日本における行政検査結果(1994年度)による食品中のBHA,BHT,プロピレングリコール,サッカリンナトリウム濃度の実態及びそれに基づいた摂取量の推定

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Original

Evaluation of the Contents of BHA, BHT, Propylene Glycol, and Sodium Saccharin in Foods and Estimation of Daily Intake Based on the Results of Official Inspection in Japan in Fiscal Year 1994*

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The concentrations of BHA, BHT, propylene glycol, and sodium saccharin in foods sold in Japan were estimated using the results of official inspections by 88 local governments, representing most of Japan, in fiscal year 1994. A total of 27,474 samples, including 1,720 imported foods, were inspected. The detection rates of BHA, BHT, propylene glycol, and sodium saccharin were 12.2%, 2.3%, 36.3%, and 11.2%, respectively. The mean concentrations of these food additives in all samples in which their use is allowed were 1.3%, 1.0%, 13.6%, and 5.0%, respectively, of the legally permitted levels. The estimated daily intakes of these food additives were 0.169, 0.051, 57.1, and 7.26 mg/person, respectively, when calculated based on their mean concentrations in foods obtained in the present study and daily food consumption data. The estimated daily intakes were 0.7%, 0.3%, 4.6%, and 2.9%, respectively, of the acceptable daily intake (ADI). The food that contributed most to the daily intake of BHA was dried fish (70.2% of BHA intake); that of BHT was chewing gum (61.6%); that of propylene glycol was raw noodles (67.3%); and that of sodium saccharin was nonalcoholic beverages (27.0%).

Key words: food additive; BHA; BHT; propylene glycol; saccharin; sodium saccharin; daily intake; surveillance in food; official inspection

Introduction

Determination of daily intake of food additives is important as it facilitates comparison of the daily intake in Japan with the acceptable daily intakes (ADIs) prescribed by the Joint FAO/WHO Food Standard Programme[1]. The International Life Science Institute-Europe has organized a workshop to assess food additive intake[2]. The "Investigation Group of the Daily Intake of Food Additives in Japan" (the Japanese group)[3]-[5] has worked for many years to estimate the daily intake of food additives by a market-basket method. The daily intake of food additives has also been reported by the Ministry of Agriculture, Fisheries and Food of the United

* Concentration of Food Additives in Foods and the Daily Intake in Japan (Report No. 4) (Report No. 3 was entitled "Evaluation of the Inorganic Food Additive (Nitrite, Nitrate, and Sulfur Dioxide) Content of Foods and Estimation of Daily Intake Based on the Results of Official Inspection in Japan in Fiscal Year 1994", published in this Journal, **39**, 78-88 (1998).)

Kingdom (UK)[6]. The UK group calculated the daily intake of food additives from the results of surveillance of the amount of food additives produced and the amounts used by food makers based on the results of a questionnaire survey. Penttilä, *et al.*[7] (the Finnish group) estimated the daily intake of food additives based on the concentration in foods reported by the National Board of Trade of Finland. We have reported the daily intake of five preservatives[8], four antifungal agents[9], and three inorganic food additives[10] estimated on the basis of the results of official inspection of foods in markets performed by local governments in Japan.

In the present paper, we have estimated the mean concentrations of some other food additives (butylated hydroxyanisole (BHA), butylated hydroxytoluene (BHT), propylene glycol, and sodium saccharin) in commercial foods based on the official inspection data in Japanese fiscal year 1994 and deduced the daily intakes per person from the mean concentrations and amounts of foods consumed.

Methods

The methods used were similar to those previously described[8].

1. Investigation methods and items studied

a) Questionnaire

A questionnaire on the results of official inspection for food additives from April 1, 1994 to March 31, 1995 (Japanese fiscal year 1994) was sent by the Division of Food Chemistry, Ministry of Health and Welfare of Japan to all 103 local governments in the country[11]. These local governments are obligated to establish public health centers by the Regional Public Health Law.

b) Items of queried

BHA, BHT, propylene glycol, and sodium saccharin were investigated in the present study. The names of both imported and domestic foods, the numbers of food samples inspected, the numbers of samples in which these food additives were detected, the maximum, minimum, and mean concentrations of the food additives in the samples in which they were detected, and the detection limit of the analysis for each food additive were recorded. Saccharin in foods was expressed as sodium saccharin except that in chewing gums, according to the Japanese Standards for Use of Food Additives[12]. Free saccharin is allowed to be used only in chewing gum.

c) Sampling of foods and methods of determining food additives

Samples were chosen by food inspectors in markets or factories manufacturing food products according to the yearly program of each local government. The gas chromatographic methods prescribed by the Environmental Health Bureau of the Ministry of Health and Welfare were used for the determination of BHA[13], BHT[14], propylene glycol[15], and saccharin[16].

2. Analyses of reports from the local governments

Data sent from the local governments were classified according to the categories of foods in the Japanese Standards for Use of Food Additives[12]. Foods in which these compounds are not allowed to be used as food additives were categorized according to the "Guide for Categorization of Food"[17]. Concentrations of these food additives were regarded as 0 mg/kg irrespective of detection limits when these food additives were not detected, as described in the previous papers[8)-10].

3. Calculations and estimation of daily intake

The daily intakes of the food additives BHA, BHT, propylene glycol, and sodium saccharin per person were estimated based on the mean concentrations of these food additives in foods obtained in the present study and the daily food consumption levels reported by the Japanese group[3),5] and the Ministry of Health and Welfare of Japan[18]. When the level of consumption of a food was unknown, 0.1 g was assumed to be the daily consumption per person, as described in previous reports[8)-10].

Results and Discussion

1. Questionnaire return and number of samples

Ninety-six of the 103 local governments (93.2%), representing most of Japan, i.e., 42 prefectures, 33 cities, and 21 wards, replied to the questionnaire. Four kinds of food additives, BHA, BHT, propylene glycol, and sodium sac-

Table 1. Number of Local Governments that Performed Inspections of BHA, BHT, Propylene Glycol, and Sodium Saccharin, and Numbers of Inspected Food Samples

Food additive	Number of local governments	Numbers of food samples		
		Domestic	Imported	Total
Butylated hydroxyanisole	72	1,812	310	2,122
Butylated hydroxytoluene	71	1,781	324	2,105
Propylene glycol	79	3,275	8	3,283
Sodium saccharin	86	18,886	1,078	19,964

Table 2. Detection Limits of BHA, BHT, Propylene Glycol, and Sodium Saccharin

Food additive	Detection limits					
	0.01 g/kg		0.001–0.01 g/kg		0.005–0.05 g/kg	
	Govern. (%)*1	Sample (%)*2	Govern. (%)*1	Sample (%)*2	Govern. (%)*1	Sample (%)*2
Butylated hydroxyanisole	57.7	53.2			87.3	86.7
Butylated hydroxytoluene	58.6	50.6			87.1	88.0
Propylene glycol	39.2	32.1	92.4	82.7		
Sodium saccharin	74.4	83.4			107.0	98.6

*1 Ratio of local governments setting the indicated detection limits to the total number of local governments that performed inspections.

*2 Ratio of samples at each detection limit to the total number of samples.

charin, described in the present paper were inspected by 88 local governments (91.7% of the 96 local governments replied). Inspection for BHA was performed by 72 local governments, for BHT by 71 local governments, for propylene glycol by 79 local governments, and for sodium saccharin by 86 local governments. The total number of foods inspected was 27,474, and included 1,720 imported foods (Table 1). Results obtained in the present study were therefore considered to be highly reliable, since inspection results were received from most of the local governments in Japan and large numbers of samples were tested.

2. Detection limits

The detection limits of these food additives are summarized in Table 2. Although the detection limits are specific to the methods in the case of standard solutions of each compound, they are affected by food components or are modified to appropriate concentrations according to the maximum allowable levels for use of these food additives. The detection limits were set at 0.01 g/kg for BHA, BHT, and sodium saccharin by more than 57.7% of the local governments that inspected them, and between 0.005 and 0.05 g/kg by more than 87.1% of the local governments. The detection limit for propylene glycol was set at 0.01 g/kg by 39.2% of the local governments, and between 0.001 and 0.01 g/kg by 92.4% of them. The percentages of the samples inspected at the detection limit of 0.01 g/kg to the total samples for BHA, BHT and sodium saccharin were 53.2%, 50.6%, and 83.4%, respectively. That of propylene glycol at the detection limit of 0.01 g/kg was 32.1%. The legally permissible limits for sodium saccharin are categorized into 31 groups of foods including saccharin for chewing gum as shown in Table 6. Some local governments then set the detection limit for sodium saccharin at several different levels depending on the standards for use of sodium saccharin. The total percentage of local governments, therefore, exceeded 100%.

3. Concentration of BHA in foods

BHA was detected in 258 (12.2%) out of 2,122 samples, including some nonpermissible foods (Table 3). The detection rate in tested foods in

Table 3 Concentration of Butylated Hydroxyanisole (BHA) in Foods and Estimated Daily Intake

Foods categorized by the standards for use	Regulation (g/kg)	Numbers of samples		Detection rate (%)	Maximum conc. (g/kg)	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 fish	1.0	20	0	0.0	0	0.000	0.0	0.000	0.0	1.8	0.000	0.0
Frozen whale meat	1.0	0								0.1		
Fats and oils	0.20	102	1	1.0	0.01	0.010	5.0	0.000	0.0	12.4	0.001	0.7
Butter	0.20	13	0	0.0	0	0.000	0.0	0.000	0.0	0.9	0.000	0.0
Dried fish	0.20	1,288	248	19.3	0.58	0.065	32.5	0.012	6.2	9.5	0.119	70.2
Salted fish	0.20	55	2	3.6	0.045	0.028	13.8	0.001	0.5	5.7	0.006	3.4
Mashed potato (dried)	0.20	0								0.4		
Nonpermissible foods*	—	644	7	1.1	0.8	0.140	—	0.002	—	737.1		
Total (or mean)	—	2,122	258	(12.2)	—	—	—	—	—	767.9		
*Breakdown (detected only)												
Chewing gum	—	18	7	38.9	0.8	0.140	—	0.054	—	0.8	0.044	25.7
Daily intake of BHA											0.169	100.0

* Nonpermissible foods and numbers of samples in which BHA was not detected were as follows.
Fish products 91, confectionery 122, meat products 27, wine 11, seasoning 25, canned or bottled food 24, noodles 7, health food 12, cheese 18, nonalcoholic beverages 7, salted vegetables 6, *etc.*

which BHA is allowed to be used was 17.0%, and the mean concentration of BHA was 1.3% of the allowable limits by the Japanese Food Sanitation Law[12]. The highest detection rate (19.3%) was observed in dried fish (248 out of 1,288 samples). The detection rates of BHA in other foods were 3.6% or less. BHA may be replaced by tocopherol[18] as an antioxidant. The mean concentration in tested dried fish was 0.012 g/kg, and was 6.2% of the legal allowable limit (0.20 g/kg). BHA was detected in 7 out of 18 imported samples of chewing gum, which is a nonpermissible food. The mean concentration in the 7 samples was 0.140 g/kg. Ohta, *et al.*[20] reported that no BHA was detected in domestic chewing gum at a detection limit of 1 mg/kg. No BHA was detected in 626 other nonpermissible foods inspected in fiscal year 1994.

4. Concentration of BHT in foods

BHT was detected in 49 out of 2,105 samples (2.3%) (Table 4). The detection rate in all foods in which the use of BHT is allowed was 2.5%, and the mean concentration of BHT was 1.0% of the allowable limit. The mean concentration in chewing gum, in which the use of BHT is permitted, was 0.039 g/kg, and was 5.3% of the allowable limit (0.75 g/kg). BHT was detected in one sample of imported crackers out of 600 nonpermissible foods. BHT in the crackers was subsequently confirmed to have been carried over from edible oil by the local government that inspected the sample.

5. Concentration of propylene glycol

Propylene glycol is allowed to be used in all processed foods as a carrier solvent or a wetting agent, and is also allowed as a softening agent for chewing gum. The Japanese Food Sanitation Law sets individual maximum concentrations for 4 categories of foods, raw noodles, smoked cuttlefish, crust of Chinese pastry, and chewing gum, and 6.0 g/kg for all other foods (categorized as "other foods" by the Law) (Table 5).

Propylene glycol was detected in 1,193 out of 3,283 samples (36.3%) (Table 5) and the mean concentration of propylene glycol was 13.6% of the allowable limit. The highest detection rate (42.4%) of propylene glycol was observed in raw noodles (1,032 out of 2,432 samples). Chewing gum was not inspected for propylene glycol in fiscal year 1994. The detection rate of propylene glycol in the "other foods" was 5.9% (24 detected samples in 407 tested samples), and the mean concentration was 0.18 g/kg corresponding to 2.9% of the limit (6.0 g/kg). The breakdown of the "other foods" in which propylene glycol was detected is also shown in Table 5. Propylene glycol was detected in 5.2% of boiled noodles at 0.16 g/kg, corresponding to 2.6% of the limit, and in 50.0% (one of 2 samples) of cakes at 2.70 g/kg, corresponding to 45.0% of the limit. Propylene glycol, at 0.19 g/kg (3.1% of the limit), was also detected in some 20 other foods out of 301 samples.

6. Concentration of sodium saccharin

Saccharin is only allowed to be used in chewing gum at a maximum concentration of 0.050 g/kg as saccharin. Another 30 categories of foods are allowed to contain sodium saccharin, and the allowable limits in these foods are expressed as sodium saccharin.

Sodium saccharin was detected in 2,239 (11.2%) out of 19,964 samples (Table 6). The detection rate in tested foods in which saccharin or sodium saccharin is allowed to be used was 12.5%, and the mean concentration was 5.0% of the allowable limits. The highest detection rate (57.1%, 493 out of 863 samples) and the highest concentration (0.281 g/kg, mean of tested samples) were observed in salted dried radish (Takuan-zuke). The highest concentration rate (35.8%) was observed in foods categorized as "other pickled foods" which means pickled foods other than Koji-zuke, Su-zuke, Takuan-zuke, Kasu-zuke, Miso-zuke, and Shoyu-zuke. In the present study, 21 chewing gum samples were inspected, but no saccharin was detected in any sample.

7. Evaluation of food additive concentrations in foods

The concentrations of BHA, BHT, propylene glycol, and sodium saccharin shown in Tables 3–6 might be over- or underestimated due to the following reasons; a) most samples inspected were permitted foods for these food additives, b) although many nonpermissible foods for these

Table 4. Concentration of Butylated Hydroxytoluene (BHT) in Foods and Estimated Daily Intake

Foods categorized by the standards for use	Regulation (g/kg)	Numbers of samples		Detection rate (%)	Maximum conc. (g/kg)	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 fish	1.0	20	0	0.0	0	0.000	0.0	0.000	0.0	1.8	0.000	0.0
Frozen whale meat	1.0	0								0.1		
Fats and oils	0.20	102	3	2.9	0.05	0.037	18.3	0.001	0.5	12.4	0.013	26.1
Butter	0.20	18	0	0.0	0	0.000	0.0	0.000	0.0	0.9	0.000	0.0
Dried fish	0.20	1,239	7	0.6	0.05	0.028	13.9	0.000	0.1	9.5	0.001	2.9
Salted fish	0.20	56	0	0.0	0	0.000	0.0	0.000	0.0	5.7	0.000	0.0
Mashed potato	0.20	0								0.4		
Chewing gum	0.75	70	38	54.3	0.55	0.073	9.7	0.039	5.3	0.8	0.032	61.6
Nonpermissible foods*	—	600	1	0.2	0.016	0.016	—	0.000	—	736.3		
Total (or mean)	—	2,105	49	(2.3)	—	—	—	—	—	767.9		
*Breakdown (detected only)												
Cracker (carry-over from oils)	—	1	1	100.0	0.016	0.016	—	0.016	—	0.3	0.005	9.4
Daily intake of BHT											0.051	100.0

* Nonpermissible foods and numbers of samples in which BHT was not detected were as follows.
Confectionery 104, fish products 98, meat products 27, seasoning 25, canned or bottled food 24, cheese 18, health food 12, noodles 7, nonalcoholic beverages 7, salted vegetables 6, boiled beans 4, dried fruits 3, *etc.*

Table 5. Concentration of Propylene Glycol (PG) in Foods and Estimated Daily Intake

Foods categorized by the standards for use	Regulation (g/kg)	Numbers of samples		Detection rate (%)	Maximum conc. (g/kg)	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	2,432	1,032	42.4	31	11.61	58.0	4.93	24.6	7.8	38.4	67.3
Smoked cuttlefish	20	78	0	0.0	0	0.00	0.0	0.00	0.0	0.4	0	0.0
Crust of Chinese pastry	12	366	137	37.4	40	8.60	71.7	3.22	26.8	1.4	4.5	7.9
Chewing gum	6.0	0								0.8		
Other foods*	6.0	407	24	5.9	11	2.97	49.4	0.18	2.9	757.5	132.5	
Total (or mean)	—	3,283	1,193	(36.3)	—	—	—	—	—	767.9	175.5	
*Breakdown (detected only)												
Boiled noodles	6.0	58	3	5.2	4	3.00	50.0	0.16	2.6	7.8	1.2	2.1
Cake	6.0	2	1	50.0	5.4	5.40	90.0	2.70	45.0	4.8	13	22.7
Other foods	6.0	301	20	(6.6)	11	2.83	47.1	0.19	3.1			
Daily intake of PG											57.1	100.0

* Other foods and number of samples in which propylene glycol was not detected were as follows.
Dried fish 25, processed foods 13, dried plum 2, buckwheat noodles (Soba) 2, dry noodles 1, butter 1, brandy 1, *etc.*

Table 6. Concentration of Sodium Saccharin in Foods and Estimated Daily Intake

Foods categorized by the standards for use	Regulation (g/kg)	Numbers of samples		Detection rate (%)	Maximum conc. (g/kg)	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)	Na Saccharin (mg)	
Chewing gum	0.050*[1]	21	0	0.0	0	0.000	0.0	0.000	0.0	0.8	0.000	0.0
Koji-pickled foods (Koji-zuke)	2.0	135	39	28.9	1.2	0.490	24.5	0.141	7.1	0.1	0.014	0.2
Vinegar-pickled foods (Su-zuke)	2.0	954	325	34.1	2.24	0.651	32.6	0.222	11.1	2.2	0.488	6.7
Salted dried radish (Takuan-zuke)	2.0	863	493	57.1	2	0.492	24.6	0.281	14.1	3.4	0.955	13.1
(Koji-zuke, Su-zuke and Takuan-zuke)	2.0	28	11	39.3	0.74	0.563	28.2	0.221	11.1	—	—	—
Powdered nonalcoholic beverages	1.5	29	5	17.2	0.66	0.570	38.0	0.098	6.6	0.5	0.049	0.7
Lees-pickled foods (Kasu-zuke)	1.2	262	77	29.4	1.3	0.482	40.2	0.142	11.8	1.2	0.170	2.3
Bean paste-pickled foods (Miso-zuke)	1.2	210	34	16.2	0.95	0.361	30.1	0.058	4.9	0.3	0.017	0.2
Soy sauce-pickled foods (Shoyu-zuke)	1.2	1,966	381	19.4	1.33	0.461	38.4	0.089	7.4	6.3	0.561	7.7
Fish products	1.2	1,309	91	7.0	1.01	0.313	26.1	0.022	1.8	5.7	0.125	1.7
Seaweed products	0.50	29	0	0.0	0	0.000	0.0	0.000	0.0	0.4	0.000	0.0
Soy sauce	0.50	701	255	36.4	0.52	0.211	42.2	0.077	15.4	20.7	1.594	21.8
Boiled down foods with soy sauce (Tsukudani)	0.50	1,091	19	1.7	0.76	0.142	28.3	0.002	0.5	3.0	0.006	0.1
Boiled beans	0.50	560	3	0.5	0.05	0.037	7.3	0.000	0.0	2.4	0.000	0.0
Fish paste products	0.30	3,829	217	5.7	0.5	0.105	34.9	0.006	2.0	13.0	0.078	1.1
Vinegar	0.30	40	2	5.0	0.06	0.052	17.3	0.003	0.9	2.5	0.008	0.1
Nonalcoholic beverages	0.30	1,341	103	7.7	1	0.159	52.9	0.012	4.1	162	1.943	27.0
Syrup	0.30	31	0	0.0	0	0.000	0.0	0.000	0.0	0.1	0.000	0.0
Sauce	0.30	259	29	11.2	0.46	0.192	64.0	0.021	7.2	3.0	0.063	0.9
Milk beverages	0.30	38	0	0.0	0	0.000	0.0	0.000	0.0	16.6	0.000	0.0

Lactic acid bacterial beverages	0.30	61	0	0.0	0	0.000	0.0	0.000	0.0	0.1	0.000	0.0
Ice candy	0.30	76	19	25.0	0.29	0.124	41.2	0.031	10.3	5.1	0.158	2.2
Ice cream	0.20	163	2	1.2	0.3	0.275	137.5	0.003	1.7	14.4	0.043	0.7
Sweetened bean paste (An)	0.20	260	0	0.0	0	0.000	0.0	0.000	0.0	7.8	0.000	0.0
Jam	0.20	76	0	0.0	0	0.000	0.0	0.000	0.0	1.1	0.000	0.0
Other pickled foods	0.20	604	87	14.4	1.5	0.497	248.3	0.072	35.8	8.9	0.641	8.7
Fermented milk	0.20	57	0	0.0	0	0.000	0.0	0.000	0.0	9.7	0.000	0.0
Flour paste	0.20	34	0	0.0	0	0.000	0.0	0.000	0.0	0.1	0.000	0.0
Soy bean paste (Miso)	0.20	393	10	2.5	0.5	0.145	72.3	0.004	1.8	15.7	0.063	0.8
Confectionery (Kashi)	0.10	1,988	13	0.7	1.15	0.510	510.0	0.003	3.3	49.3	0.148	2.3
Canned or bottled foods	0.20	333	4	1.2	0.08	0.035	17.6	0.000	0.2	9.9	0.000	0.1
Canned or bottled fish	0.20	20	0	0.0	0	0.000	0.0	0.000	0.0	0.1	0.000	0.0
Nonpermissible foods*2	—	2,203	20	0.9	0.92	0.232	—	0.002	—	402		
Total (or mean)	—	19,964	2,239	(11.2)	2.24	—	—	—	—	767.9		
*2Breakdown (detected only)												
Pre-cooked foods (Sozai)	—	395	5	1.3	0.92	0.448	—	0.006	—	20.7	0.124	1.6
Cheese	—	26	3	11.5	0.11	0.050	—	0.006	—	1.9	0.011	0.2
Daily intake of sodium saccharin											7.260	100.0

*1 Only chewing gum is allowed to contain saccharin, while other allowable foods can contain sodium saccharin.

*2 Nonpermissible foods and numbers of samples in which sodium saccharin was not detected were as follows.
Meat products 303, dried fruits 37, seasoning 20, alcoholic beverages 25, noodles 11, butter 1, health foods 25, margarine 3, rice cake 5, *etc.*

Table 7 Estimated Daily Intakes of BHA, BHT, Propylene Glycol, and Sodium Saccharin, and Ratios to ADI

Food additives	Daily intake (mg/person) (1994)	Ratio to ADI (%)	Daily intake in references (mg/person) (The year of investigation)				ADI (mg/person) as 50 kg body weight
			Japanese group[5]		UK group[6]	Finnish group[7]	
			(1994)	(1987)	(1984–1986)	(1980)	
BHA	0.169	0.7	0.002	0.007	0.4	0.17*1	25.0
BHT	0.051	0.3	0.066	0.007	0.2		15
Propylene glycol	57.1	4.6	37.3	23.6	54.1	—	1,250
Sodium saccharin	7.26	2.9	0.416	1.107	12.2*2	5.9*2	250

*1 Total of BHA and BHT
*2 As saccharin.

food additives were also inspected, some of them were foods in which these food additives might be used, c) foods containing excessive amounts of food additives and nonpermissible foods containing these food additives would be excluded from markets by the local governments, d) the concentration of these food additives in samples where they were undetectable was regarded as 0 mg/kg. Factors a) and b) would most strongly affect the results. It is therefore more likely that the results obtained are overestimated rather than underestimated.

8. Estimation of daily intake of BHA, BHT, propylene glycol, and sodium saccharin per person

The daily intake of BHA, BHT, propylene glycol, and sodium saccharin has been estimated by means of the market basket method in Japan[3)-5)], based on replies to a questionnaire sent to food and food additive producers in the UK[6)], and based on analytical results by official laboratories in Finland[7)]. In the present study, we estimated daily intake based on the analytical results of the official inspection by local governments in fiscal year 1994. The data on the daily consumption of foods by the nation estimated by the Japanese group[3), 5)] and the Ministry of Health and Welfare[18)] were utilized to estimate the daily intakes of these food additives. The Finnish method[7)] and our method for the estimation were very similar.

Estimated amounts of daily intake of these food additives per person in the present study were as follows: BHA, 0.169 mg; BHT, 0.051 mg; propylene glycol, 57.1 mg; and sodium saccharin, 7.26 mg (Table 7). These results were closer to the results of European groups[6), 7)] rather than those of the Japanese group[5)]. The estimated daily intakes of BHA, BHT, propylene glycol and sodium saccharin in the present study were 0.7%, 0.3%, 4.6%, and 2.9% of ADI, respectively. Foods that contributed most to the daily intake were dried fish for BHA (70.2% of daily intake), chewing gum for BHT (61.6%), raw noodles for propylene glycol (67.3%), and nonalcoholic beverages for sodium saccharin (27.0%).

These results obtained in the present study may be over- or underestimated as described above and in the previous papers[8)-10)]. In addition to the points mentioned above, BHA in dried fish was mainly detected in "Niboshi" which is boiled and dried sardine. Since the daily consumption of "Niboshi" is 1.3 g, the estimated daily intake of BHA from dried fish may decrease to 0.085 mg. Further, a cooking or extracting factor should be considered because "Niboshi" is mainly used for brewing stock. Migration of BHT from chewing gum into saliva may affect both estimates of the daily intake and contribution rate, because about a half of BHT in chewing gum is known to remain during chewing for 30 min[21)]. In the present study, all BHT in chewing gum was assumed to be ingested. The daily intakes of these antioxidants and contribution rates of dried fish and chewing gum may therefore be considerably overestimated.

9. Discussion of the results of the present series on the concentrations and daily intake of 16 kinds of food additives

In the series of studies on "Concentration of

Food Additives in Foods and the Daily Intake in Japan"[8)-10)], the mean concentration in foods and daily intake per person were estimated for 5 preservatives[8)], namely benzoic acid, dehydroacetic acid, *p*-hydroxybenzoic acid, propionic acid, and sorbic acid; 4 antifungal agents[9)], namely diphenyl, imazalil, *o*-phenylphenol, and thiabendazole; 3 inorganic food additives[10)], namely nitrite, nitrate, and sulfur dioxide, and the food additives reported in the present paper, namely BHA, BHT, propylene glycol, and sodium saccharin. The total number of analytical results utilized in this series was 122,952, based on the official inspection in Japan in the fiscal year 1994. The total daily intake of these 16 kinds of food additives was 116.2 mg per person. Among these 16 food additives, the highest rate of daily intake to ADI was that of nitrite (10.6%). Meat products contributed most (55.0%) to the daily intake of nitrite. The UK group[6)] considered that the preferred daily intake of a food additive is less than 10% of ADI. Daily intakes of other food additives were 4.7% of ADI for sulfur dioxide, or less. The daily intakes of inorganic food additives estimated in this series include only a minor contribution from natural components. The highest daily intake per person was observed with propylene glycol (57.1 mg, corresponding to 4.6% of ADI).

Although the method used in this series is unable to estimate the overall intake of allowed food additives, as reported by the Japanese group (9.4 g per person[5), 22)]) or by the UK group (8.0 g[6)]), the advantage of the present method is that we can obtain the concentrations in each type of foods and daily intakes from analytical results of more than one hundred thousand food samples obtained by official inspections covering most of Japan.

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