tions among the 13 fractions (Cs or K weight/fraction weight: $\mu g\ g^{-1}$), 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 $46.0E^{-6} \pm 7.5E^{-6}\ \mu g\ g^{-1}$. 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.

LITERATURE CITED

(1) Mascanzoni, D. Chernobyl's challenge to the environment: A report from Sweden. *Sci. Total Environ.* **1987**, *67*, 133–148.

(2) Elstner, E. F.; Fink, R.; Höll, W.; Lengfelder, E.; Ziegler, H. Radioactivity in mushrooms, mosses and soil samples of defined biotops in SW Bavaria. *Oecologia* **1989**, *80*, 173–177.

(3) Borio, R.; Chiocchini, S.; Cicioni, R.; Degli Esposti, P.; Rongoni, A.; Sabatini, P.; Scampoli, P.; Antonini, A.; Salvadori, P. Uptake of radiocesium by mushrooms. *Sci. Total Environ.* **1991**, *106*, 183–190.

(4) Byrne, A. R. Radioactivity in fungi in Slovenia, Yugoslavia, following the Chernobyl accident. *J. Environ. Radioact.* **1998**, *6*, 177–183.

(5) Sugiyama, H.; Shibata, H.; Isomura, K.; Iwashima, K. Concentration of radiocesium in mushrooms and substrates in the sub-alpine forest of Mt. Fuji, Japan. *J. Food Hyg. Soc. Jpn.* **1994**, *35*, 13–22.

(6) Yoshida, S.; Muramatsu, Y. Accumulation of radiocesium in basidiomycetes collected from Japanese forests. *Sci. Total Environ.* **1994**, *157*, 197–205.

(7) Yoshida, S.; Muramatsu, Y. Concentrations of alkali and alkaline earth elements in mushrooms and plants collected in a Japanese pine forest, and their relationship with ^{137}Cs. *J. Environ. Radioact.* **1998**, *41*, 183–205.

(8) Tsukada, H.; Shibata, H.; Sugiyama, H. Transfer of radiocaesium and stable caesium from substrata to mushrooms in a pine forest in Rokkasho-mura, Aomori, Japan. *J. Environ. Radioact.* **1998**, *39*, 149–160.

(9) Sugiyama, H.; Terada, H.; Shibata, H.; Morita, Y.; Kato, F. Radiocesium concentrations in wild mushrooms and characteristics of cesium accumulation by the edible mushroom (*Pleurotus ostreatus*). *J. Health Sci.* **2000**, *46*, 370–375.

(10) Dvoøák, P.; Kunová, V.; Beòová, K.; Ohera, M. Radiocesium in mushrooms from selected locations in the Czech Republic and the Slovak Republic. *Radiat. Environ. Biophys.* **2006**, *45*, 145–151.

(11) Karadeniz, O.; Yaprak, G. Dynamic equilibrium of radiocesium with stable cesium within the soil-mushroom system in Turkish pine forest. *Environ. Pollut.* **2007**, *148*, 316–324.

(12) Baeza, A.; Guillén, J.; Paniagua, J. M.; Hernández, S.; Martín, J. L.; Diez, J.; Manjón, J. L.; Moreno, G. Radiocaesium and radiostrontium uptake by fruit bodies of *Pleurotus eryngii* via mycelium, soil and aerial absorption. *Appl. Radiat. Isot.* **2000**, *53*, 455–462.

(13) Rudawska, M.; Leski, T. Macro and microelement contents in fruiting bodies of wild mushrooms from the Notecka forest in west-central Poland. *Food Chem.* **2005**, *92*, 499–506.

(14) Dupré, de Boulois, H.; Voets, L.; Delvaux, B.; Jakobsen, I.; Declerck, S. Transport of radiocaesium by arbuscular mycorrhizal fungi to *Medicago truncatula* under in vitro conditions. *Environ. Microbiol* **2006**, *8*, 1926–1934.

(15) Gaso, M. I.; Segovia, N.; Morton, O.; Lopez, J. L.; Machuca, A.; Hernandez, E. Radioactive and stable metal bioaccumulation, crystalline compound and siderophore detection in *Clavariadelphus truncates*. *J. Environ. Radioact.* **2007**, *97*, 57–69.

(16) Sugiyama, H.; Terada, H.; Isomura, K.; Tsukada, H.; Shibata, H. Radiocesium uptake mechanisms in wild and culture mushrooms. *Radioisotopes.* **1993**, *42*, 683–690 (in Japanese with English abstract).

(17) Terada, H.; Shibata, H.; Kato, F.; Sugiyama, H. Influence of alkali elements on the accumulation of radiocesium by mushrooms. *J. Radioanal. Nucl. Chem.* **1998**, *235*, 195–200.

(18) Kuwahara, C.; Fukumoto, A.; Ohsone, A.; Furuya, N.; Shibata, H.; Sugiyama, H.; Kato, F. Accumulation of radiocesium in wild mushrooms collected from a Japanese forest and cesium uptake by microorganisms isolated from the mushroom-growing soils. *Sci. Total Environ.* **2005**, *345*, 165–173.

(19) Vaughn, L. E.; Davis, R. H. Purification of vacuoles from *Neurospora crassa*. *Mol. Cell. Biol.* **1981**, *1*, 797–806.

(20) Cramer, C. L.; Ristow, J. L.; Paulus, T. J.; Davis, R. H. Methods for mycelial breakage and isolation of mitochondria and vacuoles of *Neurospora*. *Anal. Biochem.* **1983**, *128*, 384–392.

(21) Tijssen, J. P. F.; Beekes, H. W.; Steveninck, J. V. Localization of polyphosphates in *Saccharomyces fragilis*, as revealed by 4′,6-diamidino-2-phenylindole fluorescence. *Biochim. Biophys. Acta* **1982**, *721*, 394–398.

(22) Sugiyama, H.; Kato, F.; Sanekata, T.; Terada, H.; Morita, Y.; Wada, M. Growth and ^{137}Cs accumulation by mushroom (*Pleurotus ostreatus*) mycelia and elementary analysis with scanning electron microscope-X-ray microanalyzer. *Bioimages* **1998**, *6*, 109–113.

(23) Zhdanova, N. N.; Redchits, T. I.; Zheltonozhsky, V. A.; Sadovnikov, L. V.; Gerzabek, M. H.; Olsson, S.; Strebl, F.; Muck, K. Accumulation of radionuclides from radioactive substrata by some micromycetes. *J. Environ. Radioact.* **2003**, *67*, 119–130.

(24) Kato, F.; Kuwahara, C.; Oosone, A.; Ichikawa, T.; Terada, H.; Morita, Y.; Sugiyama, H. Accumulation and subcellular localization of cesium in mycelia of *Streptomyces lividans* and a Cs tolerant strains, *Streptomyces* sp TOHO-2. *J. Health Sci.* **2000**, *46*, 259–262.

(25) Dupré, de Boulois, H.; Joner, E. J.; Leyval, C.; Jakobsen, I.; Chen, B. D.; Roos, P.; Thiry, Y.; Rufylkiri, G.; Delvaux, B.; Declerck, S. Role and influence of mycorrhizal fungi on radiocesium accumulation by plants. *J. Environ. Radioact* **2008**, *99*, 785–800.

(26) Gyuricza, V.; Dupré de Boulois, H. Declerck, S. Effect of potassium and phosphorus on the transport of radiocesium by arbuscular mycorrhizal fungi. *J. Environ. Radioact.* [Online early access]. Published Online: May 15, 2008.

(27) Klionsky, D. J.; Herman, P. K.; Emr, S. D. The fungal vacuole: composition, function and biogenesis. *Microbiol. Rev.* **1990**, *54*, 266–292.

(28) Kuwahara, C.; Watanuki, T.; Matsushita, K.; Nishina, M.; Sugiyama, H. Studies on uptake of cesium by mycelia of the mushroom (*Pleurotus ostreatus*) by ^{133}Cs-NMR. *J. Radioanal. Nucl. Chem* **1998**, *235*, 191–194.

(29) Allan, R. A.; Miller, J. J. Influence of S-adenosylmethionine on DAPI-induced fluorescence of polyphosphate in the yeast vacuole. *Can. J. Microbiol.* **1980**, *26*, 912–920.

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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 $mSvy^{-1}$, 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 cecium-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 Hiratuka 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.

RESUITS

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: Bq man^{-1}day^{-1})

Year	Analytical organization	Collection time (s)	^{137}Cs NET ± SE	^{40}K NET ± SE
1993	This work	324900	0.047 ± 0.013	73.2 ± 0.5
	JCAC[a)]	71967	0.046 ± 0.012	71.5 ± 0.7
1994	This work	244200	0.046 ± 0.014	72.4 ± 2.2
	JCAC[a)]	71665	ND	75.3 ± 0.8
1995	This work	150000	0.051 ± 0.010	63.3 ± 0.4
	JCAC[a)]	70057	ND	63.7 ± 0.7
1996	This work	150000	0.075 ± 0.0090	73.3 ± 0.5
	JCAC[a)]	71678	0.076 ± 0.015	73.2 ± 0.7
1997	This work	100000	0.063 ± 0.012	68.9 ± 0.6
	JCAC[a)]	72146	ND	82.7 ± 0.8
1998	This work	100000	0.11 ± 0.013	82 ± 0.6
	JCAC[a)]	71289	0.061 ± 0.014	73.2 ± 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 (%)	n		Analytical value	S.D.	CV (%)	n	
Ca	20500	± 1650	8.05	6	1.91	1600	± 150	9.38	4	8.05
	20900	± 300				1740	± 70			
Cl	784	± 51	6.51	6		8690	± 402	4.63	4	0.11
	(690)					8700	± 400			
Cr	2.3	± 0.24	10.43	6	11.54	ND				
	2.6	± 0.30				—				
Cs	0.045	± 0.019	42.22	5		0.027	± 0.012	44.44	4	
	(0.04)					(0.014)				
Fe	287	± 43	14.98	6	4.33	41	± 7.8	19.02	4	25.77
	300	± 20				32.6	± 3.6			
K	13900	± 726	5.22	6	5.44	4130	± 2050	49.64	4	31.85
	14700	± 300				6060	± 280			
Mn	84	± 3.1	3.69	6	7.69	ND				—
	91	± 4				5.2	± 0.4			
Na	103	± 27	26.21	6	25.61	5820	± 361	6.20	4	6.88
	82	± 6				6250	± 260			
Rb	11	± 1.6	14.55	6	8.33	4.6	± 0.78	16.96	4	
	12	± 1				(4.8)				
Sc	0.060	± 0.0049	8.17	6		ND				
	[0.047 – 0.066]									
Zn	22	± 2.1	9.55	6	12.00	30.7	± 1.2	3.91	4	0.32
	25	± 3				30.8	± 1.1			

Element	NIES No.1 Pepperbush				Deflection with certified value (%)
	Analytical value	S.D.	CV (%)	n	
Ca	13400	± 832	6.21	2	2.90
	13800	± 700			
Cl	155	± 1.2	0.77	2	
Cr	0.65	± 0.20	31.18	2	
	(1.3)				
Cs	1.2	± 0.028	2.33	2	
	(1.2)				
Fe	208	± 5.9	2.82	2	1.46
	205	± 17			
K	12100	± 341	2.82	2	19.87
	15100	± 700			
Mn	1810	± 11	0.60	2	10.84
	2030	± 170			
Na	91	± 12	13.62	2	14.15
	106	± 13			
Rb	73	± 0.78	1.08	2	2.67
	75	± 4			
Sc	ND				
Zn	322	± 32.7	10.17	2	5.29
	340	± 20			

Upper row: our result, Lower row: certified value, ( ): uncertified or reference value, [ ]: literature value[1], ND: not detectable.

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

		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, chlorine (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^{-1}day^{-1} (^{137}Cs)]

		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
Electrolytes	Na	2270	2370	2390	1900	2480	2280
	Cl	3270	3480	3580	2730	3570	3330
	K	915	1090	1290	1190	1170	1130
Other trace elements	Rb	1.68	1.61	1.62	1.70	1.60	1.64
	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	0.060
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
Electrolytes	Na	1900 — 3610		2530	459	18.14	10
	Cl	2730 — 4710		3510	525	14.97	10
	K	915 — 2370		1440	436	30.25	10
Other trace elements	Rb	1.24 — 2.38		1.62	0.309	19.02	10
	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	10

with literature values[1)] 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 ^{137}Cs and cesium-134 (^{134}Cs) and natural radionuclide potassium-40 (^{40}K), *etc.*[2–4)] Figure 1 shows the time-course of ^{137}Cs 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 ^{137}Cs 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 ^{137}Cs 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 1990 s, this phenomenon was seen in less regions and the level of radioactive substances detected became lower. After 2000, however, ^{137}Cs 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 ^{137}Cs level remains constant because ^{137}Cs continues to be supplied in the form of fallout, although in low levels. The daily dietary ^{137}Cs level recorded in Kanagawa Prefecture in recent years was close to the level of ^{137}Cs 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 ^{137}Cs intake was 0.080 ± 0.056 Bq man^{-1}day^{-1}, with a median of 0.065 Bq man^{-1}day^{-1}.

The man ^{137}Cs 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^{-1}day^{-1} (median: 0.059 Bq man^{-1}day^{-1}), similar to the national level for the same period (mean: 0.061 ± 0.040 Bq man^{-1}day^{-1}, median: 0.050 Bq man^{-1}day^{-1}).[6] The maximum level in Japan during this period was 0.34 Bq man^{-1}day^{-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.

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

^{134}Cs 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^{-1}day^{-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^{-1}day^{-1} (^{137}Cs)]

		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 (%)	*n*
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 kg ± 0.405 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 kg ± 0.345 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 kg man^{-1}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^{-1}day^{-1} (^{137}Cs)]

		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 ^{137}Cs. The MHLW established the following 5 indices for nutrient intake: esti-

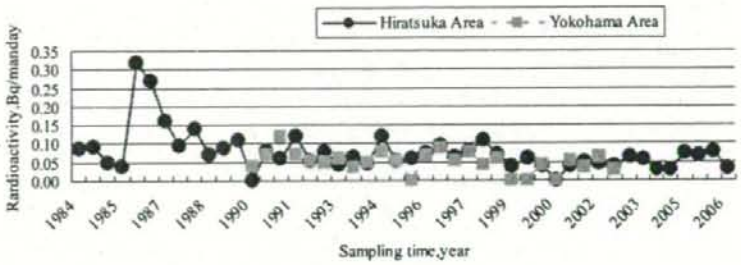

Fig. 1. ^{137}Cs Intake in Daily Diet in Kanagawa Prefecture

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) (2.2–8.6 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, *etc.*
2	Potatoes and powder	Sweet potato, potato, paste made from arum root, starch, Harusame, *etc.*
3	Sugar	Sugar, honey, maple syrup, *etc.*
4	Beans	Kidney bean, soybean, bean curd, natto, soymilk, and Yuba, *etc.*
5	Seeds	Almond, ginkgo nut, chestnut, sesame, peanut, peanut butter, *etc.*
6	Vegetables	Cucumber, radish, onion, eggplant, carrot, green pepper, spinach, *etc.*
7	Fruits	Plum, pickled ume, mandarin orange, persimmon, apple, pear, peach, *etc.*
8	Mushrooms	Enokitake, siitake, shimeji, hiratake, matsutake, kikurage, *etc.*
9	Algae	Green laver, laver, seaweed, hijiki, wakame, *etc.*
10	Seafood	Fish, shellfish, shrimp, crab, squid, octopus, fish paste products, *etc.*
11	Meat and poultry	Beef, pork, ham, bacon, sausage, chicken, frog, bees, *etc.*
12	Eggs	Quail egg, egg, egg-tofu, grilled egg, *etc.*
13	Milk	Fresh milk, process milk, powder milk, vegetable fat, cheese, ice cream, *etc.*
14	Fats and oils	Sesame oil, soybean oil, mixture oil, suet, butter, margarine, *etc.*
15	Cakes	Japanese-style confection, Danish pastries, cakes, dessert cakes, chocolates, *etc.*
16	Beverages	Alcohol, tea, coffee, cocoa, seaweed tea, carbonated drinks, *etc.*
17	Seasonings and spices	Salt, vinegar, dashi, mayonnaise, soy sauce, miso, peppers, *etc.*
18	Processed foods	Pre-packaged curry, frozen gyoza, frozen hamburg steak, frozen pilau, *etc.*

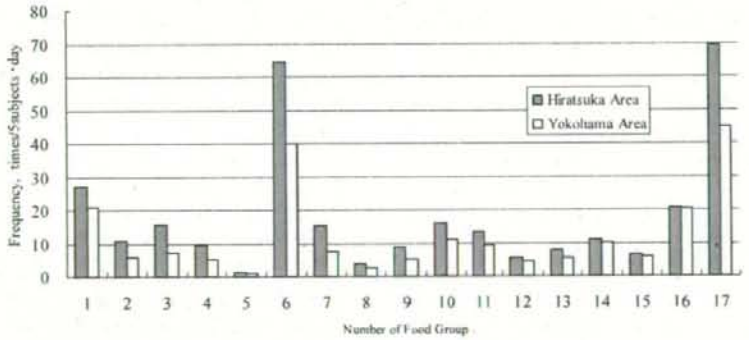

Fig. 2. Average Use Frequency of Ingredients in a Sample (for 5 persons)
The number of X-axis corresponds to Table 4. No. 18 (Processed foods) was excluded because its assessment in the menus was difficult.

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 Sugiyama[9] 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 ^{137}Cs intake was high. Among others, mushrooms are known to contain high levels of ^{137}Cs.[18–24] The contribution of group 6 to the intake of stable elements also seems to be high. During the survey period, ^{137}Cs 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 mgday^{-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 ± 0.028 Bq man^{-1}day^{-1} (national average: 0.039 ± 0.019 Bq man^{-1}day^{-1}, national maximum: 0.17 Bq man^{-1}day^{-1}, same year.[5]) When individual samples were analyzed separately, the intake was ND to 0.29 Bq man^{-1}day^{-1} and the mean of 5 samples was 0.15 ± 0.085 Bq man^{-1}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 Bq man^{-1}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

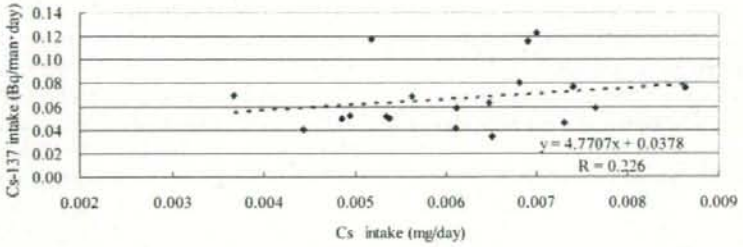

Fig. 3. Relation between Cs and ^{137}Cs Intakes in Daily Diet in Kanagawa, Japan (1990–1994)

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 ± 0.029 Bq man^{-1}day^{-1}) was greater than that in the Yokohama area (0.055 ± 0.024 Bq man^{-1}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.

REFERENCES

1) Hamaguchi, H., Numata, Y., Iwata, S., Koyama, M., Sasajima, K., Katayama, Y., Takeuchi, T., Shinogi, M., Mamuro, T., Kusaka, Y., Tsuji, H., Tamari, Y., Sagawa, T., Ohmori, S., Nagatsuka, S., Tanizaki, Y., Suzuki, T., Tomura, K., Hashimoto, Y., Bando, S. and Imahashi, T. (1977) Precision of neutron activation analysis for environmental biological materials.

BUNSEKI KAGAKU, **26**, T24–T28 (in Japanese).
2) Iijima, I., Takagi, H., Kuwahara, C. and Koyama, K. (1996) Concentration of caesium-134 and caesium-137 delivered from the Chernobyl accident to food in Kanagawa prefecture. *Bulletin of Kanagawa Prefectural Institute of Public Health*, **26**, 48–50 (in Japanese).
3) Iijima, I., Takagi, H. and Tomura, K. (2006) Concentration of artificial radionuclides and trace elements in daily intakes. *Bulletin of Kanagawa Prefectural Institute of Public Health* **36**, 41–44 (in Japanese).
4) Iijima, I., Kuwahara, C., Nakaguchi, M. and Katsube, K. (2007) Radioactivity Survey in Kanagawa prefecture. *The 49th proceeding of environmental radioactivity survey.*, Ministry of Education, Culture, Sports, Science and Technology-Japan Dec.4, Tokyo, Japan, 157–160 (in Japanese).
5) Iijima, I., Takagi, H. and Koyama, K. (1991) Transition of environmental radiation and radioactivity levels in Kanagawa prefecture. 1. Concentrations of some radionuclides in foods. *Bulletin of Kanagawa Prefectural Institute of Public Health*, **21**, 7–12 (in Japanese).
6) Japan Chemical Analysis Center (2008) *Radiation database*, http://search.kankyo-hoshano.go.jp/top.jsp
7) Igarashi, Y., Aoyama, M., Hirose, K., Miyao, T., Nemoto, K., Tomita, M. and Fujikawa, T. (2003) Resuspension: Decadal monitoring time series of the anthropogenic radioactivity deposition in Japan. *J. Radiat. Res.* (Tokyo), **4**, 319–328.
8) Aoyama, M., Hirose, K. and Igarashi, Y. (2006) Re-construction and updating our understanding on the global weapons tests ^{137}Cs fallout. *J. Environ. Monit.*, **8**, 431–438.
9) Sugiyama, H., Terada, H., Takahashi, M., Iijima, I. and Isomura, K. (2007) Contents and daily intakes of gamma-ray emitting nuclides, ^{90}Sr, and ^{238}U using market-basket studies in Japan. *J. Health Sci.*, **53**, 107–118.
10) Koyama, K., Takagi, H., Sugiyama, H., Iijima, I., Moriya, K. and Sakakibara, T. (1987) *Report of radioactivity survey in Kanagawa prefecture—1986—*, p.27 (in Japanese).
11) Cho, S. Y., Kang, S. H., Hong, W. J., Kim, H. S., Lee, J. Y. and Chung, Y. S. (2006) Duplicate diet study of Korean geriatric population by neutron activation analysis. *Japan Association of Activation Analysis; Activation Analysis*, **19**, 16–23.
12) Okada, R., Tanabe, N., Wakai, S., Kabasawa, R. and Suzuki, H. (2004) Zink intake from daily meals in preschool children and its influence on their physical growth. *Nippon kōshū Eisei Zasshi* (*Japanese Journal of Public Health*), **51**, 753–763 (in Japanese).
13) World Health Organization (WHO) (1996), *Trace elements in human nutrition and Health*, WHO press, Geneva, pp.1–2.
14) Yamagata, N. (1962) The concentration of common cesium and rubidium in human body. *J. Radiat. Res.* (Tokyo), **3–1**, 9–30.
15) Lloyd, R. D., Mays, C. W., McFarland, S. S., Zudel, W. S. and Tyler, F. H. (1973) Metabolism of ^{83}Rb and ^{137}Cs in persons with muscle disease. *Radiat. Res.*, **54**, 463–478.
16) Yoshida, S. and Muramatsu, Y., (1996) Determination of trace elements in mushrooms, plants and soils collected from Japanese forests. *Int J. Environ. Anal Chem.*, **63**, 195–206.
17) Imai, N. (2005) Geochemical map of Japan for environmental assessment. *Environment Res. Quarterly*, **139**, 25–33 (in Japanese).
18) Tsukada, H., Shibata, H. and Sugiyama, H. (1998) Transfer of radiocaesium and stable caesium from substrata to mushrooms in a pine forest in Rokkasho-mura, Aomori, Japan — The transfer of radionuclides through Nordic ecosystems to man. *J. Environ. Radioact.*, **39**, 149–160.
19) Kuwahara, C., Watanuki, T., Matsushita, K., Nishina, M. and Sugiyama, H. (1998) Studies on uptake of cesium by mycelium of the mushroom (*Pleurotus ostreatus*) by ^{133}Cs-NMR. *J. Radioanal. Nuc. Chem.*, **235**, 191–194.
20) Sugiyama, H., Kato, F., Sanekata, T., Terada, H., Morita, Y. and Wada, M. (1998) Growth and ^{137}Cs accumulation by mushroom (*Pleurtus ostreatus*) mycelia and elementary analysis with scanning electron microscope-X-ray microanalyzer. *Bioimages*, **6**, 109–113.
21) Giovani, C., Garavaglia, M. and Scruzzi, E. (2004) Radiocaesium in mushrooms from northeast Italy, 1986–2002. *Radiat. Prot. Dosimetry*, **111**, 377–383.
22) Koyama, K., Takagi, H., Iijima, I., Kuwahara, C. and Matsuzaki, M. (1991) *Report of radioactivity survey in Kanagawa prefecture—1990—*, p.20 (in Japanese).
23) Muramatsu, Y., Yoshida, S. and Sumiya, M. (1991) Concentrations of radiocesium and potassium in basidiomycetes collected in Japan. *Sci. Total Environ.*, **105**, 29–39.
24) Sugiyama, H., Shibata, H., Isomura, K. and Iwashima, K. (1994) Concentration of radiocesium in mushrooms and substrates in the sub-alpine forest of Mt. Fuji Japan. *J. Food Hyg. Soc. Japan*, **35**, 13–22.

平成19年度　分　担　研　究　報　告（訂正版）

日常食の汚染物摂取量及び汚染物モニタリング調査研究

渡邉　敬浩

平成19年度厚生労働科学研究補助金　食品の安心・安全確保推進研究事業

食品中の有害物質等の摂取量の調査及び評価に関する研究

研究分担報告書

日常食の汚染物質摂取量及び汚染物モニタリング調査研究

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研究要旨

国内に流通している食品に含まれる汚染物質の濃度，及び食事を介した当該汚染物質の摂取量を明らかにすることを目的として，全国の衛生研究所の協力のもと，汚染物モニタリング調査と，マーケットバスケット方式によるトータルダイエット試料の分析を通じた汚染物質摂取量調査を実施した．

汚染物モニタリングにおいては，全国48カ所で得られた食品中汚染物検査データ50万件を収集し，食品での汚染物の検出率，複数の汚染物による汚染状況を調査した．汚染物質摂取量調査では，全国10カ所の衛生研究所等において各地域の摂取量に応じたトータルダイエット試料をマーケットバスケット方式により調製し，継続的にモニターしている汚染物質の濃度を測定することを通じて，1日当たりの当該汚染物質摂取量を推定した．

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A．　研究目的

近年，輸入食品から鉛あるいは農薬が検出されるなど，種々の化学物質による食品の汚染，さらにヒトに対する曝露や，それに伴う健康影響に関する不安が国民の間に広がり，社会的関心が高まっている．これら化学物質のヒトへの曝露はその 90 %以上が食事を介していると考えられており，食品に含まれる有害化学物質の量とその分布状態を明らかにして食品の安全性を確保することは，食品衛生における基本課題であるが，正確な実施は非常に困難である．食品を１つの大きな集合と捉えたとき，化学物質はその中で均一に分布しておらず，特定の食品に偏って存在することが多い．従って，食品の汚染状態を正確に把握するためには，多数の食品に含まれる汚染物質の濃度データを全国的に継続して収集し，解析しなくてはならない．また，食品中の汚染物質のヒトへの曝露状態を把握するためには，単に個々の食品の濃度のみならず，日本人がその食品をどのくらい食べているか，つまり摂取量も考慮しなければならない．さらに，食品を調理加工した場合の汚染物質濃度の変化も考慮しなければならず，食品を日常的に摂取される状態とした上で分析し，得られた分析値に基づいた摂取量の推定が必要である．本研究では，前者の目的のために，汚染物モニタリング調査研究，後者の目的のためにマーケットバスケット方式による汚染物質摂取量調査研究を行った．

B．研究方法

1）汚染物質摂取量調査

日常食からの汚染物質摂取量を推定するため，日常食のモデルとしてマーケットバスケット方式によるトータルダイエット試料を調製した．調製は，地域による食品摂取パターンの違いについても考慮することを目的に，全国 10 カ所の衛生研究所及び大学で行った．各地域における個々の食品の摂取量は，平成 14 年度～16 年度に行われた国民健康・栄養調査の結果を地域別に集計した結果の平均値とした．各地の小売店から食品を購入し，茹でる，焼く等の一般的な調理加工を行ってから，一日当たりの摂取量に従って秤量し，混合・均一化して試料とした．試料中の重金属，農薬等の

汚染物質濃度を測定し，その結果得られた汚染物質の濃度と食品の摂取量から，1日の食事からの汚染物質摂取量を推定した．

2）汚染物モニタリング調査

全国 48 カ所の地方衛生研究所等から食品中の汚染物検査データ 50 万件を収集した．あらかじめ入力用のフォームを規定した上で配布し，これに各機関が入力後返送する形式でデータを収集した．入力用フォームには，誤入力をチェックするプログラム(Microsoft Excel VBA)を含めておき，機関ごとに入力者があらかじめ誤入力をチェックした後に送付するよう指示することにより，無効なデータが入らないようにした．

国立医薬品食品衛生研究所食品部に送付されたデータは再度エラーチェックを行い集計した後，食品部サーバー上に構築したデータベースに追加した．

C．研究結果

1）汚染物質摂取量調査

Table 1 に全協力機関から報告された，ヘキサクロロシクロヘキサン(HCH)類，DDT類，ディルドリン，ヘプタクロルエポキサイド，HCB，PCB，有機リン系農薬類（マラチオン，MEP，ダイアジノン），金属類（鉛，カドミウム，ヒ素，水銀，銅，マンガン，亜鉛）の総摂取量について．2003～2007年の年次推移を示す．代表値として，10機関の平均値(mean)と中央値(median)を示した．平均値については，汚染物質濃度が定量下限以下(ND)の場合に濃度を0として計算した結果(ND=0)と，定量下限の1/2の濃度として計算した結果(ND=1/2LQ)の2種類を示した．

HCH類の中で摂取量の多いα及びβ-HCH摂取量の平均値は2002から2004年にかけて減少傾向を示したが，2005年にはβ-HCHが増加し，2006年にはα，β-HCH共に増加した．2007年には，ND=0として計算した平均値は2005年以前のレベルに戻ったが，ND=1/2LQとして計算した平均値は非常に大きくなった．検出頻度としては，α-HCHが3機関で，β-HCHが2機関で検出された．一方，γ-HCHの摂取量は2004年に高値を示したが，2005から2007年には例年通りの値となった．δ-HCHを検出した機関はなかった．本年度の調査結果において，総HCH摂取量は2003及び2005年と同程度となった．

2003から2007年の間の総DDT摂取量はほぼ一定であった．異性体中では，p,p'-DDEの摂取量が最も高く，また，最も高い頻度で検出された．(p,p'-DDT，p,p'-DDD，o,p'-DDTはそれぞれ，10機関中7機関，6機関，4機関で検出された。これらに対しp,p'-DDEは10機関中8機関で検出された．)

ディルドリン摂取量は2004から2006年に比べ減少した．ヘプタクロルエポキサイドは2005年及び2006年の摂取量が高い値であったが，本年は例年通りの値となった．HCB摂取量は2004年にはやや高かったが，本年度は5年間で最も低い値であった．PCB摂取量は2005年に1 μg/man/day に近い値を示したが，本年度は2006年に続いて例年通りの摂取量となった．

有機リン系農薬のマラチオンとダイアジノンはそれぞれ1機関で検出され，MEPは全ての機関で検出されなかった．例年，有機リン系農薬の検出頻度は低く，摂取量の中央値は5年間を通じて0 μg/man/dayである．