

Contents and Daily Intakes of Gamma-Ray Emitting Nuclides, ^{90}Sr , and ^{238}U using Market-Basket Studies in Japan

Hideo Sugiyama,^{*,a} Hiroshi Terada,^a Mitsuko Takahashi,^a Ikuyo Iijima,^b and Kimio Isomura^c

^aDepartment of Environmental Health, National Institute of Public Health, 2-3-6, Minami, Wako-shi, Saitama 351-0197, Japan,

^bChemistry Division, Kanagawa Prefectural Institute of Public Health, 1-3-1, Shimomachiya, Chigasaki-shi, Kanagawa 253-0087, Japan, and ^cAtmospheric Environment Division, Hyogo Prefectural Institute of Public Health and Environmental Sciences, 2-1-29, Arata, Hyogo-ku, Kobe-shi, Hyogo 652-0032, Japan

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To investigate the contents of radionuclides in foods marketed in Japan and their daily intakes and exposure doses in adults, we performed market-basket studies concerning radionuclide intakes. The study period was 2003–2005, and the studies were performed in 13 cities in Japan. Foods including drinking water were divided into 14 food groups, and samples were prepared by common cooking procedures. γ -ray emitting nuclides (an artificial radionuclide, radioactive Cs, and natural radionuclides, ^{40}K and U series such as ^{214}Bi , and ^{212}Pb , and Th series) were measured in each food group, and artificial radionuclides, ^{90}Sr and ^{238}U , were measured in a mixed sample of 13 food groups excluding drinking water. The daily intakes in adults were calculated from the concentrations of the radionuclides and mean daily consumption of foods and drinking water. The daily ^{137}Cs and ^{40}K intakes (mBq/person · day) in the 13 cities were 12.5–<79.7 and 57309–95746, respectively. The ^{90}Sr intake from the food groups excluding drinking water was 20.8–53.6, with a mean of 39.2 (mBq/person · day) (deviation of the mean: 23%). Similarly, the daily ^{238}U intake was 5.9–31.1, with a mean of 12.6 (mBq/person · day) (deviation: 60%), showing a more than 5-fold difference between the minimum and maximum values, and there were regional differences. Since the contents of the U series, such as ^{214}Bi and ^{212}Pb , and Th series were lower than the lower detection limits in many samples, their daily intakes were not calculated. Regarding the daily intake of ^{137}Cs from each food group, the intakes from fish and shellfish, milk, meat/eggs, and mushrooms/seaweed tended to be higher. The daily ^{40}K intake from each food group varied among the areas, but the total intake from the 14 food groups was similar in all 13 cities. ^{40}K from these foods accounted for most of the annual effective dose ($\mu\text{Sv}/\text{person} \cdot \text{year}$) of γ -ray emitting nuclides, and the doses of ^{40}K , ^{90}Sr , and ^{238}U were 130–217, 0.21–0.55, and 0.10–0.51, respectively.

Key words — radionuclide, intake, dose estimation, diet, cesium

INTRODUCTION

Clarification of the contents and distribution of toxic substances in foods, and estimation and evaluation of their intakes by the public are important to secure food safety. For this purpose, studies concerning dietary intakes of chemical substances, such as Polychlorinated Biphenyl (PCB) and dioxin, and toxic elements, such as Cd, Pb, and As, have been

performed in Japan.^{1–4)} In addition to these substances, it is important to investigate the intakes of radionuclides, considering them to be toxic substances, and evaluate the dietary exposure doses based on the values obtained. Studies concerning the daily intakes of radionuclides by the public have been performed in many countries,^{5–16)} and the nuclides investigated were natural radionuclides, such as ^{238}U and ^{232}Th , in many reports. Regarding artificial radionuclides, such as ^{137}Cs and ^{90}Sr , surveys and studies concerning the contamination level of food materials and evaluation of the exposure doses to investigate the influences of past atmospheric nuclear tests and the Chernobyl nuclear plant acci-

*To whom correspondence should be addressed: Department of Environmental Health, National Institute of Public Health, 2-3-6, Minami, Wako-shi, Saitama 351-0197, Japan. Tel.: +81-48-458-6263; Fax: +81-48-458-6270; E-mail: sugiyama@niph.go.jp

dent and monitor environments of nuclear energy-related facilities are mainly performed. We have also investigated the levels of γ -ray emitting nuclides in imported foods from foreign countries in Japan.¹⁷⁾ However, fewer systematic studies of daily intakes of artificial radionuclides have been performed based on average food consumption covering all foods ingested by the public.

Duplicated portion and market-basket studies are typical study methods of intakes of toxic substances. The duplicated portion study is capable of directly evaluating toxic substance intakes, but ensuring typicality is necessary because the contents of daily food samples vary depending on taste, time, and region. In Japanese market-basket studies, raw materials of typical foods ingested by the public at high rates are purchased from distribution markets, referring to the "Current state of national nutrition" (Ministry of Health, Labour and Welfare), and toxic substances are individually investigated. This method is superior concerning the typicality of meals per day of average persons because many food groups are covered, and advantageous in that it can be easily used to judge the contributions of individual food groups. However, it is cost-, time-, and labor-intensive. For market-basket studies, the detection of differences in measured values of toxic substances including radionuclides in cooked or uncooked foods is important.

There are various procedures to investigate radionuclide intakes. The procedures are divided into those using the duplicated portion and market-basket methods, and the pretreatment is divided into cooked and uncooked methods. This study adopted the market-basket method, and purchased foods from distribution markets. The foods were boiled, stir-fried, simmered, or roasted following Japanese eating habits to prepare total diet samples reproducing everyday-foods in Japan, and the latest radionuclide intakes by adult Japanese were evaluated. Food samples were classified into 14 food groups including drinking water, as in previous total diet studies (TDS) of chemical substances and toxic elements performed in Japan. The food samples were purchased from distribution markets in 13 cities covering most regions of Japan between 2003 and 2005. The radionuclides investigated were artificial γ -ray emitting nuclides, ^{137}Cs and ^{134}Cs , and natural γ -ray emitting nuclides, ^{40}K , ^{214}Pb , ^{214}Bi , ^{228}Ac , ^{212}Pb , and ^{208}Tl , as well as ^{90}Sr and ^{238}U . All γ -ray emitting nuclides in each sample of the 14 food groups, and ^{90}Sr and ^{238}U in samples prepared

by mixing the 13 food groups excluding drinking water were measured and analyzed, and the intakes were evaluated.

MATERIALS AND METHODS

Collection and Preparation of Total Diet Samples — To reproduce typical everyday Japanese meals, foods were divided into 14 groups including drinking water. Foods were purchased between 2003 and 2005 at supermarkets by the market-basket method in 13 major cities covering most regions of Japan (Hokkaido area: Sapporo, Tohoku area: Sendai, Kanto area: Saitama, Chiba, and Yokohama, Hokuriku area: Niigata, Tokai area: Nagoya, Kinki area: Osaka and Kobe, Chugoku area: Yamaguchi, Shikoku area: Takamatsu, Northern Kyushu area: Fukuoka, and Southern Kyushu area: Naha). Figure 1 shows the sampling sites in Japan. As for drinking water, tap water was collected at each sampling site. The classification (14 food groups) and selection of foods to be sampled from each group in the 13 cities, their daily consumption, and selection of cooking methods were decided on by referring to "TDS data of toxic substances" prepared mainly by the Ministry of Health, Labor and Welfare (based on the results of the 2000 National Nutrition Survey).¹⁸⁾ Table 1 shows the 14 food groups, names of foods belonging to the food groups collected in Fukuoka in the Northern Kyushu area, and their daily consumption as an example. The food materials of each food group were boiled, stir-fried, simmered, and roasted following Japanese eating habits to prepare TDS samples reproducing typical everyday meals in each area (the weight after preparation was 5 or 12 kg in Group 1 and about 5 kg in Groups 2 to 13). The foods were cooked without the addition of food materials belonging to other food groups, such as salt, sugar, fats and oils, and seasonings. Prepared samples excluding those of fats and oils (Group 4) were subjected to ashing at 450°C for about 24 hr after freeze- or dry heat-drying. Regarding 12 food groups excluding fats and oils (Groups 1 to 3 and 5 to 13), 10–100 g of ash samples were placed in plastic containers (100 ml), and the containers were tightly sealed using a silicone sealant and left standing for 2 weeks to prepare measurement samples. As for drinking water, about 100 l were concentrated by heating to reduce the volume, and dry samples were prepared. For analysis of ^{90}Sr and ^{238}U , 13 food

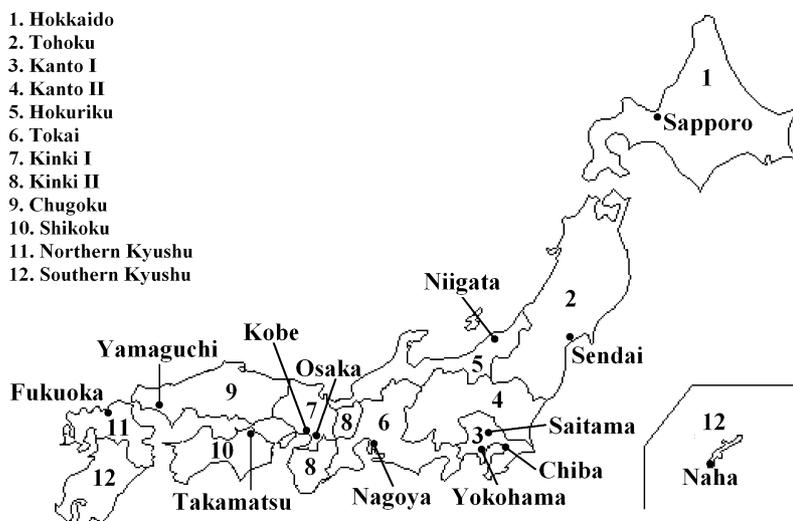


Fig. 1. Thirteen Locations in Japan where We Sampled Fourteen Foodstuff Groups during 2003–2005 for This Total Diet Study

groups excluding drinking water (Groups 1 to 13) were cooked as described above, and about 4 kg mixed samples were prepared: weights of food materials of the food groups to be mixed were calculated by their proportional distribution based on the daily consumption data, and the foods were homogeneously mixed, dried, and ashed to prepare analytical samples.

Measurement and Analysis of Diet Samples — γ -ray emitting nuclides (^{137}Cs , ^{40}K , and U and Th series) were measured in the ash samples for measurement for 200000–300000 seconds using a high-purity Ge semiconductor detector (2519 of CANBERRA Co., Meriden, Connecticut, U.S.A., EGPC20-190-R of EURYSIS Co., Lingolsheim, Cedex-France, CNVD30-35195 of OXFORD Co., Oxon, Oxford, UK, and IGC40200 and IGC25190SD of PGT Co., Princeton, New Jersey, U.S.A.) connected to Multi Channel Analyger and analytical software. As for fats and Oils, the prepared sample was added and sealed in a 1 l Marinelli container, and subjected to γ -ray spectrometry, as with the ash samples.

For ^{90}Sr measurement, the ash sample corresponding to 1–2 kg of the raw sample was heat-degraded with aqua regia and nitric acid, heat-extracted with hydrochloric acid, and filtered, and Sr was separated from the filtrate as oxalate salt precipitates. Ca was then removed by ion exchange. ^{90}Y was removed from the eluate and kept standing for 2 weeks, and ^{90}Y produced by milking was co-precipitated with ferric hydroxide and mounted on a filter as a measurement sample. Measurement samples were measured for 3600–14400 seconds

using a low-background β -ray measurement instrument (LBC-471Q, Aloka Co., Tokyo, Japan).

As for ^{238}U , about 5 g of the ash sample was heat-degraded with nitric acid and hydrogen peroxide solution, dried, dissolved with nitric acid, and filtered through a membrane filter, and the filtrate was adjusted to the specified volume with 1 M nitric acid to prepare a sample solution. Bi was added to the sample solution as an internal standard. ^{238}U in samples was analyzed by Inductively Coupled Plasma Mass Spectrometer (Agilent 7500ce, Yokogawa Analytical Systems Co., Tokyo, Japan).

Quality Control — The reliability of the measured values of the radionuclides obtained by our measurement and analysis was confirmed. The values of γ -ray emitting nuclides in foods and environmental samples (samples of Groups 8 and 11 for this study and ash of incinerated sludge) were compared with those obtained by collaborators of this study and official analytical institutions. The results are shown in Table 2. On comparison of the values in food samples measured by us and collaborators with those measured by official analytical institutions, the ^{137}Cs content was lower by a maximum of 20% at 2 institutions, the ^{40}K , ^{214}Bi , and ^{223}Ac contents were lower by a maximum of 5.3%, 8.2%, and about 30% at one institution, respectively, and the ^{208}Tl content was higher by a maximum of 17% at one institution. The measured natural radionuclide levels employed for the comparison were within relatively small variations, although the radioactivities were close to the lower detection limits. In the ash sample of incinerated sludge, the values measured by the institutions were well consistent. The relia-

Table 1. Classification of Food Group and Daily Consumption (Fukuoka City)

Food group	Food	Daily consumption (g)	
Group I	Rice	white rice (Yumetsukushi (Fukuoka), Koshihikari (Saga), Tsukushiroman (Fukuoka), Koshihikari (Niigata)), rice cake	162.5
Group II	Cereals, seeds and nuts, potatoes	pressed barley, soft flour, bread, sweet bean paste bread, cream bun, wheat noodle (boiled), macaroni spaghetti (dried), instant chinese noodle (fried), sweet corn (canned), sesame, peanut, sweet potato (raw), Irish cobbler (raw), aroid	165.0
Group III	Sugar and preserves, sweets	white sugar, strawberry jam, drops, rice cracker, shortcake, biscuit, chocolate, rice cake stuffed with bean paste	31.9
Group IV	Fats and oils	butter, margarine, vegetable oil, lard, mayonnaise	15.2
Group V	Pulses	sweet miso, soybean miso, barley miso, tofu, fried tofu, fermented soybean, boiled pinto bean	64.5
Group VI	Fruits	Satsuma mandarin (raw), Valencia orange, grapefruit, lemon, apple, banana, strawberry, persimmon, kiwi fruit, pineapple, pear, mandarin	113.9
Group VII	Green and yellow vegetables	carrot, spinach, bell pepper, tomato, pumpkin, lettuce, broccoli, green chive, garland chrysanthemum, long green onion, kidney bean, qing-geng-cai	88.3
Group VIII	Other vegetables, mushrooms, seaweeds	Japanese radish, onion, cabbage, cucumber, Chinese cabbage, eggplant, burdock, sprout, Welsh onions, salted Chinese cabbage, pickled radish, shiitake, wakame (raw)	178.7
Group IX	Seasonings and beverages	strong soy sauce, Worcestershire sauce, salt, sweet cooking rice wine, noodle sauce, sauce for grilled meat, flavor seasonings (consommé), sake, beer, distilled spirit, canned coffee, carbonated beverage (Coke), sports drink	172.2
Group X	Fish and shellfish	salmon (raw), tuna (lean), red sea bream (raw), flounder (raw), mackerel (raw), horse mackerel (raw), sardine (raw), hairtail (raw), barracuda (raw), yellowtail (raw), cuttlefish (raw), shrimp (raw), queen crab (raw), littleneck clam (raw), scallop (raw), salted salmon, dried horse mackerel, tuna in spring water, fish flake boiled in soy sauce, steamed fish paste, minced flesh, fish	85.2
Group XI	Meat and poultry, eggs	beef round, beef chuck, pork loin, ham, chicken thigh (with skin), canned whale meat, horsemeat, meat product (sausage), hen egg	119.5
Group XII	Milk and dairy products	normal milk, processed milk, processed cheese, sugar-free yogurt, sweetened yogurt	122.5
Group XIII	Others	curry roux, cream soup (powder), vinegar, roux for hashed rice	5.6
Group XIV	Drinking water	tap water	600.0
		total	1925.0

bility of the analytical values of ^{238}U and ^{90}Sr was confirmed using standards, JB-1 (Basalt), National Institute of standards and Technology 1632 (Coal), NIST (Cal Fly Ash), and milk powder of WHO:

World Health Organization/IRC: International Reference Center. The results are shown in Table 3. The values of the 2 radionuclides measured by this analytical method were well consistent with the cer-

Table 2. Comparative Analysis of γ -ray Emitting Nuclides in Food and Environmental Samples (unit: Bq/kg)

Sample	Nuclide	Analytical institution			
		Research institution A	Research institution B	Research institution C	Official analytical institution
Yokohama Group VIII ^{a)}	¹³⁷ Cs	0.049 ± 0.011 ^{c)}	0.040 ± 0.010 ^{c)}	0.031 ± 0.004 ^{c)}	Undetectable
	⁴⁰ K	56 ± 0.5	54 ± 0.6	56 ± 0.3	57 ± 0.5 ^{c)}
	²¹⁴ Pb	<0.065	0.087 ± 0.021	0.100 ± 0.010	Not measured
	²¹⁴ Bi	<0.077	<0.067	0.078 ± 0.009	0.085 ± 0.017
	²²⁸ Ac	0.160 ± 0.065	0.230 ± 0.042	0.230 ± 0.020	0.230 ± 0.037
	²¹² Pb	0.150 ± 0.018	0.140 ± 0.017	0.210 ± 0.008	Not measured
	²⁰⁸ Tl	<0.039	0.051 ± 0.010	0.055 ± 0.004	0.047 ± 0.008
Takamatsu Group XI ^{a)}	¹³⁷ Cs	0.032 ± 0.009	0.028 ± 0.004	0.028 ± 0.004	0.035 ± 0.006
	⁴⁰ K	52 ± 0.4	50 ± 0.3	51 ± 0.2	51 ± 0.4
	²¹⁴ Pb	<0.045	0.035 ± 0.011	0.059 ± 0.009	Not measured
	²¹⁴ Bi	<0.049	<0.029	0.055 ± 0.009	0.054 ± 0.012
	²²⁸ Ac	<0.140	0.080 ± 0.018	0.066 ± 0.019	Undetectable
	²¹² Pb	<0.036	0.026 ± 0.008	0.044 ± 0.007	Not measured
	²⁰⁸ Tl	<0.025	<0.014	<0.014	Undetectable
Ash of incinerated sludge ^{b)}	¹³⁷ Cs	Not measured	Not measured	Not measured	Not measured
	⁴⁰ K	Not measured	Not measured	Not measured	Not measured
	²¹⁴ Pb	0.026 ± 0.002	0.026 ± 0.003	0.025 ± 0.002	0.029 ± 0.002
	²¹⁴ Bi	0.023 ± 0.002	0.023 ± 0.007	0.026 ± 0.002	0.024 ± 0.002
	²²⁸ Ac	0.054 ± 0.005	0.057 ± 0.006	0.063 ± 0.004	0.057 ± 0.004
	²¹² Pb	0.072 ± 0.002	0.071 ± 0.002	0.073 ± 0.007	0.063 ± 0.002
	²⁰⁸ Tl	0.019 ± 0.001	0.022 ± 0.001	0.023 ± 0.001	0.021 ± 0.001

a) The concentration is presented as Bq/kg raw. b) The concentration is presented as Bq/kg dry. c) Counting error.

Table 3. Analytical Results of ²³⁸U and ⁹⁰Sr in Standards

Sample	Nuclide	Analytical method	Analytical result (Bq/kg)	Certified value	Converted ²³⁸ U value (Bq/kg)
JB-1 (Basalt)	²³⁸ U	ICP-MS ^{a)}	20 ± 0.3 ^{b)}	1.8 ppm	22
NIST 1632a (Coal)			21 ± 0.7 ^{b)}		
NIST 1633a (Coal Fly Ash)	²³⁸ U	ICP-MS ^{a)}	16 ± 0.2 ^{b)}	1.28 ± 0.02 (μ g/g)	16 ± 0.2
WHO/IRC (Milk Powder)			⁹⁰ Sr	130 ± 2 ^{b)} 130 ± 4 ^{b)}	10.2 ± 0.1 (μ g/g)
			32 ± 1.8	30.8 ± 3.1 ^{c)}	

a) Inductively coupled plasma mass spectrometry. b) Standard deviation of the measurement repeated 5 times. c) Analytical results of the International Reference Center (IRC).

tified values.

RESULTS AND DISCUSSION

Contents of Radionuclides

The γ -ray emitting nuclides measured were artificial nuclides, such as ¹³⁷Cs, and natural γ -ray emitting nuclides, for which measurement of the radioactivity in foods has recently been required, such as ²¹⁴Pb, ²¹⁴Bi, ²²⁸Ac, ²¹²Pb, and ²⁰⁸Tl. The ra-

dioactivities (per weight after cooking) of the γ -ray emitting nuclides were measured in the 13 cities. The results of Fukuoka for individual food groups are shown in Table 4 as an example. On summarizing the results of all 13 cities, only ¹³⁷Cs was measurable as an artificial γ -ray emitting nuclide, and its radioactivity was lower than 0.1 Bq/kg in most of the 13 cities and lower than the detection limit in many food groups. The highest ¹³⁷Cs level was 0.145 Bq/kg in fish and shellfish (Group 10) in Yamaguchi, and the lowest level was 0.005 Bq/kg in

Table 4. Radioactivity Concentrations of γ -ray Emitting Nuclides in Fourteen Food Groups in Fukuoka City in Japan (unit: Bq/kg raw^{a)})

Group	¹³⁷ Cs	¹³⁴ Cs	⁴⁰ K	²¹⁴ Pb	²¹⁴ Bi	²²⁸ Ac	²¹² Pb	²⁰⁸ Tl
Group I	<0.006	<0.007	6.6 ± 0.1 ^{b)}	<0.015	<0.014	<0.026	<0.011	<0.006
Group II	<0.025	<0.028	62.2 ± 0.6	<0.066	<0.062	<0.126	<0.056	<0.028
Group III	<0.010	<0.014	42.2 ± 0.3	<0.035	<0.035	<0.056	<0.027	<0.015
Group IV	<0.021	<0.022	3.2 ± 0.2	<0.105	<0.094	<0.153	<0.100	<0.044
Group V	<0.041	<0.059	90.6 ± 1.1	<0.130	<0.126	<0.223	<0.100	<0.053
Group VI	<0.023	<0.031	57.8 ± 0.6	<0.048	<0.050	<0.098	<0.048	<0.207
Group VII	<0.021	<0.035	84.5 ± 0.7	<0.053	<0.087	<0.125	<0.048	<0.026
Group VIII	0.017 ± 0.005 ^{b)}	<0.016	55.9 ± 0.3	<0.034	<0.036	0.065 ± 0.021 ^{b)}	<0.027	<0.015
Group IX	<0.039	<0.033	32.0 ± 0.6	<0.087	<0.085	<0.171	<0.066	<0.036
Group X	0.079 ± 0.011	<0.042	109.6 ± 0.9	<0.083	<0.078	<0.145	<0.058	<0.033
Group XI	0.053 ± 0.010	<0.039	85.6 ± 0.8	<0.069	0.074 ± 0.023 ^{b)}	0.160 ± 0.050	<0.055	<0.029
Group XII	<0.026	<0.030	49.1 ± 0.6	<0.069	<0.056	<0.096	<0.045	<0.027
Group XIII	<0.014	<0.019	8.0 ± 0.4	<0.052	<0.056	<0.084	<0.040	<0.025
Group XIV	<0.0003	<0.0004	0.06 ± 0.004	<0.0011	<0.0015	<0.0015	<0.0008	<0.0005

a) Per weight after cooking. b) Counting error.

Table 5. ⁹⁰Sr and ²³⁸U concentrations in TDS Mixed Samples in Japan

Location	⁹⁰ Sr (Bq/kg) ^{a)}	²³⁸ U (Bq/kg) ^{a)}
Sapporo	0.013 ± 0.0042 ^{b)}	0.0040 ± 0.00013 ^{b)}
Sendai	0.020 ± 0.0039	0.0083 ± 0.00006
Saitama	0.026 ± 0.0040	0.0042 ± 0.00002
Chiba	0.027 ± 0.0051	0.0140 ± 0.00030
Yokohama	0.031 ± 0.0054	0.0180 ± 0.00010
Niigata	0.026 ± 0.0052	0.0046 ± 0.00012
Nagoya	0.028 ± 0.0054	0.0072 ± 0.00019
Osaka	0.021 ± 0.0048	0.0046 ± 0.00011
Kobe	0.029 ± 0.0041	0.0090 ± 0.00003
Yamaguchi	0.029 ± 0.0053	0.0039 ± 0.00012
Takamatsu	0.016 ± 0.0046	0.0066 ± 0.00010
Fukuoka	0.026 ± 0.0042	0.0049 ± 0.00001
Naha	0.022 ± 0.0047	0.0098 ± 0.00019
mean ± σ	0.024 ± 0.0054	0.0076 ± 0.0043

a) Group XIV (drinking water) was excluded. The concentration is presented with per (raw) weight after cooking. b) Counting error.

rice/processed rice products (Group 1) in Sapporo. On comparison of the food groups, the highest ¹³⁷Cs level (0.045–0.145 Bq/kg) was detected in fish and shellfish (Group 10) in many cities, followed by meat/eggs and milk. A Chernobyl nuclear plant accident-derived artificial radionuclide, ¹³⁴Cs, was not detected or measured in any food or drinking water sample from any of the 13 cities.

Since about 0.0117% of a natural radionuclide, ⁴⁰K, is present in foods as an isotope of a major element of foods, K, ⁴⁰K was measured in various foods. In Fukuoka, the ⁴⁰K level per weight after cooking was 3.2–109.6 Bq/kg raw in the 13 food

groups excluding drinking water. The ⁴⁰K level in drinking water (0.019–0.141 Bq/kg raw) was generally lower than that in the other food groups, and the second lowest level was detected in oils and fats and rice/processed rice products. The levels of typical natural γ -ray emitting nuclides, ²¹⁴Pb, ²¹⁴Bi, ²²⁸Ac, ²¹²Pb, and ²⁰⁸Tl, were lower than the detection limits in many food groups in the 13 cities.

The ⁹⁰Sr radioactivity level was analyzed in samples prepared by mixing all 13 food groups excluding drinking water (Group 14). The radioactivities (per weight after cooking) are shown in Table 5. The ⁹⁰Sr level was within a range of 0.013–0.031 Bq/kg in the 13 cities, with a mean of 0.024 ± 0.005 Bq/kg, and the regional variation in cooked foods was small. The lowest and highest ⁹⁰Sr radioactivities were detected in Sapporo and Yokohama, respectively.

²³⁸U radioactivity was analyzed in a sample prepared by mixing all 13 food groups excluding drinking water (Group 14), similarly to ⁹⁰Sr. The radioactivities (per weight after cooking) are shown in Table 5. The ²³⁸U level was within a range of 0.0039–0.0180 Bq/kg in the 13 cities, with a mean of 0.0076 ± 0.0043 Bq/kg. There was 56% variation relative to the mean value, and the regional variation in cooked foods was larger than that of ⁹⁰Sr.

Daily Intakes of Radionuclides

The daily intakes of γ -ray emitting nuclides were calculated from the radioactivities measured by TDS in the 13 Japanese cities and the rates of food consumption in these areas. The daily intakes

Table 6. Daily ^{137}Cs Intakes from Fourteen Food Groups in Japan (unit: mBq/person · day)

Group	Sapporo	Sendai	Saitama	Chiba	Yokohama	Niigata	Nagoya	Osaka	Kobe	Yamaguchi	Takamatsu	Fukuoka	Naha
Group I	2.2	<4.6	1.6	<1.8	<4.1	7.4	<3.9	9.3	<2.5	<1.5	<10.3	<2.0	3.5
Group II	2.8	11.4	<2.0	<5.1	<5.1	4.9	8.0	<11.9	2.1	4.6	<5.1	<4.8	<2.6
Group III	0.8	0.7	0.6	<0.4	<0.7	0.9	1.3	<1.3	0.9	<0.6	<0.9	<0.3	<0.4
Group IV	<0.6	<0.7	<1.1	<0.4	<0.5	<0.6	<0.7	<1.2	<0.4	<0.5	<1.9	<0.3	<0.3
Group V	<2.1	<3.1	<3.1	<3.9	<3.5	<4.5	<3.6	<4.1	<1.7	<2.4	<4.7	<2.8	<2.7
Group VI	<3.2	4.7	<1.4	<3.0	<1.7	<3.5	<3.1	<3.8	<2.9	<2.3	<2.1	<2.6	<1.8
Group VII	<1.1	<3.6	<1.6	2.7	<12.0	2.5	2.5	<5.2	<2.2	<2.1	<2.5	<1.8	<2.1
Group VIII	<3.7	<4.0	<2.4	3.1	8.9	9.1	7.0	<6.1	6.1	<3.5	<14.3	2.9	<2.6
Group IX	<2.8	<4.5	<2.4	<4.5	<9.1	<6.5	<6.2	<14.5	<7.5	<2.1	<9.6	<6.7	<6.2
Group X	7.1	9.1	7.8	7.2	7.1	6.6	8.8	6.5	8.4	12.4	7.0	6.2	8.0
Group XI	5.0	4.4	<2.3	4.0	8.9	6.9	5.2	9.0	7.5	5.9	5.0	5.4	6.3
Group XII	3.7	<3.0	7.0	8.6	1.8	4.3	9.3	5.7	2.9	2.4	<5.5	<3.2	5.0
Group XIII	<0.6	<0.2	0.1	<0.1	<0.4	<1.1	<1.0	<0.5	<0.4	<0.1	0.6	<0.5	<0.1
Group XIV	2.8	<0.1	6.1	0.2	<0.2	<0.2	<0.2	<0.6	<0.1	<0.3	<0.2	<0.2	<0.2
total	24.4<T<38.6 ^{a)}	30.3<T<54.2	23.2<T<39.5	25.8<T<45.1	26.7<T<63.8	42.5<T<58.8	42.1<T<60.8	30.5<T<79.7	27.9<T<45.5	25.3<T<40.5	12.5<T<69.5	14.5<T<39.7	22.8<T<41.9

a) The minimum and maximum values were calculated by excluding and including values lower than the lower detection limits, respectively.

Table 7. Daily ^{40}K Intakes from Fourteen Food Groups in Japan (unit: mBq/person · day)

Group	Sapporo	Sendai	Saitama	Chiba	Yokohama	Niigata	Nagoya	Osaka	Kobe	Yamaguchi	Takamatsu	Fukuoka	Naha
Group I	2389	2567	1647	2045	1666	2748	2378	2992	2758	2433	2510	2336	2712
Group II	11951	15007	9310	13545	9143	12444	10870	24341	10396	8342	8426	11768	8283
Group III	1206	1078	2059	1288	1353	1163	2352	2922	2018	655	996	1384	905
Group IV	26	<18	62	47	43	87	<16	43	43	70	<41	49	54
Group V	6178	7509	7697	6833	7049	5784	7325	4957	7126	4318	4694	6299	5365
Group VI	6129	6849	5943	7627	5338	7273	6431	8987	8329	5985	4965	6581	5433
Group VII	7432	9766	9534	6993	7742	7602	7999	9968	8534	917	6405	6930	7066
Group VIII	14660	17103	8475	16062	11983	11526	11266	9530	15751	7916	26776	9678	5998
Group IX	5723	6773	4059	6071	20190	4824	5515	6171	4942	3440	9089	5510	4564
Group X	9046	9467	6992	7320	6651	9047	7219	8423	7920	8605	8012	8605	6690
Group XI	8442	7939	9030	7885	9095	6259	6583	10469	5748	8984	8055	8665	8694
Group XII	5595	6034	6484	6305	5481	6742	6549	6692	7940	5436	6995	6015	6087
Group XIII	671	208	244	648	438	465	403	166	206	164	361	274	213
Group XIV	27	11	50	61	21	29	15	85	56	44	14	36	32
total	T=79475	311<T<90329	T=71586	T=82730	T=86193	T=75993	74905<T<74921	T=95746	T=81767	T=57309	87299<T<87340	T=74130	T=62096

a) The minimum and maximum values were calculated by excluding and including values lower than the lower detection limits, respectively.

(mBq/person · day) of the 8 γ -ray emitting nuclides (^{137}Cs , ^{134}Cs , ^{40}K , ^{214}Pb , ^{214}Bi , ^{228}Ac , ^{212}Pb , and ^{208}Tl) from each food group were calculated, and the total intake calculated by adding the intakes from all food groups was evaluated. The total of measured values, excluding values lower than the detection limits, was defined as the minimum value, and the value calculated by adding the lower detection limits as non-measurable values to this minimum value was defined as the maximum value. Since the radioactivities of the natural radionuclides (^{214}Pb , ^{214}Bi , ^{228}Ac , ^{212}Pb , and ^{208}Tl) were lower than the detection limits in many samples in all 13 cities, as shown in the results of Fukuoka (Table 4),

the daily intakes of these nuclides were evaluated as very low. Thus, only the daily intakes of ^{137}Cs and ^{40}K in the 13 cities are shown in Tables 6 and 7. Since the radioactivity of undetectable samples was not handled as "0", and the lower detection limit was adopted as its value, attention should be paid to the fact that the daily intakes were overestimated. In Tables 6 and 7, the total daily intakes (T) are represented as the minimum values (calculated from the quantifiable values) < T (total) < maximum values (calculated by adding the detection limits to the minimum value). The highest minimum daily intake of ^{137}Cs (42.5 mBq/person · day) was noted in Niigata among the 13 cities in

Table 8. Regional Daily Intakes of ^{90}Sr and ^{238}U at Thirteen Locations in Japan

Location	^{90}Sr (mBq/person·day) ^{a)}	^{238}U (mBq/person·day) ^{a)}
Sapporo	20.8	6.4
Sendai	35.6	14.8
Saitama	39.9	6.4
Chiba	44.8	23.2
Yokohama	53.6	31.1
Niigata	43.4	7.7
Nagoya	45.0	11.6
Osaka	33.5	7.3
Kobe	48.2	14.9
Yamaguchi	43.8	5.9
Takamatsu	26.7	11.0
Fukuoka	40.1	7.5
Naha	34.5	15.4
mean $\pm\sigma$	39.2 \pm 8.9 (C.V. 22.7%)	12.6 \pm 7.5 (C.V. 59.5%)

a) Group XIV (drinking water) was excluded.

the 12 areas, and the highest maximum intake (<79.7 mBq/person·day) was noted in Osaka. The intake in all 13 cities was within a range of 12.5–<79.7 mBq/person·day. Fish and shellfish (Group 10), milk (Group 12), meat/eggs (Group 11), cereals/nuts and seeds/potatoes (Group 2), and other vegetables/mushrooms/seaweed (Group 8) accounted for a large part of the intake. ^{40}K was quantifiable in all foods excluding a few samples of fats and oils. Regional differences were noted in the daily intakes from individual food groups, but the total intake from all 14 food groups was 57309–95746 mBq/person·day in the 13 cities, showing no marked regional difference. As a regional characteristic, all ^{214}Pb , ^{214}Bi , ^{228}Ac , ^{212}Pb , and ^{208}Tl could be measured in other vegetables/mushrooms/seaweed (Group 8) in Chiba and Yokohama. On comparison of the composition of the food material samples (Group 8), between Chiba and the other areas, the content of seaweed was slightly higher in Chiba. The reason for this was unclear, and further investigation is necessary.

To calculate the daily intakes of ^{90}Sr and ^{238}U , samples of the individual 13 food groups excluding drinking water (Group 14) were individually cooked and homogeneously mixed, and the ^{90}Sr and ^{238}U radioactivities were measured in the mixed samples. The calculated daily intakes of ^{90}Sr and ^{238}U are shown in Table 8. The daily ^{90}Sr intake in the 13 cities was within a range of 20.8–53.6 mBq/person·

day, and the mean was 39.2 ± 8.9 mBq/person·day, showing about 20% deviation relative to the mean among the cities. The daily ^{238}U intake was calculated in the same way as for ^{90}Sr intake. The results are shown in Table 8. The daily ^{238}U intake in the 13 cities was within a range of 5.9–31.1 mBq/person·day, and the mean was 12.6 ± 7.5 mBq/person·day. The maximum daily ^{238}U intake detected in Yokohama was 5 times or greater than the minimum intake detected in Sapporo, and there was about 60% deviation relative to the mean among the cities. Since the ^{238}U content is known to be higher in seaweed than in other foods, we will further investigate the food material compositions of individual food groups and related references to clarify factors related to the regional differences in the intake.

Comparison of the Present Results with International Values

To evaluate the daily intakes of γ -ray emitting nuclides, ^{90}Sr and ^{238}U , in the 13 cities in Japan, the values were compared with those reported in other countries, as shown in Table 9. In this market-basket TDS, the daily intakes of ^{137}Cs and ^{40}K were 12.5–<79.7 and 57309–95746 mBq/person·day, respectively, and the mean intakes of ^{90}Sr and ^{238}U were 39.2 ± 8.9 and 12.6 ± 7.5 mBq/person·day, respectively. In study results of other countries, the daily ^{137}Cs intake widely varied from 530 to 315000 mBq/person·day (mean: 4050 ± 497 mBq/person·day) in TDS performed in Kiev, Rovno, and Volynsky in the Ukraine in 1994,⁵⁾ and that of ^{40}K was 25400–169000 mBq/person·day (mean: 85100 ± 1580 mBq/person·day). In the results of a nationwide duplicated portion study performed in the early 1990's by the Japan Chemical Analysis Center,⁶⁾ the ^{137}Cs and ^{90}Sr intakes were 64 and 66 mBq/person·day, respectively. The daily ^{137}Cs intake obtained in the present study was similar to that measured in the early 1990's in Japan. The ^{40}K intake was also similar to that in the Ukraine, and the variation was relatively small. The daily ^{137}Cs intake was higher in the Ukraine than in Japan, which may have been associated with the contamination caused by the Chernobyl nuclear plant accident. The mean daily ^{90}Sr intake was considered to be similar to that reported in the early 1990's, although it was slightly lower. Studies concerning the daily ^{238}U intake have been performed in many countries since the 1960's. In America, the intake was 16.2⁷⁾ and 15.9 mBq/person·day⁸⁾

Table 9. Daily Dietary Intakes of ^{137}Cs , ^{40}K , ^{90}Sr and ^{238}U in Japan and Other Countries

Nuclide	Country	Intake (mBq/person·day)	Reference
^{137}Cs	Ukraine (Kiev, Rovno, Volynsky)	530–315000 (mean: 4050 ± 4970)	5)
	Japan	12.5–<79.7 64	present study 6)
^{40}K	Japan	57309–95746	present study
	Ukraine	25400–169000 (mean: 85100 ± 1580)	5)
^{90}Sr	Japan	39.2 ± 8.9 66	present study 6)
^{238}U	U.S.A. (New York)	16.2	7)
	(New York)	15.9	8)
	(Utah)	54	9)
	U.K.	12.4	10)
	Pakistan	27	11)
	India	6.8	12)
		27	12)
	Poland	22.1	13)
	Japan	12.6 ± 7.5 8.8 14	present study 14) 15)

in New York, and 54 mBq/person·day⁹⁾ in Utah. The intake was 12.4 mBq/person·day in England,¹⁰⁾ 27 mBq/person·day in Pakistan,¹¹⁾ 6.8 and 27 mBq/person·day in India,¹²⁾ and 22.1 mBq/person·day in Poland.¹³⁾ In Japan, Shiraishi *et al.* performed a duplicated portion study in 31 areas of Japan between 1980 and 1988, and found that the mean ^{238}U intake was 8.8 mBq/person·day.¹⁴⁾ Kuwahara *et al.* analyzed uncooked food samples in Yokohama between 1985 and 1993, and found that the intake was 14 mBq/person·day.¹⁵⁾ Shiraishi *et al.* previously investigated ^{238}U mainly by the duplicate portion method, and additionally investigated the contribution of ^{238}U from each of 18 food groups using the market-basket method.¹⁶⁾ The daily ^{238}U intake in the present study was not markedly different from the values in these past studies in Japan, and the value was not particularly high in comparison with international data.

Annual Effective Dose Estimation

Basically, the exposure doses (Sv) of radionuclides through food ingestion are dependent on the nuclide intakes through food ingestion (Bq) and the coefficient for conversion to the effective dose (mSv/Bq). The exposure dose was estimated based on the following example of the generally used cal-

culational equation.

Radionuclide intake, i (Bq), is provided by the equation below.

$$A_{m,i} = C_{m,i} \cdot M_m \cdot fm_m \cdot fd_m \cdot t_m, \text{ where:}$$

$A_{m,i}$: Intake of radionuclide, i , through ingestion of food, m (Bq)

$C_{m,i}$: Concentration of radionuclide, i , in the food to be evaluated, m , at the time of sampling (Bq/kg)

t_m : Duration of ingestion of food, m (d)

M_m : Intake of food, m , per day (kg/d)

fm_m : Market dilution coefficient of food, m (–)

fd_m : Decontamination coefficient of food, m , by cooking (–)

In this equation, when the radionuclide has a long physical half-life, physical attenuation during the period between the food sampling and measurement can be disregarded.

Accordingly, the internal exposure dose, H (mSv), through food ingestion is provided by the equation below.

$$H = \sum_m \sum_i K_i \cdot A_{m,i}, \text{ where:}$$

H : Effective dose through food ingestion (mSv)

K_i : Conversion coefficient to effective dose through oral ingestion of radionuclide, i (mSv/Bq)

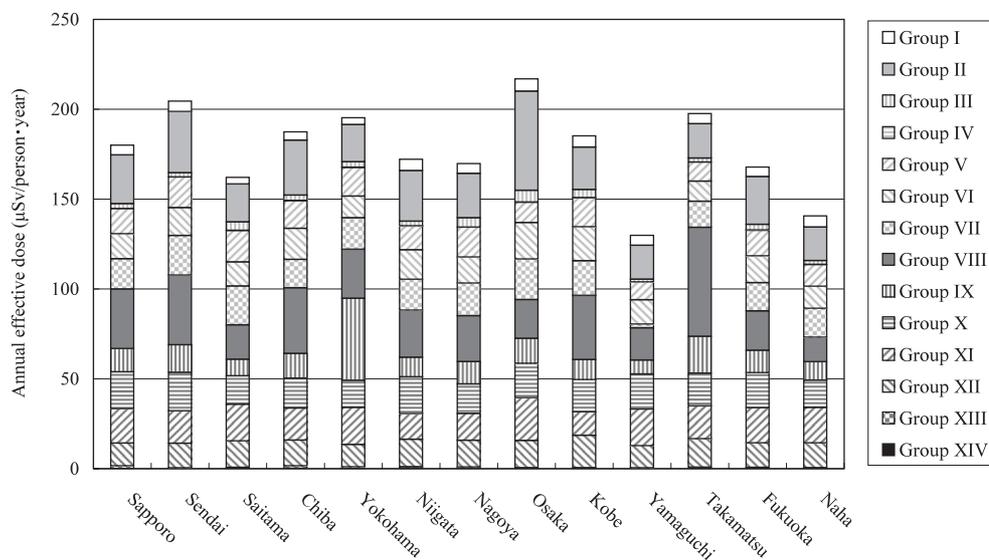


Fig. 2. Annual Effective doses of γ -ray Emitting Nuclides (^{40}K , ^{137}Cs , and the Others) due to Ingestions of the Daily Total Diet for Adult Members of the Public in Thirteen Cities in Japan during 2003–2005

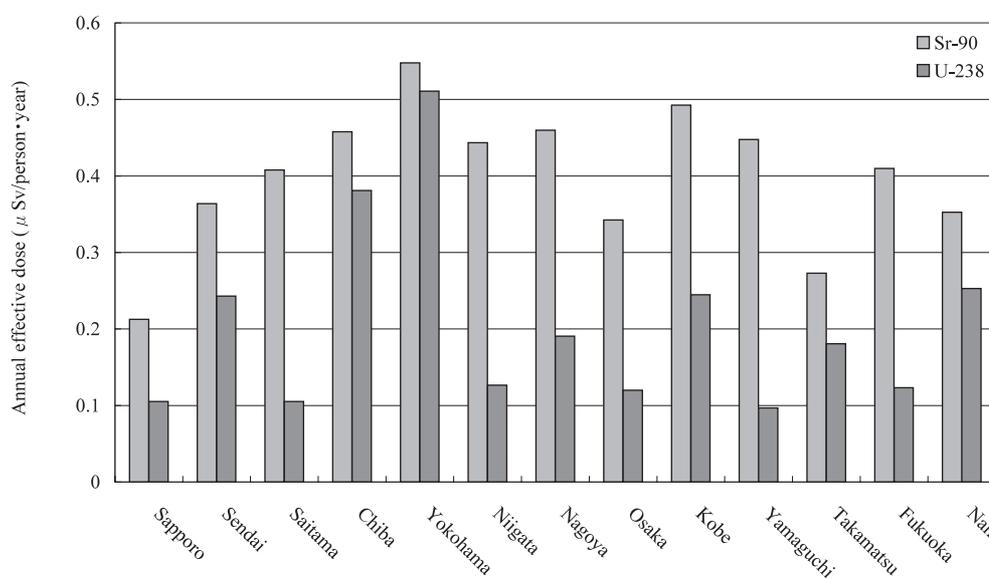


Fig. 3. Annual Effective doses of ^{90}Sr and ^{238}U Excluding Drinking Water due to Ingestions of Daily Diets for Adult Members of the Public in Thirteen Cities in Japan during 2003–2005

The exposure dose through the ingestion of each radionuclide in adults was estimated by the above calculation. For the dose conversion coefficients, the values for adults published in International Commission on Radiological Protection (ICRP) Publication 72¹⁹⁾ by the International Commission on Radiological Protection were applied. The annual effective doses through ingestion of γ -ray emitting nuclides in adults are shown in Fig. 2. The effective doses of the radionuclides were as follows: The effective dose of ^{137}Cs (dose conversion coefficient: 1.3×10^{-5} mSv/Bq)

was estimated to be 0.049 – <0.378 μSv . The exposure dose of ^{40}K (dose conversion coefficient: 6.2×10^{-6} mSv/Bq) was similarly estimated to be 130 – 217 μSv . As for natural radionuclides, ^{214}Pb (dose conversion coefficient: 1.4×10^{-7} mSv/Bq), ^{214}Bi (1.1×10^{-7} mSv/Bq), ^{228}Ac (4.3×10^{-7} mSv/Bq), and ^{212}Pb (6.0×10^{-6} mSv/Bq), the minimum-maximum effective dose of ^{214}Pb was 0 – <0.007 μSv , and the highest measured value was 0.001 μSv . Those of ^{214}Bi were 0 – 0.007 and 0.001 μSv , those of ^{228}Ac were 0 – <0.057 and 0.015 μSv , and those of ^{212}Pb were

0–<0.282 and 0.112 μSv , respectively. Those of ^{208}Tl were not calculated because its dose conversion coefficient was not presented in the ICRP Publication. Based on these findings, most of the effective dose through ingestion of γ -ray emitting nuclides in adults is derived from ^{40}K , and the contribution of ^{137}Cs and natural radionuclides (^{214}Pb , ^{214}Bi , ^{228}Ac , and ^{212}Pb) to the dose is small.

Figure 3 shows the annual effective doses of ^{90}Sr and ^{238}U ($\mu\text{Sv}/\text{person} \cdot \text{year}$) in the 13 cities. The exposure dose of an artificial radionuclide, ^{90}Sr (dose conversion coefficient: 2.8×10^{-5} mSv/Bq), was 0.21–0.55 μSv in the 13 cities, with a mean of 0.40 μSv . This value was well consistent with that published in the 2000 annual report by the United Nations Science Committee on the Effects of Atomic Radiation²⁰⁾ (0.56 μSv ; UNSCEAR 2000). As for ^{238}U (dose conversion coefficient: 4.5×10^{-5} mSv/Bq), the exposure dose was 0.10–0.51 μSv in the 13 areas, and the mean value for the 13 cities was estimated to be 0.21 μSv . The estimated annual effective dose of ^{238}U in adults was similar to that reported by UNSCEAR 2000 (0.14–0.30 μSv) and the mean value in everyday meals reported by the Japan Chemical Analysis Center (mean: 0.23 μSv , 0.09–0.46 μSv).²¹⁾

Since the lower detection limit was adopted as the maximum concentration when quantification was not possible in the calculation of the annual effective dose, the values were overestimated. Even though this point was taken into consideration, the exposure dose in adults obtained was far lower than the dose limit for the public (1 mSv/year, ICRP 1990 recommendation), and similar to or lower than the mean annual effective dose of natural radionuclide exposure through ingestion in adults (0.29 mSv, UNSCEAR 2000).

In conclusions, various foods were sampled by the market-basket method in 13 cities in 12 areas throughout Japan, cooked by boiling, stir-frying, simmering, and roasting according to Japanese eating habits, reproducing typical everyday meals in the areas investigated, to prepare total diet samples for analysis. Although regional differences to a maximum of about 5 times were noted in the daily ^{238}U intake, the daily intakes of γ -ray emitting radionuclides, ^{90}Sr , and ^{238}U were generally low, and their contribution to the exposure dose in adults was small. Out of the toxic substances contained in foods, ^{238}U and ^{232}Th have been investigated in many studies concerning daily intakes of radionuclides and evaluation of exposure to nuclides in

Japan. Under such circumstances, this study clarified the latest states concerning the daily intakes of various radionuclides, not only U used in the nuclear power-related field but also artificial radionuclides (γ -ray emitting nuclides and ^{90}Sr) derived from past atmospheric nuclear tests and the Chernobyl nuclear plant accident and natural γ -ray emitting nuclides. The results of this study are valuable basic data which can be used to ensure the safety and security of foods against radionuclides.

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