

Figure 4. Distribution of K in each fraction of *P. ostreatus*-Y1 mycelia cultured on YM agar following density-gradient centrifugation. The basic K content in YM medium was 8.4 mM.

Measurement of Cs and K Contents in *P. ostreatus* Mycelia Fraction by Cell Fractionation. *P. ostreatus* mycelia incubated on YM agar medium (0.4% yeast extract, 1% malt extract, 0.4% glucose, and 1.8% agar, pH 7.2) containing 15 mM CsCl at 27 °C for 20 days were harvested and washed with distilled water. The mycelia were disrupted using a Bead-Beater (Biospec Products, Bartlesville, OK, USA). The chamber was filled with 1 M sorbitol (Sigma), and mycelia were homogenized with glass beads (diameter: 0.5 mm) in a vessel filled with crushed ice for 1 min. The homogenized mycelia were fractionated according to the revised method for isolating vacuolar and mitochondrial pellets (19, 20). The homogenized mycelia were centrifuged at 600g for 5 min (15 °C), and the supernatant was additionally centrifuged at 15,000g for 20 min (15 °C) to obtain the sediment. The sediment was suspended in 6 mL of 1 M sorbitol. Eighteen milliliters of 1.8 M sucrose (Sigma) – 1 M sorbitol, 12 mL of 1.8 M sucrose and 6 mL of mycelia suspension in 1 M sorbitol were added to a centrifuge tube in the order of lower to upper layers, and then centrifuged at 43,000g for 120 min (4 °C). In addition to the above-mentioned mycelia suspension, control samples were also prepared according to the procedures described above, except that the top layer was changed to 1 M sorbitol solution containing 5 mM CsCl or 1 M sorbitol containing 5 mM KCl (Wako). A total of 12 layers (3-mL aliquots of both mycelia suspension and controls) were pipetted into clean tubes to fractionate the liquid and sediment layers. These fractions were digested individually with approximately 20 mL of 14 M HNO₃ (Wako) and 2 mL of 30% H₂O₂ (Wako) at 160 °C for 4–12 h on a hot plate. The digested samples were diluted with up to 10 mL of 0.5 N HCl (Wako and MilliQ Element), and then the Cs and K contents were analyzed separately by flame atomic absorption spectrometry (Hitachi A-2000) using hollow cathode lamps (Hamamatsu Photonics, Cs; 852.1 nm absorption lines and Hitachi, K; 766.5 nm absorption lines). Similarly, flame atomic absorption spectrometry of the control CsCl and KCl fractions was performed without the wet digestion. The K content in the YM medium as a blank value was 8.4 mM.

Detection of Polyphosphate by Staining of *P. ostreatus* Mycelia. *P. ostreatus* was inoculated onto a YM agar plate (0.4% yeast extract, 1% malt extract, 0.4% glucose, and 1.8% agar, pH 7.2) containing 15 mM CsCl and incubated at 27 °C for seven days. The control sample (without Cs) was also incubated under the same conditions. Cultivated mycelia were carefully scraped from the plate and stained with 50 µg mL⁻¹ of DAPI (ABD Bioquest) for 10 min. Stained mycelia were

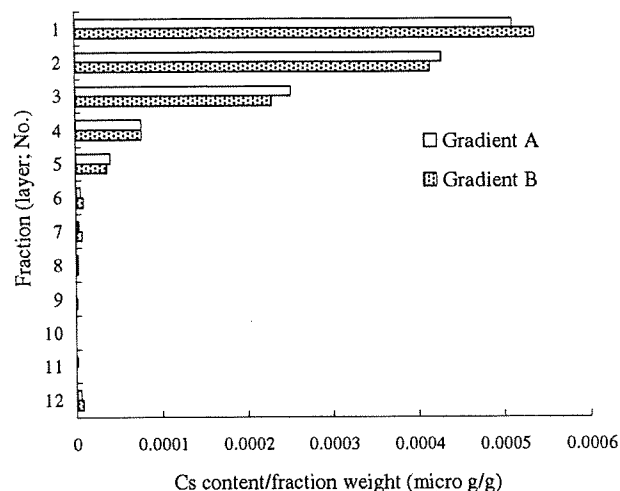


Figure 5. Distribution of Cs in each fraction of the control solution containing 5 mM CsCl following density-gradient centrifugation.

washed with distilled water and observed by fluorescence microscopy using a confocal laser scanning microscope (Carl Zeiss LSM 510 META). By DAPI staining, the fluorescence of polyphosphate (polyphosphate-DAPI) and DNA-DAPI can be observed around 526 nm (yellowish fluorescence) and 456 nm (bluish fluorescence), respectively (21). The presence of fluorescence corresponding to polyphosphate in mycelia and its morphology were observed.

RESULTS AND DISCUSSION

SEM Observation and Qualitative Elemental Analysis of *P. ostreatus* Cultivated Mycelia. SEM equipped with EDX, which can detect the presence of elements in the microstructure of plant tissue, is one of the most effective techniques in elemental analysis. As shown in **Figure 1a**, a nonuniform distribution of circular white spots was observed in *P. ostreatus* mycelia cultivated in the presence of 25 mM CsCl. In addition, the spectra obtained from elemental analysis revealed that the Cs, P, and Mg concentrations of these spots were clearly higher compared to that of other regions without spots (**Figure 1b**). By SEM-EDX analysis, it was shown previously that the Cs content of the mycelial root (old regions of the hyphae) of *P. ostreatus* is approximately 5-times higher than that of the mycelial tip (growing regions of the hyphae) (22). In addition, in the previous study on Cs accumulation by soil microorganism (23) concerning the transfer of Cs from soil to mushroom (24), the presence of white spots at regular intervals was also confirmed using the mycelia of a soil filamentous bacterium (*Streptomyces lividans*) by SEM, and semiquantitative elemental analysis by EDX revealed that the Cs and P contents in these spots were as high as in *P. ostreatus* mycelia used in the present study. K, a chemical analogue of Cs, and P, an essential element transported by arbuscular mycorrhizal fungi (AMF), have been suggested to influence the uptake, translocation, and transfer of radiocesium by AMF (25). It was demonstrated that the transport of ¹³⁴Cs was affected by P independently of the concentration of K, and increasing the concentration of P in the hyphal compartment resulted in higher uptake and accumulation of ¹³⁴Cs in roots and shoots of *M. truncatula* plants (26). From these results, it was considered that Cs trapped in *P. ostreatus* mycelia was accumulated in specific regions with higher levels of P and concentrated. As specific regions with higher levels of P are present in mycelia, vacuoles accumulating polyphosphate have been suggested (27). The white spots where

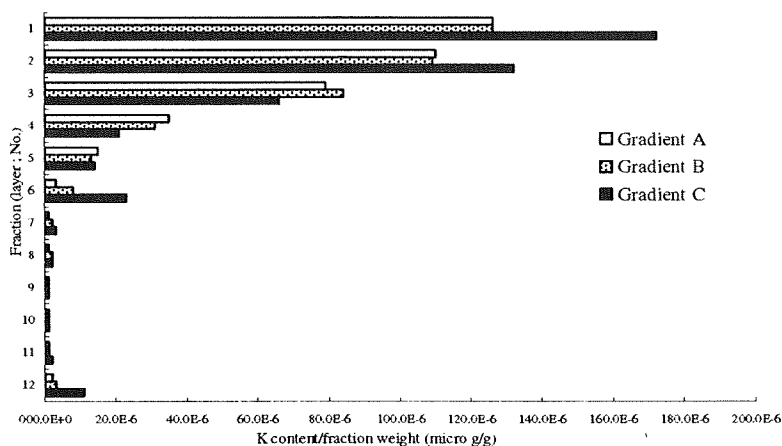


Figure 6. Distribution of K in each fraction control solution containing 5 mM KCl following density-gradient centrifugation.

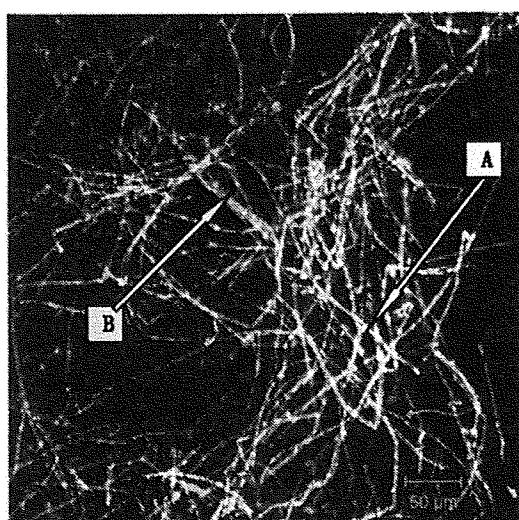


Figure 7. Image of 4',6-diamidino-2-phenylindole (DAPI)-stained *P. ostreatus*-Y1 mycelia cultured on a YM agar plate containing 15 mM CsCl with laser-scanning confocal microscopy. A, polyphosphate-DAPI (yellowish granular fluorescence) around 526 nm; B, DNA-DAPI (bluish granular fluorescence) around 456 nm.

the Cs and P contents were higher than other regions. observed in the mycelia cultivated with CsCl by SEM could be a result of Cs⁺ trapped by polyanionic polyphosphate.

Uptake of ¹³⁷Cs into *P. ostreatus* Cultivated Mycelia and Elution Fractions. The ¹³⁷Cs contents in mycelia and the elution fractions after washing were investigated. The rate of uptake of Cs into *P. ostreatus* mycelia cultured for seven days and elution by washing was investigated using ¹³⁷Cs. Figure 2 shows the ¹³⁷Cs concentrations in water used for ¹³⁷Cs elution and mycelia at each time point in the second wash that was retained after the first wash. Mycelia of *P. ostreatus* cultivated in YM liquid medium containing ¹³⁷Cs with shaking were harvested by centrifugation and washed with distilled water. In Figure 2, error bars indicates the mean value ± the standard deviation of three replicates. The ¹³⁷Cs concentrations in each 2 mL aliquot of water at 0, 30, 60, 90, and 180 min after the start of elution were almost constant. These results suggested that the elution of ¹³⁷Cs from mycelia into distilled water occurred rapidly after the start of elution and that the increase or decrease in the amount of ¹³⁷Cs eluted with time was small. The ¹³⁷Cs activities in the water used for the ¹³⁷Cs elution experiment were approximately 4–6 times higher than those in the mycelia after washing. The previous ¹³³Cs NMR studies showed that the

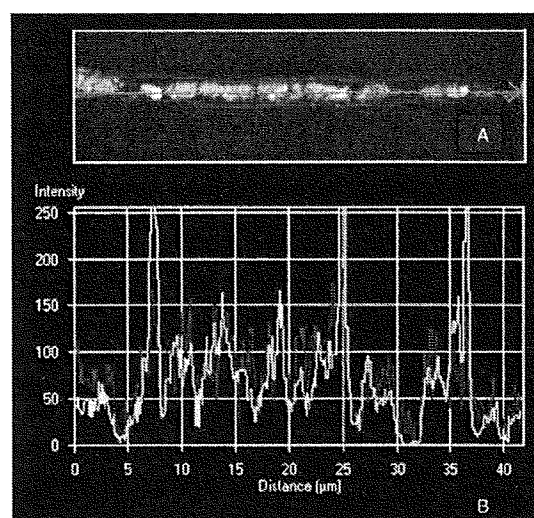


Figure 8. Image and fluorescence intensity caused by polyphosphate-4',6-diamidino-2-phenylindole (DAPI)-stained *P. ostreatus*-Y1 mycelia cultured on a YM agar plate containing 15 mM CsCl by laser-scanning confocal microscopy (526 nm). (A) Image of fluorescence from polyphosphate-DAPI in the mycelium. (B) Fluorescence intensity of the mycelium. The scale of the horizontal axis of the graph (B) is the same scale as the fluorescence microscopy image of *P. ostreatus* mycelia (A).

spectra exhibited two resonance signals arising from the mycelia and the fruiting bodies, whereas one signal was detected from CsCl standard solution (0 ppm) and the other lower magnetic field. This suggested that Cs existed in a different state in the samples (9, 18). Taking these results into consideration, part of the ¹³⁷Cs was concluded to be in its ionized form in the mycelia and could be eluted by washing, but the other Cs form was trapped by polyphosphate and remained in the mycelia even after washing.

Presence of Cs and K in *P. ostreatus* Mycelia Fraction as Assessed by Cell Fractionation. After density gradient centrifugation of the mycelial homogenate of *P. ostreatus*, 13 fractions were obtained. Yellowish suspensions, fractions 3 and 13 (sediment), were regarded as containing mitochondrial and vacuolar material, respectively (19, 20). However, confirmation of the composition by electron microscopy was not performed. The results of atomic absorption spectrometry of Cs and K in each fraction are shown in Figure 3 (for Cs obtained from four experiments) and Figure 4 (for K obtained from five experiments), respectively. In both graphs, the contents of Cs or K derived from *P. ostreatus* mycelia are expressed as concentra-

tions among the 13 fractions (Cs or K weight/fraction weight: $\mu\text{g g}^{-1}$), which consisted of 3-mL aliquots of fractions 1–12 obtained from the upper liquid layers and the sediment layer (fraction 13). The concentrations of Cs and K in the controls without mycelia are shown in **Figure 5** (for Cs obtained from duplicate experiments) and **Figure 6** (for K obtained from triplet experiments), respectively. As shown in **Figure 3**, it was found that the Cs concentration derived from *P. ostreatus* mycelia was higher in fractions 1–3. The Cs concentrations of the sediment containing vacuolar pellets were relatively less variable with the mean among 4 samples being $46.0\text{E}^{-6} \pm 7.5\text{E}^{-6} \mu\text{g g}^{-1}$. Similar to the results obtained with mycelia, a tendency toward clearly higher Cs concentrations in the upper fractions 1–3 was also observed in the control experiment of Cs. Subsequently, the Cs ratio, which compared the Cs content in each fraction to the total Cs content obtained by summing the values of all fractions and the sediment, showed that approximately 90% of Cs was present in fractions 1–3. Therefore, the Cs content in the lower nine liquid layers (fractions 4–12) was negligible. The existence of ionized Cs in the liquid layers was confirmed by comparison with the results of the Cs control experiment. It was considered that the larger amount of ionized Cs compared to the results of experiments on the uptake of ^{137}Cs into *P. ostreatus* cultivated mycelia and the water used for the ^{137}Cs elution experiment mentioned above can be attributed to the disruption of mycelia. However, the localization of Cs was also observed in the sediment-containing vacuolar pellets. It was suggested that Cs transferred into the vacuolar compartment could be trapped by polyanionic polyphosphate as mentioned above. NMR spectroscopy of both *P. ostreatus* mycelia and the fruiting body showed the existence of ionized Cs and another Cs form (9, 28). In addition, as the localizations of Cs and P were observed in the SEM-EDX experiment as mentioned above, polyphosphate could be related in Cs accumulation and localization in *P. ostreatus* mycelia. The distribution of K, which is a chemical analogue of Cs, derived from mycelia in each liquid fraction was slightly different from that of Cs, and no particularly high concentrations of K were observed in liquid fractions 1–12. However, the concentration of K in the sediment was higher in a similar manner to the results of Cs. Consistent with the results obtained for Cs, higher K concentrations in the upper liquid fractions 1–3 were also observed in the control experiment, showing that a different distribution of K was obtained with the results from mycelia. It was shown previously that the concentrations of K in the mycelia of *P. ostreatus* decreased with the increase in Cs concentrations in the media containing 0–15 mM Cs (17). Each K concentration in YM medium in the previous study and the present study was 0.02% K and 8.4 mM K (approximately 0.03% K), respectively, from the inherent content in the medium. It was considered that the above observations with K and Cs appear to show an inverse relationship, which could result from the effect of high Cs concentration (15 mM Cs) in the medium. However, it could not be clarified why the higher K concentration in the sediment was observed in a manner similar to the result of Cs distribution, and further investigation will be necessary.

Staining of *P. ostreatus* Mycelia Indicates the Presence of Polyphosphate. From the results mentioned above, it was decided to focus on the relationship between the presence of polyphosphate and Cs in *P. ostreatus* mycelia. Polyphosphate was detected by fluorescence microscopy using DAPI-stained mycelia. Fluorescence microscopy images of *P. ostreatus* mycelia cultured in medium containing 15 mM CsCl stained with DAPI are shown in **Figures 7** and **8**. As shown in **Figure**

7, yellowish granular fluorescence resulting from polyphosphate-DAPI (**A** in **Figure 7**) and bluish fluorescence resulting from DNA-DAPI (**B** in **Figure 7**) were observed in images of the mycelia. The aromatic fluorescent dye DAPI is a well-known reagent for the analysis of DNA, and the binding of DAPI to polyphosphate causes a shift in the fluorescence emission maximum from 456 to 526 nm (29). The fluorescence intensity around the region of yellowish granular fluorescence is shown in **Figure 8**. The scale of the horizontal axis of the graph is the same scale as the one in the fluorescence microscopy image of *P. ostreatus* mycelia. It can be noted that the fluorescent intensity is particularly strong at the granular sites. Thus, it was recognized that, in *P. ostreatus* mycelia, polyphosphate non-uniformly localizes in a granular fashion. Tijssen reported that the spectrum of yeast (*S. fragilis*)-bound DAPI had a maximum at about 526 nm, highly suggestive of a reaction with polyphosphate, and fluorescence microscopy confirmed the localization of the fluorescent DAPI-polyphosphate complex in yeast vacuoles (21). In this study, as shown by the results of SEM-EDX in *P. ostreatus* mycelia, Cs and P localized at the same sites as observed for the white spots, and therefore, a relationship between the fluorescence intensity of polyphosphate-DAPI and Cs in vacuoles or other organelles was inferred. A more detailed investigation of this relationship, for example, how the percentage of Cs included in polyphosphate relates to the total Cs content in mycelia, will be necessary in the future.

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Evaluation of Cesium-137 (^{137}Cs) and Elements Intake from Daily Diets in Residents of Kanagawa Prefecture, Japan

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The present study was undertaken to investigate the amounts of artificial radionuclides ingested through the daily diet by inhabitants of Kanagawa Prefecture, Japan. To this end, the level of cesium-137 (^{137}Cs) contained in the daily diets of females (combined samples from 5 females) aged 40–69 living in urban and suburban districts of Kanagawa Prefecture was measured. Furthermore, 11 elements (Ca, Cl, Cr, Cs, Fe, K, Mn, Na, Rb, Sc, and Zn) in the diets were quantified by instrumental neutron activation analysis. The study revealed that the intake of ^{137}Cs increased markedly in 1986, the year the Chernobyl accident occurred, and it tended to decrease gradually thereafter. In recent years, the reduction in the dietary intake of ^{137}Cs has been slowing down. When the intake of nutrients ingested by inhabitants of the two survey areas was compared with the Dietary Reference Intakes for Japanese (2005), the amount ingested in each of the two survey areas approximately satisfied or was slightly lower than the reference levels. Among others, the amount of Ca and Fe ingested in the urban district was particularly low (52 and 64% of the national reference, respectively). For many elements, the amount ingested was greater in the suburban than in the urban district. When the ingredients of the subjects' meals were classified into food groups, the meals prepared by suburban inhabitants were found to be composed of more diverse ingredients, covering all food groups, compared to those prepared by urban inhabitants. The daily food intake was also greater in the suburban district. When the daily dietary samples from individual subjects were analyzed separately, without being combined, both the level of ^{137}Cs and the amount of stable elements contained in the separate samples showed greater variations from day to day than those in the combined samples. There was no correlation between the dietary intake of ^{137}Cs and that of Cs.

Key words — daily diet, major and trace element, radionuclide, ingestion, gamma-ray spectrometry, instrumental neutron activation analysis

INTRODUCTION

In 1963, we began daily diet sampling (combined samples from 5 individuals) from adults living in 3 fixed monitoring areas in the middle of the southern part of Kanagawa Prefecture, *i.e.*, Hiratsuka City, Oiso Town (Naka County), and Ninomiya Town (Naka County, hereinafter referred to collectively as the “Hiratsuka area”), within the framework of the Japanese governmental survey on

the effects of nuclear bomb experiments. As defined above, the Hiratsuka area is composed of one middle-scale city and two towns of Kanagawa Prefecture, Japan. It is composed of an urban and a suburban district (a mixture of farming and fishing zones). From 1984 onwards, radioactivity analysis has also been carried out by us. Environmental radionuclides are incorporated into the human body via the diet and respiration, contributing to the internal exposure dose. According to the 2000 report of the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), the mean internal exposure dose of natural radionuclides via foods was 0.29 mSv y^{-1} , corresponding to about 1/4 of the exposure dose of inhaled radon,

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which accounts for the highest percentage of the total exposure. Although the report states that exposure to natural radionuclides accounts for most of the exposure, radiation accidents, *etc.*, are matters of concern as nuclear power-related facilities are operated worldwide. In the Chernobyl nuclear power plant accident that occurred in 1986 in the former Soviet Union, a huge amount of artificial radionuclides entered the atmosphere and spread extensively, causing environmental and food contamination on a global scale.

Triggered by this accident, in 1990 we expanded the daily diet radioactivity survey area, adding Yokohama City (an urban district of Kanagawa Prefecture) to the Hiratsuka area (two survey areas in total). Furthermore, for the 5-year period from 1990, the radioactivity survey was accompanied by a survey of the intakes of nutrients and trace elements by females aged 40–69 living in the urban and suburban districts.

In this paper, we describe mainly after 1990 and show the already reported papers for the former 1990. It will report on our analysis of the time-course of dietary cesium-137 (^{137}Cs) intake by inhabitants of Kanagawa Prefecture, presence/absence of differences in the intake of ^{137}Cs and elements between urban and suburban districts of Kanagawa Prefecture, association between various dietary ingredients and nutrient intake, association between artificial radionuclide and stable elements, and so on. Because radionuclide levels often vary greatly among daily dietary samples, we quantified ^{137}Cs and stable elements contained in diets for individual subjects in 1998 to analyze the relationship between inter-individual variance regarding radionuclide intake and the diet of individuals.

MATERIALS AND METHODS

Samples for Radionuclide Analysis —

Sampling Location: Kanagawa Prefecture is located in the southwest of Tokyo Metropolitan. Yokohama City is the Prefecture capital of Kanagawa. Hiratsuka City is a suburban district located in about 20 km southwest of Yokohama City. Oiso Town and Ninomiya Town are located in west side of Hiratsuka City.

Regional Samples: Daily diets were collected by the duplicate portion method together with simple daily menus from the following subjects twice a year (summer and winter): five adults living

in the Hiratsuka area (Hiratsuka City, Oiso Town, and Ninomiya Town, Kanagawa Prefecture), with a sampling period from 1984 to 2006, and five adults living in the Yokohama area (Konan-ku, Yokohama City, Kanagawa Prefecture), with a sampling period from 1990 to 2002. For each sampling occasion, the subjects were advised to prepare their usual menus, avoiding special menus. Bulky and fibrous foods in daily diets were cut into small pieces with a cooking knife. Materials usually not ingested, such as bone, skin, and seeds, were removed. Portions of diets were transferred to ceramic dishes for individual subjects and dried at 105°C for 10–12 hr in a circulating hot air drying oven, followed by 24-hr incineration at 450°C in an electric furnace. Ashes of samples from 5 subjects were mixed well to yield a sample for radionuclide analysis.

Individual Samples: The samples collected from 5 subjects in 1998 from each survey area were individually incinerated without being combined, to yield individual samples for radionuclide analysis.

Samples for Instrumental Neutron Activation Analysis — From each of the 40 samples for radionuclide analysis (20 samples collected in the summer and winter of 1990 through 1994 from Hiratsuka and Yokohama areas and 20 individual samples collected in the summer and winter of 1998), about 100 mg was taken, weighed precisely, and tightly packed into a polyethylene bag, to yield a sample for neutron activation analysis.

Standard Material for Comparison: A standard sample for atomic absorption spectrometry (Wako Pure Chemical Industries, Ltd., Osaka, Japan) was adsorbed onto cellulose filter paper No. 7 (Toyo Roshi Kaisha, Ltd., Tokyo, Japan), air-dried, and packed tightly into a polyethylene bag.

Samples for Accuracy Control of Artificial Radionuclide Analysis — The combined samples, collected in the summer of 1993 through 1998 from the Hiratsuka area, were used.

Samples for Accuracy Control of Neutron Activation Analysis — Each of standard reference materials (SRM) 1571 ORCHARD LEAVES (OL) (supplied by National Institute of Standards and Technology; NIST, Gaithersburg, MD, U.S.A.), SRM1548 TOTAL DIET (TD) (NIST), and CRM No. 1 PEPPERBUSH (PB) (supplied by National Institute for Environmental Studies; NIES, Tsukuba, Japan), about 300 mg was taken, weighed precisely and packed tightly into a polyethylene bag.

Radionuclide Analysis— Each combined or individual sample after incineration was packed into a container made of styrol resin U-8 (As One corporation, Osaka, Japan) and subjected to radionuclide quantification by gamma-ray spectrometry. For accuracy control, the samples we analyzed were sent to the Japan Chemical Analysis Center (JCAC), where they were also subjected to gamma-ray spectrometry.

Analysis of Stable Elements— Using a TRIGA II type atomic reactor of the Institute for Atomic Energy, Rikkyo University, samples were individually irradiated with neutrons for 1 min through an f-hole for nuclides with short half-lives. For nuclides with intermediate to long half-lives, the incinerated sample (packed into a polyethylene bag), the standard sample for comparison, and the standard sample for precision control were placed in one polyethylene capsule. The capsule was put on the rotary sample shelf and exposed to neutrons for 6–24 hr.

Instruments— The artificial radionuclides and the intermediate to long half-life nuclides produced by neutron activation were counted using a Ge semiconductor detector (pureGeCNVDS30-35195, Oxford Co., Oak Ridge, TN) connected to a multichannel analyzer (Multiport, Oxford Co.), followed by quantification using an analysis program (Gamma-truck or Assayer, Oxford Co.). Short half-life nuclides produced by neutron irradiation were counted using a Ge semiconductor detector (Ortec Co., Oak Ridge, TN) connected to a multichannel analyzer (MCA) (M7800, Seiko EG&G, Tokyo, Japan), followed by the quantification of stable elements using an analysis program (Emulation Program, Seiko EG&G).

Counting Period— Artificial radionuclides were counted for 80000–324900 sec. Short half-life nuclides produced by neutron activation (^{38}Cl , ^{49}Ca , and ^{56}Mn) were counted for 5 min. Intermediate half-life nuclides (^{24}Na , and ^{42}K) were left standing for about one week after irradiation and subsequently counted for 10000–50000 sec. Long half-life nuclides (^{51}Cr , ^{134}Cs , ^{59}Fe , ^{86}Rb , ^{46}Sc , and ^{65}Zn) were left standing for about one month after irradiation to induce the decay of intermediate half-life nuclides and subsequently counted for 80000 sec.

RESULTS

Precision Control

The precision control of artificial radionuclides in daily diets has been conducted at a testing/research facility of a major local government in Japan and JCAC since 1993. Table 1 shows the results of the radioactivity analysis of daily diets in 1993 through 1998. The results were consistent between the two institutions, verifying the quality of analysis. However, the analytical values of ^{137}Cs obtained at the JCAC in 1994, 1995, and 1997 were not significant because they were within three times the error ($3 \times$ error) arising from differences in measurement conditions (*e.g.*, collection time). In Tables 1–7, the count was expressed as ND (not detectable) when the net count was less than three times the count error. In neutron activation analysis, the 2 types of standard sample were placed into a capsule, identical to the capsule used for test samples, in each session of neutron irradiation, and

Table 1. Comparative Radioactivity Analysis of Daily Diet (unit: $\text{Bq man}^{-1}\text{day}^{-1}$)

Year	Analytical organization	Collection time (s)	^{137}Cs	^{40}K
			NET \pm SE	NET \pm SE
1993	This work	324900	0.047 \pm 0.013	73.2 \pm 0.5
	JCAC ^{a)}	71967	0.046 \pm 0.012	71.5 \pm 0.7
1994	This work	244200	0.046 \pm 0.014	72.4 \pm 2.2
	JCAC ^{a)}	71665	ND	75.3 \pm 0.8
1995	This work	150000	0.051 \pm 0.010	63.3 \pm 0.4
	JCAC ^{a)}	70057	ND	63.7 \pm 0.7
1996	This work	150000	0.075 \pm 0.0090	73.3 \pm 0.5
	JCAC ^{a)}	71678	0.076 \pm 0.015	73.2 \pm 0.7
1997	This work	100000	0.063 \pm 0.012	68.9 \pm 0.6
	JCAC ^{a)}	72146	ND	82.7 \pm 0.8
1998	This work	100000	0.11 \pm 0.013	82 \pm 0.6
	JCAC ^{a)}	71289	0.061 \pm 0.014	73.2 \pm 0.7

ND: not detectable, a) Japan Chemical Analysis Center.

Table 2. Analytical Results of Elements in Certified Reference Materials (Unit: mgkg⁻¹)

Element	NIST SRM1571 Orchard Leaves				Deflection with certified value (%)	NIST SRM1548 Total Diet				Deflection with certified value (%)
	Analytical value	S.D.	CV (%)	<i>n</i>		Analytical value	S.D.	CV (%)	<i>n</i>	
Ca	20500 20900	± 1650 ± 300	8.05	6	1.91	1600 1740	± 150 ± 70	9.38	4	8.05
Cl	784 (690)	± 51	6.51	6		8690 8700	± 402 ± 400	4.63	4	0.11
Cr	2.3 2.6	± 0.24 ± 0.30	10.43	6	11.54	ND —				
Cs	0.045 (0.04)	± 0.019	42.22	5		0.027 (0.014)	± 0.012	44.44	4	
Fe	287 300	± 43 ± 20	14.98	6	4.33	41 32.6	± 7.8 ± 3.6	19.02	4	25.77
K	13900 14700	± 726 ± 300	5.22	6	5.44	4130 6060	± 2050 ± 280	49.64	4	31.85
Mn	84 91	± 3.1 ± 4	3.69	6	7.69	ND 5.2				—
Na	103 82	± 27 ± 6	26.21	6	25.61	5820 6250	± 361 ± 260	6.20	4	6.88
Rb	11 12	± 1.6 ± 1	14.55	6	8.33	4.6 (4.8)	± 0.78	16.96	4	
Sc	0.060 [0.047–0.066]	± 0.0049	8.17	6		ND				
Zn	22 25	± 2.1 ± 3	9.55	6	12.00	30.7 30.8	± 1.2 ± 1.1	3.91	4	0.32
Element	NIES No.1 Pepperbush				Deflection with certified value (%)					
	Analytical value	S.D.	CV (%)	<i>n</i>						
Ca	13400 13800	± 832 ± 700	6.21	2	2.90					
Cl	155	± 1.2	0.77	2						
Cr	0.65 (1.3)	± 0.20	31.18	2						
Cs	1.2 (1.2)	± 0.028	2.33	2						
Fe	208 205	± 5.9 ± 17	2.82	2	1.46					
K	12100 15100	± 341 ± 700	2.82	2	19.87					
Mn	1810 2030	± 11 ± 170	0.60	2	10.84					
Na	91 106	± 12 ± 13	13.62	2	14.15					
Rb	73 75	± 0.78 ± 4	1.08	2	2.67					
Sc	ND									
Zn	322 340	± 32.7 ± 20	10.17	2	5.29					

Upper row: our result, Lower row: certified value, (): uncertified or reference value, []: literature value¹⁾, ND: not detectable.

Table 3. Element Intake in Hiratsuka Area, Kanagawa Prefecture, Japan (1990–1994) [Unit: mgday⁻¹, Bq man⁻¹day⁻¹ (¹³⁷Cs)]

		Hiratsuka Area					
		Summer					
Classification		1990	1991	1992	1993	1994	Avg.
Mineral	Ca	529	317	352	491	265	391
Trace elements	Cr	0.0369	0.0554	0.0485	0.00888	0.0298	0.0359
	Mn	2.59	2.17	2.70	1.94	2.68	2.42
	Fe	6.35	14.8	8.56	9.51	11.1	10.1
	Zn	6.64	8.05	7.74	7.46	7.31	7.44
Electrolytes	Na	2980	3350	2900	2890	3930	3210
	Cl	4060	4480	3950	4020	5340	4370
	K	1670	2150	1980	1700	1990	1900
Other trace elements	Rb	1.66	1.80	1.68	1.87	1.70	1.74
	Sc	0.000497	0.000723	0.000372	0.000748	0.00174	0.000816
	Cs	0.00608	0.00611	0.00494	0.00610	0.00731	0.00611
Radionuclide	¹³⁷ Cs	ND	0.059	0.052	0.047	0.046	0.051
		Winter					
Classification		1990	1991	1992	1993	1994	Avg.
Mineral	Ca	240	456	485	362	555	420
Trace elements	Cr	0.0871	ND	0.0800	0.0298	0.0582	0.0638
	Mn	1.72	1.63	2.13	1.69	2.18	1.87
	Fe	6.63	5.21	6.97	6.51	6.60	6.4
	Zn	5.74	6.05	5.32	5.94	6.56	5.92
Electrolytes	Na	2030	2930	2990	3010	2980	2790
	Cl	2780	4170	4500	4480	4460	4080
	K	1630	900	1830	1810	1810	1600
Other trace elements	Rb	1.91	1.94	2.11	1.93	2.03	1.98
	Sc	0.000628	0.000297	0.000567	0.000620	0.000439	0.000510
	Cs	0.00681	0.00700	0.00862	0.00649	0.00691	0.00717
Radionuclide	¹³⁷ Cs	0.080	0.12	0.076	0.064	0.12	0.092
Classification		Range		Avg.	S.D.	CV (%)	n
Mineral	Ca	240	— 555	405	112	27.71	10
Trace elements	Cr	ND	— 0.0871	0.0483	0.0251	51.92	9
	Mn	1.63	— 2.70	2.14	0.406	18.96	10
	Fe	5.21	— 14.8	8.22	2.90	35.23	10
	Zn	5.32	— 8.05	6.68	0.925	13.84	10
Electrolytes	Na	2030	— 3930	3000	466	15.54	10
	Cl	2780	— 5340	4220	645	15.28	10
	K	900	— 2150	1750	338	19.33	10
Other trace elements	Rb	1.66	— 2.11	1.86	0.152	8.14	10
	Sc	0.000297	— 0.00174	0.000663	0.000405	61.14	10
	Cs	0.00494	— 0.00862	0.00664	0.000966	14.56	10
Radionuclide	¹³⁷ Cs	ND	— 0.12	0.074	0.029	38.91	9

they were analyzed simultaneously for the purpose of accuracy control. The results are shown in Table 2. The values for each standard sample were approximately consistent with the certified values. Consistency of the analytical values in this study with the certified values at a 10% or less deflection was noted for 5 of the 8 elements in SRM1571 (OL) analysis: calcium (Ca), iron (Fe), potassium (K), manganese (Mn), rubidium (Rb); for 4 of the 7 elements in SRM1548 (TD) analysis: Ca, chlo-

rine (Cl), sodium (Na), and zinc (Zn); and for 4 of the 7 elements in NIES No. 1 (PB) analysis: Ca, Fe, Rb, and Zn. Elements whose analytical values were out of the certified range (plus uncertainty margin) were Cr and Zn in SRM1571 analysis. The analytical values for Na in SRM1571 analysis, Fe and K in SRM1548 analysis, and Na in NIES No. 1 analysis were consistent with the certified values (within the range of statistical error). The analytical value for scandium (Sc) in SRM1571 (OL) was presented

Table 4. Element Intake in Yokohama Area, Kanagawa Prefecture, Japan (1990–1994) [Unit: mgday⁻¹, Bq man⁻¹day⁻¹ (¹³⁷Cs)]

		Yokohama Area					
		Summer					
Classification		1990	1991	1992	1993	1994	Avg.
Mineral	Ca	294	216	330	455	375	334
Trace elements	Cr	0.0954	0.0653	0.0300	0.0255	0.0183	0.0469
	Mn	2.36	1.11	1.77	3.93	1.86	2.21
	Fe	5.71	4.66	5.26	9.72	5.55	6.18
	Zn	5.20	4.31	5.80	11.3	7.28	6.78
Electrolytes	Na	2580	2280	2920	3610	2500	2780
	Cl	3540	3030	3840	4710	3340	3690
	K	1330	1860	1750	2370	1390	1740
Other trace elements	Rb	1.24	1.28	1.54	2.38	1.57	1.60
	Sc	0.000214	0.000312	0.000324	0.000742	0.000357	0.000390
	Cs	0.00443	0.00518	0.00534	0.00765	0.00538	0.00560
Radionuclide	¹³⁷ Cs	0.040	0.12	0.051	0.059	0.050	0.064
		Winter					
Classification		1990	1991	1992	1993	1994	Avg.
Mineral	Ca	274	272	372	305	197	284
Trace elements	Cr	0.0298	0.221	0.0911	ND	ND	0.114
	Mn	1.52	1.74	1.23	1.91	1.31	1.54
	Fe	3.96	7.57	5.27	5.53	5.09	5.48
	Zn	4.35	4.96	4.90	7.25	5.40	5.37
	Na	2270	2370	2390	1900	2480	2280
Electrolytes	Cl	3270	3480	3580	2730	3570	3330
	K	915	1090	1290	1190	1170	1130
	Rb	1.68	1.61	1.62	1.70	1.60	1.64
Other trace elements	Sc	0.000349	0.000259	0.000209	0.000310	0.000161	0.000258
	Cs	0.00368	0.00562	0.00485	0.00652	0.00740	0.00561
	Radionuclide	¹³⁷ Cs	0.069	0.069	0.050	0.034	0.076
Classification		Range		Avg.	S.D.	CV (%)	n
Mineral	Ca	197	— 455	309	77.5	25.09	10
Trace elements	Cr	ND	— 0.221	0.0721	0.0673	93.47	8
	Mn	1.11	— 3.93	1.87	0.812	43.33	10
	Fe	3.96	— 9.72	5.83	1.65	28.23	10
	Zn	4.31	— 11.3	6.08	2.11	34.77	10
	Na	1900	— 3610	2530	459	18.14	10
Electrolytes	Cl	2730	— 4710	3510	525	14.97	10
	K	915	— 2370	1440	436	30.25	10
	Rb	1.24	— 2.38	1.62	0.309	19.02	10
Other trace elements	Sc	0.000161	— 0.000742	0.000324	0.000161	49.69	10
	Cs	0.00368	— 0.00765	0.00561	0.00126	22.41	10
	Radionuclide	¹³⁷ Cs	0.034	— 0.12	0.062	0.024	39.43

with literature values¹⁾ because no certified or reference value was available regarding this element. These findings ensured the quality of neutron activation analysis.

Radionuclide Intake from Daily Diets

Gamma-ray-emitting radionuclides detected from daily dietary samples collected from the 2 areas of Kanagawa Prefecture in 1984–2006 were the artificial radionuclides ¹³⁷Cs and cesium-134

(¹³⁴Cs) and natural radionuclide potassium-40 (⁴⁰K), etc.^{2–4)} Figure 1 shows the time-course of ¹³⁷Cs intake. Its intake increased markedly in 1986 because of the Chernobyl nuclear power plant accident taking place in the suburbs of Kiev in the former Soviet Union in the same year. After that year, the intake of ¹³⁷Cs tended to decrease overall despite small variations (increases and decreases) from year to year. Artificial radionuclides produced and emitted into the environment as a result of

Table 5. Dietary Reference Intakes in Japan for 2005–2009 (unit: mgday⁻¹)

Classification		EAR	RDA ^{a)}	RDA ^{b)}	AI ^{a)}	AI ^{b)}	DG ^{a)}	DG ^{b)}	UL ^{a)}	UL ^{b)}	DG (high blood pressure prevention)
Mineral	Ca	—	—	—	600	700	—	600	—	2300	—
Trace elements	Cr	0.025	0.030	0.030	—	—	—	—	—	—	—
	Mn	—	—	—	3.5	3.5	—	—	11	11	—
	Fe	9	10.5	10.5	—	—	—	—	40	45	—
	Zn	6	7	7	—	—	—	—	30	30	—
Electrolytes	Na	600	—	—	—	—	< 3100	< 3100	—	—	—
	Cl ^{c)}	910	—	—	—	—	< 4900	< 4900	—	—	—
	K	—	—	—	1600	1600	2800	3100	—	—	3500

a): age 30–49, standard value for women, b) age 50–69, standard value for women, c) converted value from the sodium chloride equivalent, EAR: estimated average requirement, RDA: recommended dietary allowance, AI: adequate intake, DG: tentative dietary goal for preventing lifestyle-related diseases, ND: not detectable.

atmospheric nuclear tests before the Chernobyl accident can be detected even at present, although in trace amounts, from environmental samples collected in Japan, such as fallout and soil and seabed sediments. The level of ¹³⁷Cs intake from daily diets has been approximately constant in recent years. Artificial radionuclides detected in Japanese environments are primarily nuclear fission products of atmospheric tests performed by the People's Republic of China.⁵⁾ In Japan, the phenomenon called "spring maximum" was often seen across the country before 2000.⁶⁾ This phenomenon pertains to the descending of radioactive substances from the stratosphere to the troposphere in spring, resulting in the detection of high levels of radioactive substances in the monthly fallout.⁶⁾ In the late 1990s, this phenomenon was seen in less regions and the level of radioactive substances detected became lower. After 2000, however, ¹³⁷Cs began to be detected from monthly fallouts also in high altitude districts and areas not facing the Sea of Japan in February and some other months when yellow sand from China reaches Japan.^{7,8)} It seems likely that the daily dietary ¹³⁷Cs level remains constant because ¹³⁷Cs continues to be supplied in the form of fallout, although in low levels. The daily dietary ¹³⁷Cs level recorded in Kanagawa Prefecture in recent years was close to the level of ¹³⁷Cs contained in the total diet samples collected in Japan by Sugiyama *et al.* using the market basket method (0.012–<0.077 Bq day⁻¹).⁹⁾ In the Hiratsuka area, the mean ¹³⁷Cs intake was 0.080 ± 0.056 Bq man⁻¹day⁻¹, with a median of 0.065 Bq man⁻¹day⁻¹.

The man ¹³⁷Cs intake from daily diets in Kanagawa Prefecture during the five-year period from 1990 to 1994, during which time the quantitative analysis of stable elements was performed, was

0.067 ± 0.027 Bq man⁻¹day⁻¹ (median: 0.059 Bq man⁻¹day⁻¹), similar to the national level for the same period (mean: 0.061 ± 0.040 Bq man⁻¹day⁻¹, median: 0.050 Bq man⁻¹day⁻¹).⁶⁾ The maximum level in Japan during this period was 0.34 Bq man⁻¹day⁻¹ (recorded in a high altitude area).⁶⁾ In this area, the level of radioactive fallout derived from Chinese atmospheric nuclear tests or yellow sand dispersion is very high for meteorological reasons, and this is probably reflected in the daily diets.

¹³⁷Cs intake in the Hiratsuka area for this period averaged 0.073 ± 0.029 Bq man⁻¹day⁻¹ (median: 0.064 Bq man⁻¹day⁻¹). The same parameter in the Yokohama area for the same period averaged 0.055 ± 0.024 Bq man⁻¹day⁻¹ (median: 0.056 Bq man⁻¹day⁻¹). Thus, ¹³⁷Cs intake during this period tended to have been smaller in Yokohama area than in Hiratsuka area.

¹³⁴Cs is a characteristic radionuclide released into environments as a result of the fire and explosion of the nuclear reactor during the Chernobyl accident. Immediately after the accident, it was detected in various foods and environmental samples.¹⁰⁾ However, since its half-life is short (2.06 years), it was not detected in daily diets in the winter of 1986 in Kanagawa Prefecture, although it had been detected from daily diets in the same prefecture in July of the same year (0.12 Bq man⁻¹day⁻¹).¹⁰⁾

Stable Elements in Daily Diets

Food Intake Weight and Age

Stable elements were surveyed in 100 subjects in total during the five-year period. All subjects in the Hiratsuka area were housewives in their 40–60s (mean: 52.5 ± 5.8 years). They were mostly wives of white collar workers, but some were living near farmland or a fishing port, or engaged in

Table 6. Individual Elements Intakes in Hiratsuka Area, Kanagawa Prefecture, Japan (1998) [Unit: mgday⁻¹, Unit: Bq man⁻¹day⁻¹ (¹³⁷Cs)]

		Hiratsuka Area					
		Summer					
Classification	person No.	No. 1	No. 2	No. 3	No. 4	No. 5	Avg.
Mineral	Ca	576	587	506	571	656	579
Trace elements	Mn	3.95	2.81	2.07	2.89	2.60	2.86
	Fe	4.62	6.39	5.26	5.34	5.79	5.48
	Zn	6.10	7.42	7.97	9.46	6.97	7.58
Electrolytes	Na	4270	2560	3720	3780	2590	3380
	Cl	6800	4130	5860	5880	4060	5350
	K	2230	2260	2340	2790	2670	2460
Other trace elements	Rb	1.88	2.27	2.07	2.50	1.95	2.13
	Sc	0.000222	0.000312	0.000410	0.000360	0.000261	0.000313
	Cs	0.00769	0.00898	0.00944	0.0129	0.00583	0.00897
Radionuclide	¹³⁷ Cs	0.090	0.098	0.13	0.22	0.080	0.12
		Winter					
Classification	person No.	No. 1	No. 2	No. 3	No. 4	No. 5	Avg.
Mineral	Ca	184	798	409	1000	764	631
Trace elements	Mn	2.25	2.70	2.23	7.17	2.46	3.36
	Fe	3.43	8.77	6.78	46.3	6.59	14.4
	Zn	5.88	6.90	7.18	9.93	6.88	7.35
Electrolytes	Na	1810	2370	3860	3230	2790	2810
	Cl	2230	3430	5490	4210	4080	3890
	K	770	2970	3110	4650	2550	2810
Other trace elements	Rb	0.889	2.27	1.50	3.13	2.41	2.04
	Sc	0.000227	0.000484	0.000403	0.0133	0.00231	0.00334
	Cs	0.00562	0.00646	0.00535	0.0154	0.0115	0.00887
Radionuclide	¹³⁷ Cs	ND	ND	ND	0.29	ND	0.29
Classification	person No.	Range		Avg.	S.D.	CV (%)	<i>n</i>
Mineral	Ca	184	— 1000	605	223	36.91	10
Trace elements	Mn	2.07	— 7.17	3.11	1.52	48.80	10
	Fe	3.43	— 46.3	9.93	12.9	129.54	10
	Zn	5.88	— 9.93	7.47	1.32	17.67	10
Electrolytes	Na	1810	— 4270	3100	793	25.57	10
	Cl	2230	— 6800	4620	1360	29.44	10
	K	770	— 4650	2630	960	36.52	10
Other trace elements	Rb	0.89	— 3.13	2.09	0.603	28.89	10
	Sc	0.000222	— 0.0133	0.00183	0.00408	223.03	10
	Cs	0.00535	— 0.0154	0.00892	0.00342	38.40	10
Radionuclide	¹³⁷ Cs	ND	— 0.29	0.15	0.085	56.14	6

agriculture or fishing. In the Yokohama area, the subjects changed only once during the 5-year period. For this reason, the total number of subjects was 50, but the actual number was 9, all of whom belonged to white collar worker's families living in apartments in urban districts. In the Yokohama area, all subjects except for one male student living with his family (who collaborated with the survey for a short period) were housewives in their 30–50 s, with a mean age of 42.1 ± 5.3 years. In the Hiratsuka area, the weight of the daily food intake averaged $2.053 \text{ kg} \pm 0.405 \text{ kg}$ (coefficient of variation

(CV): 19.75%), with the maximum being 3.089 kg and the minimum being 1.174 kg. In the Yokohama area, it averaged $1.808 \text{ kg} \pm 0.345 \text{ kg}$ (CV: 19.08%), with the maximum being 2.683 kg and the minimum being 1.264 kg. The mean weight of the diet was thus 0.2 kg greater in the Hiratsuka area (higher mean age of subjects) than in the Yokohama area. The intake in the Hiratsuka area was similar to the weight presented in the National Nutrition Survey performed every year ($2.041 \text{ kg man}^{-1} \text{ day}^{-1}$ in 2001), while that in the Yokohama area was smaller. A comparison with the National Nutrition Survey

Table 7. Individual Elements Intakes in Yokohama Area, Kanagawa Prefecture, Japan (1998) [Unit: mgday⁻¹, Unit: Bq man⁻¹day⁻¹ (¹³⁷Cs)]

Yokohama Area							
Summer							
Classification	person No.	No. 1	No. 2	No. 3	No. 4	No. 5	Avg.
Mineral	Ca	374	464	420	322	731	462
Trace elements	Mn	1.12	2.53	3.57	2.53	2.86	2.52
	Fe	4.08	7.49	8.46	4.62	2.31	5.39
	Zn	5.04	7.44	8.00	8.23	6.31	7.00
Electrolytes	Na	1790	4150	2580	2860	1440	2560
	Cl	2540	6190	3860	3770	1590	3590
	K	1420	2680	1810	2130	1920	1990
Other trace elements	Rb	0.954	1.67	1.27	1.29	1.52	1.34
	Sc	0.000167	0.000352	0.000207	0.000226	0.000141	0.000219
	Cs	0.00613	0.00769	0.00456	0.00436	0.00448	0.00544
Radionuclide	¹³⁷ Cs	ND	0.078	ND	0.055	ND	0.067
Winter							
Classification	person No.	No. 1	No. 2	No. 3	No. 4	No. 5	Avg.
Mineral	Ca	201	513	320	492	527	411
Trace elements	Mn	2.64	3.42	4.69	4.03	2.75	3.51
	Fe	4.60	6.16	4.38	8.58	3.73	5.49
	Zn	5.03	7.17	9.17	7.73	8.41	7.50
Electrolytes	Na	1790	6950	1720	4040	5520	4000
	Cl	2310	9820	2580	6030	8020	5750
	K	1740	ND	1480	1820	1530	1640
Other trace elements	Rb	1.63	1.51	1.60	1.62	1.40	1.55
	Sc	0.000363	0.000550	0.000312	0.000303	0.000298	0.000365
	Cs	0.00313	0.00638	0.00587	0.00680	0.00732	0.00590
Radionuclide	¹³⁷ Cs	ND	ND	0.25	0.085	ND	0.17
Classification	person No.	Range		Avg.	S.D.	CV (%)	n
Mineral	Ca	201	— 731	436	146	33.38	10
Trace elements	Mn	1.12	— 4.69	3.01	0.98	32.44	10
	Fe	2.31	— 8.58	5.44	2.13	39.12	10
	Zn	5.03	— 9.17	7.25	1.40	19.24	10
Electrolytes	Na	1440	— 6950	3280	1850	56.40	10
	Cl	1590	— 9820	4670	2740	58.67	10
	K	1420	— 2680	1840	389	21.13	9
Other trace elements	Rb	0.954	— 1.67	1.45	0.223	15.41	10
	Sc	0.000141	— 0.000550	0.000292	0.000118	40.50	10
	Cs	0.00313	— 0.00769	0.00567	0.00148	26.05	10
Radionuclide	¹³⁷ Cs	ND	— 0.25	0.12	0.090	76.57	4

ND: not detectable.

for the period corresponding to the present survey was not performed, because the National Nutrition Survey for this period takes into account the water used for cooking.

Element Intake by Area

In the present study, the concentration of several minerals and trace elements, present as nutrients in daily dietary samples, was measured, followed by the calculation of their daily intake from diets per person. The Ministry of Health, Labour and Welfare (MHLW) presented the reference daily intakes

per person of various nutrients in the 'Dietary Reference Intakes from Japanese (2005).' These reference values are applicable to the 5 years from 2005 to 2009. Tables 3, 4, and 5 shows the results of our analysis of elements in comparison with the reference values contained in the said MHLW reference. For Rb, Sc, and Cs (lower column), no national reference values are available, but our data on these elements are presented here as reference information together with data on the intake of the artificial radionuclide ¹³⁷Cs. The MHLW established the following 5 indices for nutrient intake: esti-

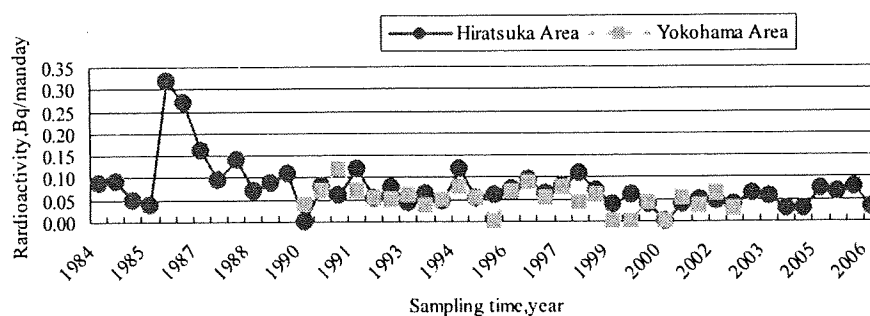


Fig. 1. ¹³⁷Cs Intake in Daily Diet in Kanagawa Prefecture

mated average requirement (EAR), recommended dietary allowance (RDA), adequate intake (AI), tentative dietary goal for preventing lifestyle-related diseases (DG), and the tolerable upper intake level (UL). In the present survey, the intake of Ca was slightly lower than the national reference. The intake of Zn (a trace element) in both survey areas satisfied the national reference level (EAR). It was close to the value reported by Cho *et al.* (females: 6.0 mgday^{-1}).¹¹⁾ The intake of iron (Fe) was 64–91% of the EAR, and tended to be smaller in the Yokohama area. Regarding this, Okada *et al.*¹²⁾ reported on the basis of a survey of daily diets during infancy that a group with a lower Zn intake had a lower energy intake and showed an insufficient intake of Fe and Ca as well. In the present survey, Zn intake satisfied the reference level, while intakes of Fe and Ca tended to be insufficient. The intake of Mn, for which the MHLW established AI and UL, was 54–60% of the AI. The Mn intake was slightly lower than the value reported by the World Health Organization (WHO) ($2.2\text{--}8.6 \text{ mgday}^{-1}$).¹³⁾ Intakes of Na (an electrolyte) and Cl (a component of sodium chloride) were lower than the DG in both areas, probably reflecting a tendency to refrain from taking dietary sodium chloride. The Cl intake was calculated by conversion of the reference sodium chloride intake. The K intake was approximately equal to the AI. The mean Cr intake was 1.6–2.4 times as large as the RDA, but its variation coefficient was high. The Cr intake never exceeded the UL (0.25 mgday^{-1}) specified in the '6th Revision Recommended Dietary Allowances for Japanese: Dietary Intake References.'

Rb (not serving as a nutrient) resembles K and Cs in terms of its chemical properties and has been attracting close attention as an element whose kinetics in vivo possibly resemble those of radioactive cesium.^{14–16)} The intake of Rb did not differ

between the two survey areas. Sc is derived from the Earth's crust, abundantly contained with Fe in basic rocks, such as basalt, and abundant in soil in Eastern Japan which is rich in basalt.¹⁷⁾ In soil, *etc.*, the concentration of Sc closely correlates with that of Fe. Although the level of Sc contained in dietary samples was very low, it was also detected in vegetables, dairy products, *etc.*, at a similar level, suggesting that Sc is taken up from the soil into plants and animals. When the correlation of the intake was analyzed between Rb and Cs and between Fe and Sc using the 20 analyzed samples, a correlation was noted between Rb and Cs ($r = 0.691$) and between Fe and Sc ($r = 0.660$). It seems likely that similarities exist between Rb and Cs and between Fe and Sc in terms of the likelihood of transfer to foods.

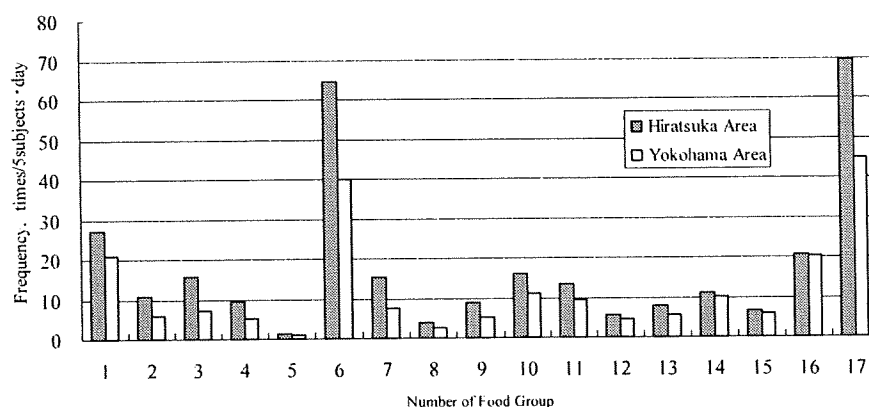
In inter-area comparison, the intakes of all elements, excluding intake during the summer of 1993 in Hiratsuka area, were greater in the Hiratsuka area than in the Yokohama area. The mean intake of each of the 7 elements, except for Cr, was about 1.4 times larger in the Hiratsuka than in the Yokohama area. The mean weight of the diet in the Hiratsuka area was about 1.1 times that of the Yokohama area. It therefore seems likely that some factors other than the weight of food intake are involved in the greater intake of elements in the Hiratsuka area.

Relationship between Meal Ingredients and Element Intake

The ingredients described in all 100 menus prepared during the 5-year survey period were classified based on the 5th Revision Standard Tables of Food Consumption in Japan. Table 8 shows the classification of the main foods. Since the frequency of use was low for some foods groups, the mean number of ingredients used for each food group per sample (combined samples from 5 subjects) is presented for each survey area in Fig. 2. Food group

Table 8. The Main Food Classification in the Fifth Revision Standard Tables of Food Composition in Japan

Number	Classification	Main food
1	Grain	Wheat, bread, noodles, rice, <i>etc.</i>
2	Potatoes and powder	Sweet potato, potato, paste made from arum root, starch, Harusame, <i>etc.</i>
3	Sugar	Sugar, honey, maple syrup, <i>etc.</i>
4	Beans	Kidney bean, soybean, bean curd, natto, soymilk, and Yuba, <i>etc.</i>
5	Seeds	Almond, ginkgo nut, chestnut, sesame, peanut, peanut butter, <i>etc.</i>
6	Vegetables	Cucumber, radish, onion, eggplant, carrot, green pepper, spinach, <i>etc.</i>
7	Fruits	Plum, pickled ume, mandarin orange, persimmon, apple, pear, peach, <i>etc.</i>
8	Mushrooms	Enokitake, shiitake, shimeji, hiratake, matsutake, kikurage, <i>etc.</i>
9	Algae	Green laver, laver, seaweed, hijiki, wakame, <i>etc.</i>
10	Seafood	Fish, shellfish, shrimp, crab, squid, octopus, fish paste products, <i>etc.</i>
11	Meat and poultry	Beef, pork, ham, bacon, sausage, chicken, frog, bees, <i>etc.</i>
12	Eggs	Quail egg, egg, egg-tofu, grilled egg, <i>etc.</i>
13	Milk	Fresh milk, process milk, powder milk, vegetable fat, cheese, ice cream, <i>etc.</i>
14	Fats and oils	Sesame oil, soybean oil, mixture oil, suet, butter, margarine, <i>etc.</i>
15	Cakes	Japanese-style confection, Danish pastries, cakes, dessert cakes, chocolates, <i>etc.</i>
16	Beverages	Alcohol, tea, coffee, cocoa, seaweed tea, carbonated drinks, <i>etc.</i>
17	Seasonings and spices	Salt, vinegar, dashi, mayonnaise, soy sauce, miso, peppers, <i>etc.</i>
18	Processed foods	Pre-packaged curry, frozen gyoza, frozen hamburg steak, frozen pilau, <i>etc.</i>

**Fig. 2.** Average Use Frequency of Ingredients in a Sample (for 5 persons)

The number of X-axis corresponds to Table 4. No. 18 (Processed foods) was excluded because its assessment in the menus was difficult.

18 was excluded from this analysis because its assessment in the menus was difficult. Group 17 includes seasonings, and these seasonings were used in most menus, resulting in a high frequency of use in both Hiratsuka and Yokohama areas. The frequency of use was marked for group 6 (vegetables) in both survey areas. Although the method of food grouping adopted in the above-mentioned study by Sugiyama⁹⁾ differs from that employed in the present study, Sugiyama reported that the contribution of other vegetables (vegetables excluding greenish-yellow vegetables), mushrooms, and algae to the ¹³⁷Cs intake was high. Among others, mushrooms are known to contain high levels of ¹³⁷Cs.^{18–24)} The contribution of group 6 to the in-

take of stable elements also seems to be high. During the survey period, ¹³⁷Cs was often detected from imported foods, but it was not possible to identify imported foods in the menus.

For all food groups, the number of ingredients used for the menu was greater in the Hiratsuka area than in the Yokohama area. Considering the use of diverse ingredients, it seems likely that the amounts of minerals, *etc.*, ingested in the Hiratsuka area were close to those shown in the references, and that these elements were ingested appropriately in this area. In the Yokohama area, a mean age was about 10 years lower than that for the Hiratsuka area. Although there were few elements whose intake differed markedly from the references, the intake of

Cr was 0.5 to 3.5 times higher than the reference (RDA, $0.030 \text{ mg day}^{-1}$) in both areas, indicating a large variance among different samples. Leaching from pans, kitchen knives, *etc.*, is also possible, but the exact cause of this variation has not been identified.

Regarding the 11 elements, we compared the data in summer with those in winter for the Hiratsuka area. This comparison revealed 7 elements (Na, Cl, *etc.*) whose intake tended to be larger during summer. When the frequency of the use of ingredients in the Hiratsuka area was compared between summer and winter, the use of vegetables during summer was more frequent than that during winter in all survey years other than 1993. During the summer, the frequency of use was also high for algae, fats and oils, beverages, *etc.*, while the frequency tended to be lower for potatoes, sugars, and fruits. These results suggest that, during the summer, when diverse types of vegetable are available, the menus incorporated large amounts of vegetables, resulting in a greater intake of many elements during summer compared to the winter.

Inter-individual Variation in Intakes of Radionuclides and Stable Elements

The ^{137}Cs intake from daily diets varied from year to year in both Yokohama and Hiratsuka areas. Following this finding, we analyzed the intake of stable elements from daily diets in 1998 for each of the 20 subjects from the Hiratsuka and Yokohama areas. Tables 6 and 7 shows the amounts of elements and ^{137}Cs ingested during the summer and winter by each subject. For ^{137}Cs and 10 of the 11 elements in the Hiratsuka area and 7 elements in the Yokohama area, CV in the mean of 10 samples was higher than that shown in Tables 3, 4 and 5. The annual average daily ^{137}Cs intake per person in the Hiratsuka area (determined from combined samples from 5 subjects in 1998) was $0.090 \pm 0.028 \text{ Bq man}^{-1}\text{day}^{-1}$ (national average: $0.039 \pm 0.019 \text{ Bq man}^{-1}\text{day}^{-1}$, national maximum: $0.17 \text{ Bq man}^{-1}\text{day}^{-1}$, same year.⁵⁾ When individual samples were analyzed separately, the intake was ND to $0.29 \text{ Bq man}^{-1}\text{day}^{-1}$ and the mean of 5 samples was $0.15 \pm 0.085 \text{ Bq man}^{-1}\text{day}^{-1}$. Thus, the mean of individual samples was not identical to the mean of combined samples, and this discrepancy is attributable to the following factors: the weight of food ingested and the incineration rate varied among individual samples; and total radioactivity was low in individual samples because of the

small weight of the incinerated material, leading to a high percentage of samples rated as ND. In the analysis of ^{137}Cs intake by individuals, the intake by Subject No. 4 was particularly high, and the intake by the same subject during winter was high to a lesser extent. The high ^{137}Cs intake of this subject is attributable to the higher weight of food ingested (about 30% larger on average) than the other subjects. The intake of 8 elements (^{137}Cs , Ca, Mn, Fe, K, *etc.*) by this subject was maximal during winter. The exact cause of this finding remains unknown because ^{137}Cs was not measured for each dietary ingredient. In the analysis of 10 individual samples in the Yokohama area, the maximum ^{137}Cs uptake was $0.25 \text{ Bq man}^{-1}\text{day}^{-1}$ (recorded in Subject No. 3 during winter). This level exceeded the national maximum for the same year, as is the case with the maximum ^{137}Cs intake in the Hiratsuka area. Regarding this, it is desirable to conduct a quantitative analysis of individual ingredients in addition to analysis of the type and frequency of ingredients used in diets. We found that the choice of ingredients and menus by individual subjects, the volume of food ingested by individual subjects, the lifestyle of individual subjects, and so on, serve as factors causing variations in the intake of elements, *etc.*, resulting in annual changes in the ^{137}Cs intake. If combined samples from multiple subjects (averaged samples) are further investigated from now on, it will be possible to continue the collection of monitoring data which can represent a given district and facilitate the detection of abnormalities, *etc.*

Stable Cesium and ^{137}Cs

The relationship between Cs and ^{137}Cs in daily dietary samples was analyzed during the period from 1990 to 1994. The results are shown in Fig. 3. ^{137}Cs was not detectable in only 1 of the 20 samples. During this survey period, ^{137}Cs levels in foods remained high under the influence of the Chernobyl accident. The coefficient of determination for the correlation was as low as 0.226 for the 19 daily dietary samples, indicating the absence of a correlation. It was thus shown that equilibrium between Cs and ^{137}Cs in many ingredients of the diet had not been reached by the first half of the 1990 s. In the analysis of individual samples during the summer of 1998 in the Hiratsuka area, a positive correlation was noted between the intakes of Cs and ^{137}Cs ($r = 0.740$). This suggests that, more than 10 years after the Chernobyl accident, equilibrium between Cs and ^{137}Cs had been achieved in some

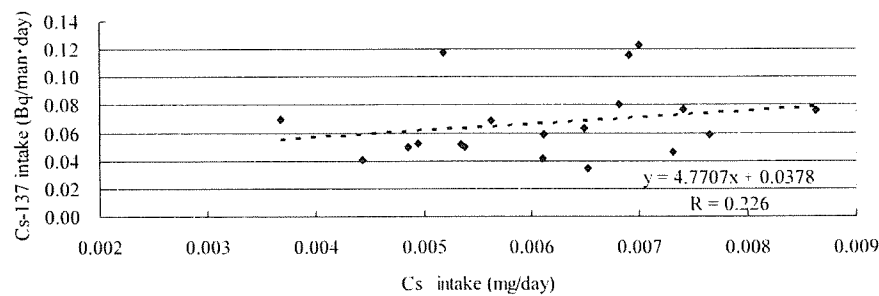


Fig. 3. Relation between Cs and ^{137}Cs Intakes in Daily Diet in Kanagawa, Japan (1990–1994)

ingredients, but that the extensive diffusion of ^{137}Cs in the environment reduced the level of ^{137}Cs in dietary ingredients, making the correlation with Cs less clear. In view of the finding that a positive correlation was noted between the intakes of Cs and ^{137}Cs from daily diets composed of diverse ingredients during some limited periods or in some limited districts, we may say that, in many ingredients of the diet, there is no equilibrium between Cs and ^{137}Cs but that the kinetics of ^{137}Cs are similar to those of Cs in some ingredients in which significant levels of ^{137}Cs are detected.

DISCUSSION

Radionuclide levels in daily diets of inhabitants of Kanagawa Prefecture were investigated for the period from 1984 to 2006. The level of ^{137}Cs in daily diets rose in 1986, the year of the Chernobyl accident, and it tended to decrease gradually thereafter. In recent years, however, the decrease in the daily dietary ^{137}Cs level has slowed down or ceased due to factors such as the dispersion of yellow sand containing ^{137}Cs from China. In the analysis of the daily dietary samples for the 5-year period from 1990 to 1994, the mean ^{137}Cs intake in the Hiratsuka area ($0.073 \pm 0.029 \text{ Bq man}^{-1} \text{ day}^{-1}$) was greater than that in the Yokohama area ($0.055 \pm 0.024 \text{ Bq man}^{-1} \text{ day}^{-1}$). For this 5-year period, we quantified stable elements contained in daily dietary samples and analyzed the amounts of inorganic elements ingested by females in their 40–60s living in Kanagawa Prefecture. The mean intake was greater in the Hiratsuka area for 7 of the 8 nutrients (Ca, Cr, Mn, Fe, Zn, Na, Cl, and K), excluding Cr. It was found that the menus in the Hiratsuka area used more diverse ingredients (including vegetables) and the daily amount of food

ingested was greater in the Hiratsuka area, possibly leading to a greater intake of ^{137}Cs and stable elements in this area. When compared to the Dietary Reference Intakes for Japanese (2005), the amount of nutrients ingested was approximately equal to or slightly smaller than the amount specified in the reference. The ingestion of sodium chloride was suppressed to levels below the DG in both areas. No element exceeded the upper limit of the amount to be ingested shown in the reference. Following the finding of the variation in dietary ^{137}Cs intake, we analyzed the intake of stable elements in individual subjects, and found that the amount of many elements ingested varied more greatly among individual than among combined samples. We found that the choice of ingredients and menus by individual subjects, the volume of diet ingested by individual subjects, the lifestyle of individual subjects, and so on, serve as factors leading to a variation in the intake of elements, *etc.*, resulting in annual changes in ^{137}Cs intake. No correlation was found between the dietary intake of Cs and that of ^{137}Cs , suggesting that an equilibrium between Cs and ^{137}Cs has not been reached in many food ingredients or that a decrease in the environmental ^{137}Cs level made it difficult to detect ^{137}Cs in dietary ingredients and to assess the relationship between ingredients and ^{137}Cs .

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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 \pm σ : 0.66 \pm 0.53) and 68.5-94.2 (81.5 \pm 8.5) Bq/d and the committed effective dose was 0.15-0.81 (0.29 \pm 0.24) and 0.16-0.21 (0.18 \pm 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 1.66×10^{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 (1.66×10^{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 concern-

ing 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