

# 全国の行政検査結果(1998年)を基にした食品中の無機食品添加物(亜硝酸塩,硝酸塩,二酸化硫黄),酸化防止剤(BHA,BHT),品質保持剤(プロピレングリコール),および甘味料(サッカリンナトリウム)の濃度実態および摂取量の推定

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Report

## Estimation of Inorganic Food Additive (Nitrite, Nitrate and Sulfur Dioxide), Antioxidant (BHA and BHT), Processing Agent (Propylene Glycol) and Sweetener (Sodium Saccharin) Concentrations in Foods and Their Daily Intake Based on Official Inspection Results in Japan in Fiscal Year 1998\*

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The mean concentration and daily intake of inorganic food additives (nitrite, nitrate, and sulfur dioxide), antioxidants (BHA and BHT), a processing agent (propylene glycol), and a sweetener (sodium saccharin) were estimated based on the results of an analysis of 34,489 food samples obtained in official inspections by 106 local governments in Japan in fiscal year 1998. The ratios of mean concentrations of these seven food additives to each allowable limit were 20.0%, 53.9%, 15.5%, 6.2%, 0.4%, 18.5%, and 5.7%, respectively. The daily intakes of these food additives estimated from their concentrations in foods and the daily consumption of foods were 0.205, 0.532, 4.31, 0.119, 0.109, 77.5, and 7.27 mg per person, respectively. These amounts were 6.8%, 0.3%, 12.3%, 0.5%, 0.7%, 6.2%, and 2.6% of the acceptable daily intake (ADI), respectively, when body weight was assumed to be 50 kg. No remarkable differences in the daily intakes of these seven food additives or the ratios to the ADI were observed compared with the results based on the official inspections in fiscal years 1994 and 1996.

**Key words:** food additive; daily intake; nitrite; nitrate; sulfur dioxide; BHA; BHT; propylene glycol; sodium saccharin; official inspection

### Introduction

Estimates of the daily intake of 16 food additives based on the results of official inspections in Japan in fiscal years 1994 and 1996 have already been reported<sup>1), 2)</sup>. We repeated the same surveillance study on the same 16 food additives based on the results of official inspections in fiscal year 1998, and reported the estimates of the daily intake of five preservatives<sup>3)</sup> and four antifungal agents<sup>4)</sup> in fiscal year 1998. In the present study, we estimated the mean concentrations of three inorganic food additives (nitrite, nitrate, and sulfur dioxide), two antioxidants (butylated hydroxyanisole (BHA) and butylated hydroxytoluene (BHT)), a processing agent (propylene glycol) and a sweetener (sodium saccharin) in foods, and deduced the daily in-

takes of these seven food additives per person from the mean concentrations and amounts of food consumed.

### Methods

The methods used were the same as those described previously<sup>3)-6)</sup>, but this time the results of new official inspections performed in fiscal year 1998 and a food consumption list reported by the Food Sanitation Investigation Council in 2000<sup>7)</sup> were used.

#### 1. Methods of investigation and items studied

##### a) Questionnaire

A questionnaire concerning the results of official inspections for food additives from April 1, 1998 to March 31, 1999 (Japanese fiscal year 1998) was sent by the Division of Food Chemistry, Ministry of Health and Welfare of Japan (present name: Standards Division, Department of Food Sanitation, Ministry of Health, Labour and Welfare)<sup>8)</sup> to all 114 local governments that are obliged to establish public health centers by the Regional Public Health Law.

##### b) Items queried

The following food additives were investigated in

\* Concentration of Food Additives in Foods and the Daily Intake in Japan (Report No. 11). [Report No. 10 was entitled "Estimation of Concentrations of Antifungal Agents Allowed as Food Additives in Foods and Their Daily Intake Based on Official Inspection Results in Japan in Fiscal Year 1998", published in this Journal, 43, 49-56 (2002).]

this study: nitrite (sodium nitrite), nitrate (sodium nitrate and potassium nitrate), sulfur dioxide (sodium sulfite, sodium hydrosulfite, sulfur dioxide, potassium pyrosulfite, sodium pyrosulfite, potassium hydrogen sulfite solution, and sodium hydrogen sulfite solution), BHA, BHT, propylene glycol, and sodium saccharin (saccharin and sodium saccharin). The substances in parentheses above are permitted to be used as food additives in Japan. The concentration of sulfites was calculated as sulfur dioxide in accordance with Japan's Specifications and Standards for Food Additives<sup>9)</sup>. The names of both imported and domestic foods, the numbers of food samples inspected, the numbers of samples in which the above seven food additives were detected, the maximum, minimum, and mean concentrations in the samples in which they were detected, and the detection limit of the analysis for each food additive were queried.

### c) Sampling of foods and analytical methods for food additives

In accordance with the annual program of each local government, samples were chosen by food inspectors in markets or factories manufacturing food products. Basically, the following methods were used to determine food additives as required by the Guide for Food Sanitation Inspection<sup>10)</sup> by the Environmental Health Bureau of the Ministry of Health and Welfare (present name: Department of Food Sanitation, Ministry of Health, Labour and Welfare): colorimetry for nitrite, colorimetry after reduction by cadmium column for nitrate, the alkaline titration method or colorimetry after distillation for sulfur dioxide, HPLC for BHA and BHT, GC for propylene glycol, and HPLC for sodium saccharin.

### 2. Analyses of reports from local governments

Data sent back from the local governments were classified according to the categories of foods in Japan's Specifications and Standards for Food Additives<sup>9)</sup>. Foods in which these food additives are not allowed to be used were categorized according to the Guide for Categorization of Food<sup>11)</sup>. As described in the previous paper<sup>3)</sup>, when the concentrations of these food additives were below the detection limit, they were regarded as 0 mg/kg.

### 3. Calculations and estimation of daily intake

The daily intakes of these seven food additives per person were estimated based on both the mean concentrations of these additives in all foods (including non-permissible foods) obtained in the present study and the daily food consumption levels reviewed in 2000 by the Food Sanitation Investigation Council<sup>17)</sup>, Ministry of Health and Welfare. When the consumption level of a kind of food was unknown, 0.01 g was applied as the daily consumption per person, as described in the report by the Council.

## Results

### 1. Questionnaire returns and number of samples

Replies were obtained from 108 of the 114 local governments (94.7%, *i.e.*, 45 prefectures, 41 cities, and 22 wards), representing most of Japan. The seven food additives relevant to the present paper were inspected by 106 local governments (98.1% of the 108 local governments which replied). Inspections were performed for nitrite, nitrate, sulfur dioxide, BHA, BHT, propylene glycol, and sodium saccharin by 100, 8, 96, 71, 73, 83, and 92 local governments, respectively. The total number of food samples inspected for these food additives was 34,489, of which 4,791 (13.9% of total samples) were samples of imported food (Table 1). The results obtained in the present study were therefore considered to be highly reliable, since inspection results were received from most of the local governments countrywide, and large numbers of samples were tested, except for nitrate for which inspections of only 182 samples by 8 local governments were performed.

### 2. Concentrations of seven food additives in foods

The relevant seven food additives, nitrite, nitrate, sulfur dioxide, BHA, BHT, propylene glycol, and sodium saccharin, were detected in 8,219 (23.8%) out of a total of 34,489 food samples. Concentrations of each food additive in foods were shown in Tables 2 to 8.

#### a) Nitrite

Nitrite was detected in 3,790 (75.7%) of 5,004 samples tested (Table 2). The detection rate in the samples of allowable foods (total of seven categories of foods in Table 2) was 78.1%, and the mean concentration in tested samples was 20.0% of the allowable residual

**Table 1.** Number of Local Governments that Performed Inspections for Relevant Food Additives and the Number of Food Samples Inspected

Food additives	Number of local governments	Number of samples		
		Domestic	Imported	Total
Nitrite	100	4,741	263	5,004
Nitrate	8	182	16	198
Sulfur dioxide	96	5,536	1,925	7,461
Butylated hydroxyanisole	71	1,460	586	2,046
Butylated hydroxytoluene	73	1,481	592	2,073
Propylene glycol	83	2,446	20	2,466
Sodium saccharin	92	13,852	1,389	15,241
Total	106	29,698	4,791	34,489

Table 2. Concentration of Nitrite in Foods and Estimated Daily Intake

Foods categorized by the standards for use	Regulation (g/kg)	Number of samples		Detection rate (%)	Detected samples		Tested samples		Daily intake per person		Contribution to total intake (%)
		Tested	Detected		Mean conc. (g/kg)	Ratio to limit (%)	Mean conc. (g/kg)	Ratio to limit (%)	Food (g)	Nitrite (mg)	
Meat products	0.070	3,686	3,050	82.7	0.0139	19.9	0.0115	16.5	11.71	0.135	65.8
Whale meat bacon	0.070	9	8	88.9	0.0291	41.6	0.0259	37.0	0.01	0.000	0.1
Fish meat ham and sausage	0.050	105	45	42.9	0.0099	19.8	0.0042	8.5	0.33	0.001	0.7
Salted salmon ovary "sujiko"	0.0050	156	121	77.6	0.0053	107.0	0.0041	83.0	0.31	0.001	0.6
Salted salmon roe "ikura"	0.0050	228	96	42.1	0.0021	42.3	0.0009	17.8	0.31	0.000	0.1
Salted pollack roe "tarako"	0.0050	479	319	66.6	0.0022	43.5	0.0014	29.0	1.28	0.002	0.9
("Sujiko", "ikura", and "tarako")	0.0050	69	55	79.7	0.0017	34.4	0.0014	27.4	—	—	—
Nonpermissible foods*	—	272	96	35.3	0.0032	—	0.0011	—	—	—	—
Total (or average)	—	5,004	3,790	(75.7)	—	(25.6)	—	(20.0)	673.71	0.140	—
Breakdown of nonpermissible foods (detected only)	—	151	80	53.0	0.0025	—	0.0013	—	20.33	0.027	13.3
Processed marine products	—	60	7	11.7	0.0070	—	0.0008	—	14.50	0.012	5.7
Fish meat paste other than fish meat ham and sausage	—	17	5	29.4	0.0045	—	0.0013	—	19.70	0.026	12.8
Pre-cooked foods "sozai"	—	—	—	—	—	—	—	—	—	—	—
Daily intake of nitrite	—	—	—	—	—	—	—	—	—	0.205	100.0

\* Breakdown of nonpermissible foods in which nitrite was not detected: nonpermissible roe products (4 samples) and rice wine ("seishu") (4), etc.

Table 3. Concentration of Nitrate in Foods and Estimated Daily Intake

Foods categorized by the standards for use	Regulation (g/kg)	Number of samples		Detection rate (%)	Detected samples		Tested samples		Daily intake per person		Contribution to total intake (%)
		Tested	Detected		Mean conc. (g/kg)	Ratio to limit (%)	Mean conc. (g/kg)	Ratio to limit (%)	Food (g)	Nitrate (mg)	
Meat products	0.070	81	57	70.4	0.0658	94.0	0.0463	66.2	5.03	0.233	43.8
Whale meat bacon	0.070	0	0	—	—	—	—	—	0.03	—	—
Cheese	0.20	8	3	37.5	0.0080	4.0	0.0030	1.5	2.24	0.007	1.3
Rice wine ("seishu")	0.10	11	2	18.2	0.0085	8.5	0.0015	1.5	21.22	0.033	6.2
Nonpermissible foods*	—	98	54	55.1	0.0083	—	0.0046	—	645.19	—	—
Total (or average)	—	198	116	(58.6)	—	(86.9)	—	(53.9)	673.71	0.272	—
Breakdown of nonpermissible foods (detected only)	—	81	49	60.5	0.0089	—	0.0054	—	36.86	0.198	37.2
Processed marine products	—	4	2	50.0	0.0030	—	0.0015	—	34.54	0.052	9.7
Canned or bottled foods	—	5	1	20.0	0.0020	—	0.0004	—	19.70	0.008	1.5
Pre-cooked foods ("sozai")	—	8	2	25.0	0.0030	—	0.0008	—	1.90	0.001	0.3
Roe	—	—	—	—	—	—	—	—	—	—	—
Daily intake of nitrate	—	—	—	—	—	—	—	—	—	0.532	100.0

limits<sup>9)</sup>. The highest detection rate, 88.9%, was observed in whale meat bacon (8 out of 9 samples), and the mean concentration in tested whale meat bacon was 0.0259 g/kg, 37.0% of the allowable limit (0.070 g/kg). The concentration of nitrite in meat products (3,686 samples) was 0.0115 g/kg, corresponding to 16.5% of the allowable limit (0.070 g/kg), and was almost the same as those in fiscal years 1994<sup>12)</sup> and 1996<sup>5)</sup>. The highest concentration ratio to the allowable limit (83.0% of the limit, 0.0050 g/kg) was observed in salted salmon ovary "sujiko". Nitrite is known to be a natural component, so low nitrite levels in foods are considered to represent the natural component. Although 20 samples of confectionery were inspected and 0.020 g/kg of nitrite was found in fiscal year 1996<sup>5)</sup>, no confectionery was inspected in fiscal year 1998. No nitrite was detected in 4 samples of nonpermissible roe products, 4 samples of rice wine "sake", etc.

#### b) Nitrate

Nitrate was detected in 116 out of 198 samples (58.6%) (Table 3). The number of samples tested was 136 in fiscal year 1994<sup>12)</sup> and 209 in fiscal year 1996<sup>5)</sup>. The detection rate in all foods in which the use of nitrate is allowed was 62.0%, and the mean concentration in allowable samples was 53.9% of the allowable residual limits. The mean concentration in 81 samples of meat products was 0.0463 g/kg, corresponding to 66.2% of the limit (0.070 g/kg). Whale meat bacon was not inspected in fiscal year 1998. Nitrate was detected in 49 of 81 samples of processed marine products (60.5%), in which the use of nitrate is not allowed, and the mean concentration of tested samples was 0.0054 g/kg.

#### c) Sulfur dioxide

Sulfur dioxide was detected in 1,828 of 7,461 samples (24.5%) (Table 4). The detection rate of sulfur dioxide in all allowable foods was 25.4%, and the mean concentration in tested samples was 15.5% of the allowable residual limits. The highest detection rate (97.9%, 46 of 47 samples), except that of molasses (100%, 1 of 1 sample), was observed in kon-jak flower, "kon'nyakuko", at a mean concentration of 0.405 g/kg corresponding to 45.0% of the residual limit. Sulfur dioxide was also detected in a high percentage of dried gourd strips, "kampyo" (364 out of 380 samples, 95.8%), and wine (463 of 522 samples, 88.7%), and the mean concentration in tested samples was 33.5% of the limit in the former and 16.9% in the latter. Kon-jak flour is an ingredient of devil's tongue, "kon'nyaku", and dried gourd strips are eaten after rinsing with water and boiling with bouillon of soy sauce. Then, the concentration of sulfur dioxide in cooked foods would be lower than that of the ingredients. The detection rate in "other foods" was 9.6% (detected in 470 of 4,890 samples tested), and the mean concentration was 0.004 g/kg, corresponding to 15.0% of the limit (0.030 g/kg). No sulfur dioxide was detected in the following allowable foods; starch syrup, "mizuame", (6 samples), candied cherry (5), natural fruit juice (3), dried mashed potato

(2), and gelatin (2). Sesame (5 samples), beans (5), and hops (2), in which the use of sulfur dioxide is prohibited, were not found to contain sulfur dioxide.

#### d) BHA

BHA was detected in 188 (9.2%) of 2,046 samples tested (Table 5). The detection rate in the tested samples in which BHA is allowed to be used was 17.4%, and the mean concentration of BHA was 6.2% of the limits. BHA was detected only in dried marine products among allowable foods, and the detection rate was 20.8% (181 of 871 samples). The mean concentration in all tested dried marine products was 0.0147 g/kg, and the concentration was 7.3% of the allowable limit (0.20 g/kg). No BHA was detected in 132 samples of fats and oils including margarine, 15 samples of butter, and 21 samples of frozen or salted marine products. Frozen whale meat and dried mashed potato were not inspected in fiscal year 1998. BHA in confectionery and processed marine products, which are nonpermissible foods, may be carried over from their ingredients because of their low concentrations. BHA is not allowed to be used in chewing gum in Japan, but it was found in 3 samples of imported chewing gum. BHA was not detected in nonpermissible foods such as seasonings (87 samples), frozen food (74), pre-cooked foods "sozai" (57), meat products (40), and vegetable products (34).

#### e) BHT

The standard for the use of BHT is almost the same as that of BHA, except that BHT is allowed to be used in chewing gum<sup>9)</sup>. BHT was detected in 27 (1.3%) of 2,073 samples tested. The detection ratio in all tested allowable foods was 1.8%, and the mean concentration was 0.4% of the limits (Table 6). BHT was detected in fats and oils, dried marine products, and chewing gum; the detection rates in the tested samples of these allowable foods were 3.0%, 0.8%, and 61.5%, and mean concentrations were 0.0020, 0.0006, and 0.0525 g/kg, respectively. These concentrations were 1.0%, 0.3%, and 7.0% of the limits. BHT was tested for 10 samples of frozen marine products, 20 samples of butter, and 20 samples of salted marine products, but was not detected. No sample of frozen whale meat or dried mashed potato was inspected in 1998. The mean concentration of BHT in detected samples in confectionery, which is a nonpermissible food, was at almost the same level as that in fats and oils. Pre-cooked foods "sozai" contributed mainly (53.7%) to the total intake of BHT. No BHT was detected in nonpermissible foods such as marine products (153 samples), seasonings (88), frozen food (74), meat products (40), and vegetable products (33).

#### f) Propylene glycol

Propylene glycol is allowed in all processed foods. It was detected in 802 (32.5%) of 2,466 samples tested (Table 7). The mean concentration of propylene glycol was 18.5% of the allowable limit. The highest detection rate (38.6%) was observed in raw noodles (681 of 1,765 samples) except dried noodles (1 sample) and fats and oils for noodle kneading (1 sample), and the concentration was 21.8% of the allowable limit (20 g/kg). No

Table 4. Concentration of Sulfur Dioxide in Foods and Estimated Daily Intake

Foods categorized by the standards for use	Regulation (g/kg)	Number of samples		Detection rate (%)	Detected samples		Tested samples		Daily intake per person		Contribution to total intake (%)
		Tested	Detected		Mean conc. (g/kg)	Ratio to limit (%)	Mean conc. (g/kg)	Ratio to limit (%)	Food (g)	Sulfur dioxide (mg)	
Dried gourd strips "kampyo"	5.0	380	364	95.8	1.747	34.9	1.673	33.5	0.55	0.920	21.3
Dried fruits (except raisins)	2.0	404	161	39.9	0.542	27.1	0.216	10.8	0.31	0.067	1.6
Kon-jak flour "kon'nyaku-ko"	0.90	47	46	97.9	0.414	46.0	0.405	45.0	—	—	—
Dried mashed potato	0.50	2	0	0.0	0.000	0.0	0.000	0.0	0.48	0.000	0.0
Gelatin	0.50	2	0	0.0	0.000	0.0	0.000	0.0	0.01	0.000	0.0
Dijon mustard	0.50	2	1	50.0	0.067	13.4	0.034	6.7	0.01	0.000	0.0
Wine	0.35	522	463	88.7	0.067	19.1	0.059	16.9	1.30	0.077	1.8
Miscellaneous alcoholic beverages	0.35	8	2	25.0	0.040	11.4	0.010	2.9	0.01	0.000	0.0
Molasses "toumitsu"	0.30	1	1	100.0	0.267	89.0	0.267	89.0	0.01	0.003	0.1
Candied cherry	0.30	5	0	0.0	0.000	0.0	0.000	0.0	0.01	0.000	0.0
Tapioca starch	0.25	7	2	28.6	0.002	0.8	0.001	0.2	0.01	0.000	0.0
Starch syrup "mizuame"	0.20	6	0	0.0	0.000	0.0	0.000	0.0	0.01	0.000	0.0
Natural fruit juice	0.15	3	0	0.0	0.000	0.0	0.000	0.0	4.86	0.000	0.0
Sweetened beans "amanatto"	0.10	82	49	59.8	0.029	29.1	0.017	17.4	0.01	0.000	0.0
Boiled beans	0.10	362	84	23.2	0.026	26.0	0.006	6.0	2.66	0.016	0.4
Shelled prawns	0.10	415	165	39.8	0.034	34.4	0.014	13.7	3.03	0.041	1.0
Frozen un-boiled shelled crab	0.10	19	13	68.4	0.025	25.3	0.017	17.3	0.01	0.000	0.0
Other foods	0.030	4,890	470	9.6	0.047	155.6	0.004	15.0	660.43	2.964	68.7
Nonpermissible foods*	—	304	7	2.3	0.035	—	0.001	—	—	—	—
Total (or average)	—	7,461	1,828	(24.5)	—	(60.9)	—	(15.5)	673.71	4.089	—
Breakdown of nonpermissible foods (detected only)	—	293	7	2.4	0.035	—	0.001	—	269.69	0.226	5.2
Vegetables	—	—	—	—	—	—	—	—	—	—	—
Daily intake of sulfur dioxide	—	—	—	—	—	—	—	—	—	4.31	100.0

\* Breakdown of nonpermissible foods in which sulfur dioxide was not detected: sesame (5 samples), beans (5) and hops (2)

Table 5. Concentration of BHA in Foods and Estimated Daily Intake

Foods categorized by the standards for use	Regulation limit (g/kg)	Number of samples		Detection rate (%)	Detected samples		Tested samples		Daily intake per person		Contribution to total intake (%)
		Tested	Detected		Mean conc. (g/kg)	Ratio to limit (%)	Mean conc. (g/kg)	Ratio to limit (%)	Food (g)	BHA (mg)	
Frozen marine products	1.0	11	0	0.0	0.000	0.0	0.0000	0.0	1.80	0.000	0.0
Frozen whale meat	1.0	0	0	—	—	—	—	—	0.01	—	—
Fats and oils	0.20	132	0	0.0	0.000	0.0	0.0000	0.0	10.32	0.000	0.0
Butter	0.20	15	0	0.0	0.000	0.0	0.0000	0.0	1.04	0.000	0.0
Dried marine products	0.20	871	181	20.8	0.071	35.3	0.0147	7.3	7.47	0.110	91.8
Salted marine products	0.20	10	0	0.0	0.000	0.0	0.0000	0.0	11.47	0.000	0.0
Dried mashed potato	0.20	0	0	—	—	—	—	—	0.47	—	—
Nonpermissible foods*	—	1,007	7	0.7	0.060	—	0.0004	—	642.93	—	—
Total (or mean)	—	2,046	188	(9.2)	—	(35.3)	—	(6.2)	675.51	0.110	—
Breakdown of nonpermissible foods (detected only)	—	11	3	27.3	0.110	—	0.0300	—	0.03	0.001	0.8
Chewing gum	—	271	3	1.1	0.022	—	0.0002	—	21.74	0.005	4.4
Confectionery, "kashi"	—	151	1	0.7	0.027	—	0.0002	—	20.18	0.004	3.0
Processed marine products	—	—	—	—	—	—	—	—	—	—	—
Daily intake of BHA	—	—	—	—	—	—	—	—	—	0.119	100.0

\* Breakdown of nonpermissible foods in which BHA was not detected: seasonings (87 samples), frozen food (74), pre-cooked foods "sozai" (57), meat products (40), vegetable products (34), bottled or canned foods (28), noodles (28), cereals (27), etc.

Table 6. Concentration of BHT in Foods and Estimated Daily Intake

Foods categorized by the standards for use	Regulation limit (g/kg)	Number of samples		Detection rate (%)	Detected samples		Tested samples		Daily intake per person		Contribution to total intake (%)
		Tested	Detected		Mean conc. (g/kg)	Ratio to limit (%)	Mean conc. (g/kg)	Ratio to limit (%)	Food (g)	BHT (mg)	
Frozen marine products	1.0	10	0	0.0	0.0000	0.0	0.0000	0.0	1.80	0.0000	0.0
Frozen whale meat	1.0	0	0	—	—	—	—	—	0.01	—	—
Fats and oils	0.20	132	4	3.0	0.0650	32.5	0.0020	1.0	10.32	0.0203	18.6
Butter	0.20	20	0	0.0	0.0000	0.0	0.0000	0.0	1.04	0.0000	0.0
Dried marine products	0.20	872	7	0.8	0.0691	34.6	0.0006	0.3	7.47	0.0041	3.8
Salted marine products	0.20	20	0	0.0	0.0000	0.0	0.0000	0.0	11.47	0.0000	0.0
Dried mashed potato	0.20	0	0	—	—	—	—	—	0.47	—	—
Chewing gum	0.75	13	8	61.5	0.0854	11.4	0.0525	7.0	0.03	0.0016	1.4
Nonpermissible foods*1	—	1,006	8	0.8	0.0624	—	0.0005	—	642.90	—	—
Total (or mean)	—	2,073	27	(1.3)	—	(24.4)	—	(0.4)	675.5	0.0260	—
Breakdown of nonpermissible foods (detected only)*2	—	275	5	1.8	0.0622	—	0.0011	—	21.74	0.0246	22.5
Confectionery, "kashi"	—	57	2	3.5	0.0850	—	0.0030	—	19.70	0.0588	53.7
Pre-cooked food, "sozai"	—	—	—	—	—	—	—	—	—	—	—
Daily intake of BHT	—	—	—	—	—	—	—	—	—	0.1094	100.0

\*1 Breakdown of nonpermissible foods in which BHT was not detected: marine products (153 samples), seasonings (88), frozen foods (74), meat products (40), vegetable products (33), noodles (28), bottled or canned foods (28), cereals (24), pickled foods (20), nuts (13), milk products (11), dried fruits (11), etc.

\*2 Foods categorized as "nonpermissible foods" or "other foods" by the local governments were omitted from the breakdown.

Table 7. Concentration of Propylene Glycol (PG) in Foods and Estimated Daily Intake

Foods categorized by the standards for use	Regulation limit (g/kg)	Number of samples		Detection rate (%)	Detected samples		Tested samples		Daily intake per person		Contribution to total intake (%)
		Tested	Detected		Mean conc. (g/kg)	Ratio to limit (%)	Mean conc. (g/kg)	Ratio to limit (%)	Food (g)	PG (mg)	
Raw noodles	20	1,765	681	38.6	11.29	56.5	4.36	21.8	7.80	34.0	43.9
Smoked cuttlefish	20	54	0	0.0	0.00	0.0	0.00	0.0	0.40	0.0	0.0
Crust of Chinese pastry	12	303	94	31.0	7.79	64.9	2.42	20.1	1.40	3.4	4.4
Chewing gum	6.0	0	0	—	—	—	—	—	0.03	—	—
Other foods*1	6.0	344	27	7.8	2.51	41.9	0.20	3.3	664.08	—	—
Total (or mean)	—	2,466	802	(32.5)	—	(57.0)	—	(18.5)	673.71	—	—
Breakdown of other foods (detected only)*2	6.0	238	21	8.8	2.52	42.0	0.22	3.7	25.00	5.6	7.2
Boiled noodles	6.0	13	2	15.4	3.10	51.7	0.48	7.9	11.79	5.6	7.3
Steamed noodles	6.0	1	1	100.0	5.00	83.3	5.00	83.3	5.79	29.0	37.4
Dried noodles	6.0	1	1	100.0	0.10	1.7	0.10	1.7	—	—	—
Fats and oils for noodle kneading	—	—	—	—	—	—	—	—	—	—	—
Daily intake of PG	—	—	—	—	—	—	—	—	—	77.5	100.0

\*1 Breakdown of other foods in which propylene glycol was not detected: marine products (28 samples), wine (6), etc.

\*2 Foods categorized as "other foods" by the local governments were omitted from the breakdown.



Table 8. Concentration of Sodium Saccharin in Foods and Estimated Daily Intake

Foods categorized by the standards for use	Regulation (g/kg)	Number of samples		Detected samples		Tested samples		Daily intake per person		Contribution to total intake (%)	
		Tested	Detected	Detection rate (%)	Mean conc. (g/kg)	Ratio to limit (%)	Mean conc. (g/kg)	Ratio to limit (%)	Food (g)		Sodium saccharin (mg)
Chewing gum*1	0.050	5	0	0.0	0.000	0.0	0.000	0.0	0.03	0.00	0.0
Koji-pickled foods, "koji-zuke"	2.0	107	21	19.6	0.538	26.9	0.106	5.3	0.01	0.00	0.0
Vinegar-pickled foods, "su-zuke"	2.0	560	163	29.1	0.704	35.2	0.205	10.2	0.97	0.20	2.7
Salted dried radish, "takuan-zuke"	2.0	566	343	60.6	0.517	25.9	0.314	15.7	3.91	1.23	16.9
Powdered nonalcoholic beverages	1.5	7	2	28.6	0.390	26.0	0.111	7.4	0.01	0.00	0.0
Lees-pickled foods, "kasu-zuke"	1.2	107	38	35.5	0.443	36.9	0.157	13.1	0.27	0.04	0.6
Bean paste-pickled foods, "miso-zuke"	1.2	121	36	29.8	0.485	40.4	0.144	12.0	0.09	0.01	0.2
Soy sauce-pickled foods, "shoyu-zuke"	1.2	1,288	221	17.2	0.435	36.3	0.075	6.2	2.45	0.18	2.5
Processed marine products	1.2	850	44	5.2	0.309	25.8	0.016	1.3	33.72	0.54	7.4
Seaweed products	0.50	30	1	3.3	0.810	162.0	0.027	5.4	0.43	0.01	0.2
Soy sauce	0.50	525	204	38.9	0.289	57.8	0.112	22.5	23.16	2.60	35.8
Boiled down foods with soy sauce, "tsukudani"	0.50	622	5	0.8	0.142	28.4	0.001	0.2	0.98	0.00	0.0
Boiled beans	0.50	257	1	0.4	0.020	4.0	0.000	0.0	2.66	0.00	0.0
Fish paste products	0.30	2,169	166	7.7	0.106	35.4	0.008	2.7	14.30	0.12	1.6
Vinegar	0.30	32	2	6.3	0.160	53.3	0.010	3.3	2.68	0.03	0.4
Nonalcoholic beverages	0.30	1,145	104	9.1	0.205	68.4	0.019	6.2	69.10	1.29	17.7
Nonalcoholic beverages, consumed in 5-fold or more dilution	1.5	16	13	81.3	0.690	46.0	0.561	37.4	0.01	0.01	0.1
Syrup	0.30	13	0	0.0	0.000	0.0	0.000	0.0	0.01	0.00	0.0
Worcester sauce	0.30	157	18	11.5	0.183	60.9	0.021	7.0	3.14	0.07	0.9
Milk beverages	0.30	39	0	0.0	0.000	0.0	0.000	0.0	27.78	0.00	0.0
Lactic acid bacteria beverages	0.30	32	0	0.0	0.000	0.0	0.000	0.0	0.01	0.00	0.0
Ice candy	0.30	82	6	7.3	0.137	45.6	0.010	3.3	5.10	0.05	0.7
Ice cream	0.20	131	4	3.1	0.070	35.0	0.002	1.1	1.97	0.00	0.1
Sweetened bean paste, "an"	0.20	95	0	0.0	0.000	0.0	0.000	0.0	—	—	—
Jam	0.20	134	0	0.0	0.000	0.0	0.000	0.0	1.16	0.00	0.0
Other pickled foods	0.20	585	50	8.5	0.581	290.4	0.050	24.8	15.12	0.75	10.3
Fermented milk	0.20	76	0	0.0	0.000	0.0	0.000	0.0	0.01	0.00	0.0
Flour paste	0.20	52	1	1.9	0.070	35.0	0.001	0.7	—	—	—
Soy bean paste, "miso"	0.20	360	8	2.2	0.141	70.6	0.003	1.6	14.36	0.05	0.6
Confectionery	0.10	1,819	2	0.1	0.340	340.0	0.000	0.4	21.74	0.01	0.1
Canned or bottled foods	0.20	315	0	0.0	0.000	0.0	0.000	0.0	34.54	0.00	0.0
Nonpermissible foods*2	—	2,944	15	0.5	0.169	—	0.001	—	393.99	—	—
Total (or mean)	—	15,241	1,468	(9.6)	—	(48.4)	—	(5.7)	673.71	—	—
Breakdown of nonpermissible foods (detected only)											
Seasonings	—	367	11	3.0	0.175	—	0.005	—	13.56	0.07	1.0
Pre-cooked foods "sozai"	—	777	4	0.5	0.150	—	0.001	—	19.70	0.02	0.2

Table 8. Continued

Foods categorized by the standards for use	Regulation (g/kg)		Number of samples		Detection rate (%)		Detected samples		Tested samples		Daily intake per person		Contribution to total intake (%)
			Tested	Detected	Mean conc. (g/kg)	Ratio to limit (%)	Mean conc. (g/kg)	Ratio to limit (%)	Food (g)	Sodium saccharin (mg)			
Daily intake of sodium saccharin												7.27	100.0

\*1 Saccharin is only allowed in chewing gum, while other allowable foods can contain only sodium saccharin.

\*2 Breakdown of nonpermissible foods in which sodium saccharin was not detected: meat products (627 samples), processed vegetables or fruits (150), dry fruits (148), frozen foods (120), alcoholic beverages (89), processed cereals (86), milk products (64), condensed or diluted seasoned soy sauce ("tare" or "tsuyu") (62), foods soaked in syrup (48), noodles (31), boiled vegetables (24), etc.

Table 9. Estimated Daily Intake of Relevant Food Additives and Ratios to ADI

Food additives	Present study (1998)	Ratio to ADI (%)	Daily intake (mg/person)									ADI (mg) (as 50 kg b.w.)
			Japan*1 (1996)	Japan*2 (1994)	Finland*3 (1980)	U.K.*4 (1984-1986)	Japan*5 (1998)	Japan*6 (1996-1997)	Japan*7 (1997-1999)	Japan*8 (2000)		
Nitrite	0.205	6.8	1.07	0.318	3.5	2	0.310	3.837	0.89	0.339	3	
Nitrate	0.532	0.3	1.43	0.430	6.4	1.3	0.251	190.8	189	289	185	
Sulfur dioxide	4.31	12.3	1.45	1.63	4	18.4	7.83	—	0.057	0	35	
Butylated hydroxyanisole	0.119	0.5	0.105	0.17	0.17*9	0.4	0.260	—	0	0	25	
Butylated hydroxytoluene	0.109	0.7	0.220	0.051	—	0.2	0.347	—	0.013	0	15	
Propylene glycol	77.5	6.2	41.4	57.1	5.9	54.1	34.7	—	31.7	10.6	1,250	
Sodium saccharin	7.27	2.6	7.64	7.26	2.5	2.5	3.69	—	3.22	0.760	280	

\*1 Based on the official inspection results<sup>2)</sup>

\*2 Based on the official inspection results<sup>1)</sup>

\*3 Based on the official analysis and data in journals<sup>13)</sup>

\*4 Based on the production and usage amounts<sup>14)</sup>

\*5 Based on the production and usage amounts<sup>15)</sup>

\*6 In middle-aged persons based on the duplicate portion method<sup>16)</sup>

\*7 Based on the market basket method<sup>17), 18)</sup>

\*8 In middle-aged persons based on the market basket method<sup>7)</sup>

\*9 Sum of butylated hydroxyanisole and butylated hydroxytoluene

sample of chewing gum was inspected in 1998. In "other foods", propylene glycol was detected in boiled noodles (21 of 238 samples, 8.8%), and the mean concentration in all tested boiled noodles was 0.22 g/kg, corresponding to 3.7% of the allowable limit (6.0 g/kg). A dried noodle sample (1 of 1 sample) contained 5.00 g/kg, corresponding to 83.3% of the limit (6.0 g/kg). No propylene glycol was detected in smoked cuttlefish (54 samples), marine products (28), wine (6), etc.

#### g) Sodium saccharin

Sodium saccharin was detected in 1,468 (9.6%) of 15,241 samples tested (Table 8). The detection rate in the tested samples in which sodium saccharin (including saccharin) is allowed to be used was 11.8%, and the mean concentration of sodium saccharin was 5.7% of the allowable limit. The highest detection rate (81.3%) was observed in nonalcoholic beverages consumed in a 5-fold or more dilution, the mean concentration in all tested samples was 0.561 g/kg, and the concentration was 37.4% of the allowable limit (1.5 g/kg). It was the highest concentration ratio to the allowable limit, though sodium saccharin is diluted to less than 0.11 g/kg before intake. Sodium saccharin was not detected in some allowable foods such as canned or bottled foods (315 samples), jam (134), sweetened bean paste "an" (95), and fermented milk (76). Saccharin was not detected in 5 samples of chewing gum in which saccharin, but not sodium saccharin, is allowed. Sodium saccharin was detected in 15 of 2,944 samples of nonpermissible foods, namely 11 samples of 367 seasonings and 4 samples of 777 pre-cooked foods "sozai". No sodium saccharin was detected in the following nonpermissible foods: meat products (627 samples), processed vegetables or fruits (150), dry fruits (148), frozen foods (120), alcoholic beverages (89), processed cereals (86), milk products (64), etc.

### 3. Estimation of the daily intake per person

The estimated daily intakes based on the official inspection results in fiscal year 1998 are summarized in Table 9, together with the results reported in previous papers based on the official inspection in fiscal years 1994<sup>1)</sup> and 1996<sup>2)</sup>, those reported in comparative references<sup>7), 13)-18)</sup>, and the ADIs<sup>19)</sup>.

The estimated daily intake of the seven food additives per person was as follows: nitrite, 0.205 mg; nitrate, 0.532 mg; sulfur dioxide, 4.31 mg; BHA, 0.119 mg; BHT, 0.109 mg; propylene glycol, 77.5 mg; and sodium saccharin, 7.27 mg. These values are 6.8%, 0.3%, 12.3%, 0.5%, 0.7%, 6.2%, and 2.6%, respectively, of the ADIs when body weight is assumed to be 50 kg. The present results on the daily intake of these seven food additives were similar to the results in fiscal years 1994<sup>1)</sup> and 1996<sup>2)</sup>, and those of other reports<sup>13)-17)</sup>, except for some daily intakes which were estimated by the market-basket method<sup>7), 18)</sup>. Foods that contributed most to the daily intake of these seven food additives were; meat products which accounted for 65.8% of the daily intake of nitrite, meat products which accounted for 43.8% of

nitrate, dried gourd strips "kampyo" which accounted for 21.3% of sulfur dioxide, dried marine products which accounted for 91.8% of BHA, pre-cooked foods "sozai" which accounted for 53.7% of BHT, raw noodles which accounted for 43.9% of propylene glycol, and soy sauce which accounted for 35.8% of sodium saccharin.

### Discussion

The estimated daily intakes of the seven food additives (nitrite, nitrate, sulfur dioxide, BHA, BHT, propylene glycol, and sodium saccharin) per person based on the results of official inspections of 34,489 food samples in fiscal year 1998 were 0.205, 0.532, 4.31, 0.119, 0.109, 77.5, and 7.27 mg, corresponding to 6.8%, 0.3%, 12.3%, 0.5%, 0.7%, 6.2%, and 2.6% of the ADIs, respectively. These results were similar to those obtained in the previous reports<sup>1), 2)</sup>, indicating that the yearly trend of the daily intake of these seven food additives is stable, and that intake amounts have been maintained below 10% of the ADIs in most cases.

The estimated daily intake of nitrate was only 0.532 mg in the present study, but it has been reported as 289 mg in adults by the market-basket method<sup>7)</sup>, and 190.8 mg in middle-aged men by the duplicate portion method<sup>16)</sup>. The results obtained by the market-basket method and the duplicate portion method include nitrate in vegetables but the estimation based on the official inspection does not, because fresh vegetables and vegetable products are not required to be inspected. The nitrate found in this study would have been mainly introduced as a food additive, because most of the samples tested were meat products and marine products. The estimated intake of nitrate reported in Finland<sup>13)</sup>, 6.4 mg, was obtained from official analytical results, by using a similar method to that used in the present paper. The estimated daily intake of nitrate based on the amount of production and usage was 1.3 mg in the U.K.<sup>14)</sup>, and 0.251 mg in Japan<sup>15)</sup>. The amounts of nitrate estimated in 1994, 1996, and 1998 were closer to the results obtained by the production and usage amount method in Japan than those obtained by the market-basket method and the duplicate portion method. Nitrate intake estimated by the market-basket method and the duplicate portion method may reflect the total intake amount, and that by the present method and the production and usage amount method may mainly reflect the intake of nitrate as a food additive.

The UK estimated<sup>14)</sup> the daily intake of sulfur dioxide at 92.7 mg per person based on the results of a preliminary surveillance of production amounts, and the estimate was corrected to 18.4 mg per person based on the determination in wine and some other foods. On the other hand, results obtained by the market-basket method in Japan were 0.057 mg<sup>18)</sup> or 0 mg<sup>7)</sup> per person. The daily intake of sulfur dioxide by the production and usage amount method may be overestimated due to decomposition during the processing and handling of food, and that by the market-basket method may be underestimated because of decomposition during prep-

aration of the homogenates of many kinds of food. The use of BHA and BHT is limited to a few kinds of food, and the concentration was low. The daily intake of BHA and BHT was estimated as 0 mg by the market-basket method<sup>7)</sup>, though their usage was found in dried marine products, and fats and oils in the present study. The results indicate that the present method is advantageous to estimate the daily intake of food additives used at low concentrations and in limited kinds of food.

The mean concentrations of food additives in foods and estimated daily intakes of those food additives reported in the present series of studies include both over- and underestimation factors for the reasons described below.

(a) Sampling in the official inspection is basically done for foods in which food additives are permitted to be used and nonpermissible foods in which food additives might be used. Therefore, if foods were chosen at random for the inspection, the detection rates and the mean concentration may be lower than the present results.

(b) Foods containing excessive amounts of food additives and nonpermissible foods in which a food additive has been used are excluded from markets by the local governments. The present results include these foods, which would be banned from sale.

(c) Concentrations of food additives in foods in which they were not detected were calculated as 0 g/kg. Some foods may contain small amounts of these food additives naturally or by carryover at levels lower than the detection limits.

Among these factors affecting the mean concentration of food additives, (a) and (b) may result in overestimation and (c) may be a cause of underestimation. When non-detectable samples were presumed to contain additives at the level of the detection limits, the estimated daily intakes of four antifungal agents were still less than 1% of the ADI<sup>20)</sup>. Factors (a) and (b) would most strongly affect the results. It is therefore more likely that the results obtained in the present series are overestimated rather than underestimated.

In general, the daily intake of food additives is relatively constant when the estimation is performed by the same method, but the results given by other methods sometimes differ. It is difficult to directly compare the results of the present study with those obtained using another method and those in other countries because of differences in conditions for surveillance and in dietary habits. However, the results shown in Table 9 will be extremely useful in understanding daily intake levels of food additives to assure food safety.

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