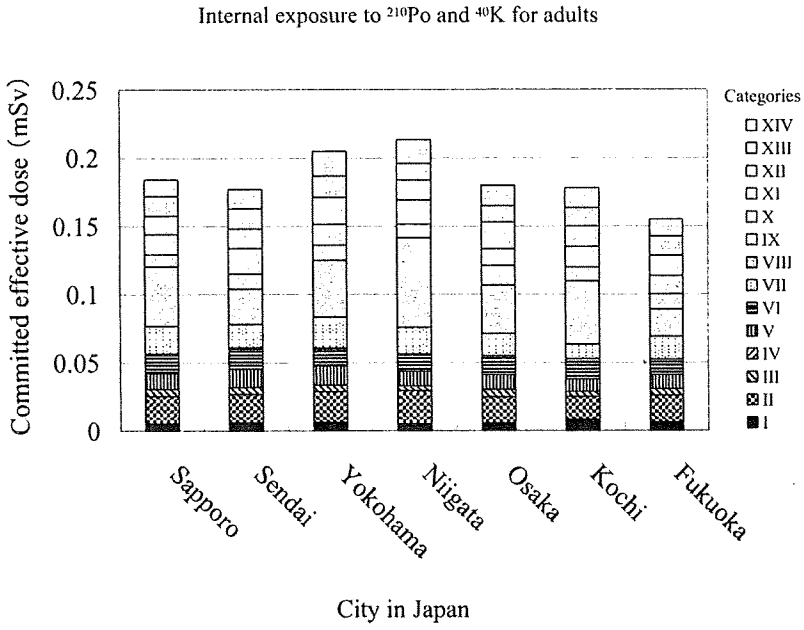

Fig. 3. Contribution of fourteen food categories for committed effective doses to intakes of ^{40}K for adults of seven cities in Japan. I: Rice; II: Grains, potatoes, seeds and nuts; III: Sugar and preserves, sweets; IV: Fats and oils, V: Legumes; VI: Fruits; VII: Green, yellow vegetables; VIII: Other vegetables, mushrooms, seaweeds; IX: Beverages; X: Fish and shellfish; XI: Meat and poultry, eggs; XII: Milk and dairy products; XIII: Seasoning and spices; XIV: Drinking water.

for ^{208}Tl was not undertaken because ^{208}Tl was not detected in any food tested. In the present study, the contribution of natural radionuclides excluding ^{210}Po and ^{40}K to the human exposure level was shown to be very small.

DISCUSSION

At present, data on the ^{210}Po radioactivity level in foods are scant in both Japan and other countries. Only a limited number of reports are available on evaluation of the ^{210}Po exposure level. UNSCEAR 2000 contains reference data on ^{210}Po levels contained in foods (15-2,000 mBq/kg). According to that report, the ^{210}Po level is the highest in fish products and the level is below 100 mBq/kg in leafy vegetables, grains, meats, root vegetables/fruits, and dairy products. Examples of reports showing high ^{210}Po levels in marine products are cited below. *Cerastoderma glaucum* and *Tapes Philippinarum*, which are seafoods collected in Venice, Italy, had ^{210}Po levels of 51.6-108 Bq/kg and 37.3-72.0 Bq/kg fresh weight, respectively (Jia *et al.*, 2001). Crops harvested in Syria had ^{210}Po levels < 0.1-15 Bq/kg dry weight, with the highest level recorded in parsley (Othman and Yassine, 1995), clearly lower than the levels of the above-mentioned marine products. According to a study in Goa, India, the ^{210}Po level was higher for fishes and prawn, at 1.60-9.65 Bq/kg wet, as compared to vegetables and rice (0.05-0.26 Bq/kg wet) (Avadhani *et al.*, 2001). In the same study, the level in leafy vegetables was higher than that in other vegetables. Representative data from Japan pertain to the analysis of ^{210}Po in fish and shellfish collected in the north-east region of Japan (Yamamoto *et al.*, 1994). In that study, the ^{210}Po level was 0.6-26.0 Bq/kg wet in fishes and 0.5-220 Bq/kg wet in molluscs, echinoderms and chordatas. Among others, the internal organs of shellfish had high levels of ^{210}Po, and the level tended to be higher in fish such as *Pneumaotophous japonics*, *Thunnus thynnus*, *Sardinops melanosticus*, and *Hippoglossoides dubius* (14.4-25.8 Bq/kg wet). The level in algae was not very high (2.8-4.3 Bq/kg wet). Recently, new data were reported on ^{210}Po contained in foodstuffs distributed in Japan. According to that report, the ^{210}Po level was high in fish and shellfish (0.02-120 Bq/kg fresh weight), and it was particularly high in dried sardines (Ota *et al.*, 2009). In the present study, ^{210}Po in each food category was analyzed using samples from Yokohama, revealing that the ^{210}Po level was highest in fish/shellfish (1.60 Bq/kg), followed by seasoning/spices (0.20 Bq/kg) and other vegetables/mushrooms/seaweeds (0.19 Bq/kg). In Osaka, the ^{210}Po level was 5.40 Bq/kg in fish/shellfish and 0.12 Bq/kg in other vegetables/mushrooms/seaweeds. These results indicate that the ^{210}Po level in fish/shellfish is higher than that in other foodstuffs in

Japan, similar to the findings reported from overseas. The amount of annual ^{210}Po intake (UNSCEAR, 2000) is larger in Japan (220 Bq/y, equivalent to 0.6 Bq/d) than in any other country (22 in USA, 18 in Argentina, 68-130 in China, 40 in Italy, 44 in Poland, 40-55 in Russia, and 28-44 Bq/y in the UK). In the present study, the amount of daily ^{210}Po intake was 0.34-1.84 Bq/d (mean: 0.66), approximately equal to the amount reported by UNSCEAR 2000. On evaluation of the amount of daily ^{210}Po intake from each of 13 food categories in Yokohama, the contribution was the greatest from fish/shellfish (68% of the total intake). This percentage is much higher than that reported for seafood in Ireland (45%) yielded by a similar evaluation of the contribution of each food category (RPII.: 2008). Yamamoto reported that the amount of ^{210}Po ingested in fish/shellfish caught in the north-east region of Japan was 0.48-0.69 Bq/d, indicating high levels of exposure to ^{210}Po from them. The large amount of daily ^{210}Po intake by Japanese people seems to reflect their dietary habits, characterized by a larger consumption of fish/shellfish, which tend to contain high ^{210}Po levels, as compared to people in Western countries.

The survey results on ^{40}K levels in foods are presented. According to the above-mentioned study in Syria, the ^{40}K level in agricultural products was 128-1,800 Bq/kg dry weight, with the highest level recorded in marrow (*Cucurbita pepo*) (Othman and Yassine, 1995). In a survey of natural radionuclide levels in 5 kinds of fish/shellfish caught in the north-east region of Japan, the levels of ^{40}K were 61.9-119 Bq/kg wet (Yamamoto *et al.*, 1994). Thus, these two studies yielded similar results, as already pointed out above. A study on natural radionuclides conducted in Korea in 1998-2000 and 2005 involved the detailed analysis of daily ^{40}K intake. In that study, representative Korean foodstuffs were divided into 8 categories, and the total amount of daily ^{40}K intake was calculated on the basis of the results from the analysis of individual food categories and the amount of food consumed. The amount of daily ^{40}K intake was 39.41 Bq/d, accounting for an overwhelming majority of the total daily intake per day (39.46 Bq/d). The contribution to daily ^{40}K intake was highest with rice (5.72 Bq/d) among all foodstuffs analyzed. The amount of daily ^{40}K intake in Korea was smaller than that in other countries or shown in the ICRP report (ICRP, 1975; Choi *et al.*, 2008). In that survey, ^{210}Po was not evaluated.

In the present study, the level of ^{40}K intake by Japanese adults was 68.5-94.2 (mean: 81.5) Bq/d, which was much larger than the levels in Korea. The amount of ^{40}K intake was particularly large from other vegetables/mushrooms/seaweeds (Category VIII; 8.8-29.1 Bq/d), followed by grains/potatoes/seeds/nuts, and green/yellow vegetables, differing from the tendency seen in Korea. The total of committed effective dose of ^{210}Po and ^{40}K in 7 cities of Japan was 0.47 mSv. This dose is higher than the global average of effective dose (0.29 mSv/y), but is within the typical range (0.2-0.8 mSv/y; UNSCEAR, 2000). The mean for ^{210}Po was 0.29 mSv, which was much higher than the global average for the uranium and thorium series (0.12 mSv/y). The mean dose of ^{40}K in the 7 Japanese cities was 0.18 mSv, which was comparable to the global average (0.17 mSv/y; UNSCEAR, 2000). We may therefore say that the large contribution of ^{210}Po is a characteristic of the dietary exposure for adult Japanese. In a similar way, a recent report showed that the effective dose of 8 nuclides (^{238}U, ^{232}Th, ^{226}Ra, ^{210}Pb, ^{210}Po, ^{90}Sr, ^{137}Cs and $^{239+240}$Pu) derived from the diet among adult Japanese was higher than the value shown in UNSCEAR 2000, and that ^{210}Po contained in marine products made a large contribution (Ota *et al.*, 2009). A study conducted in Goa on vegetarian and non-vegetarian meal is noteworthy. For the vegetarian meal, the ^{210}Po level was 0.08 Bq/kg wet and the committed effective dose was 49.1 μSv/y, while the non-vegetarian meal had a ^{210}Po level of 0.18 Bq/kg wet and a committed effective dose of 94.6 μSv/y (Avadhani *et al.*, 2001). In the above-mentioned study in Korea, the total annual internal dose from ingestion of food samples was 110 μSv/y, of which 101 μSv/y came from ^{40}K. This value was lower than that in Japan.

These results indicate that differences in dietary habits affect the exposure level to two major natural radionuclides (^{210}Po and ^{40}K). Some of the previous reports involved evaluation of the exposure level on the basis of the analysis of foodstuffs, on the other hand, in the present study, ^{210}Po and ^{40}K radioactivity levels were determined from dietary food samples (prepared by cooking foodstuffs), thus yielding data on exposure levels close to the levels prevailing under practical settings. The findings of a large amount of daily ^{210}Po intake and high committed effective dose can be viewed as reflecting dietary habits unique to Japanese people, who prefer fish and shellfish. To make the relationship between the kind of food and level of ^{210}Po intake or exposure level more clear, it would be desirable to conduct this kind of analysis for each food category involving more survey areas in Japan.

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