Original Article

FLUORIDE ANALYSIS OF FOODS FOR INFANTS AND ESTIMATION OF DAILY FLUORIDE INTAKE

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Received 28 April, 2004/Accepted for Publication 10 May, 2004

Abstract

The mean daily fluoride intake in infants was estimated on the basis of their intake of commercial foods for infants in Japan and evaluated in order to establish the effectiveness and safety criteria for water fluoridation, which is practiced as a preventive measure for dental caries suitable in life stages from children to the elderly. Based upon the intakes of foods for infants, the mean daily fluoride intake was estimated to be 0.166 mg in infants aged 3–4 months, 0.202 mg in those aged 5–6 months, and 0.266 mg in those aged 7–8 months. The mean daily fluoride intake per kg of body weight at these ages was in the range of 0.023–0.029 mg/kg, which was about half of the standard daily fluoride intake for infants and children advocated by Ophaug *et al.*, as 0.05–0.07 mg/kg.

From our results, the daily fluoride intake of infants from foods in Japan is estimated to be equivalent to or lower than the values of previous reports in non-fluoridated areas. Consequently, our data support the argument that water fluoridation and the appropriate use of fluoride for dental caries prevention in Japan are needed on the basis of scientific criteria in terms of fluoride exposure related to food intake during tooth formation.

Key words: Fluoride application—Daily fluoride intake—Infant foods

INTRODUCTION

Numerous epidemiological studies have demonstrated that fluoride application has a marked preventive effect against dental caries in children. WHO approved fluoride as a preventive measure of dental caries in 1969¹⁷⁾ and recommended its appropriate use^{48,49)}. Moreover, water fluoridation as a measure for

the prevention of dental caries has recently been shown to be effective from childhood throughout the entire lifespan to old age^{11,17)}.

In planning fluoride applications for oral health, the daily fluoride intakes (DFIs)²⁾ from drinking water and food stuffs must be evaluated as basic data for human body as an index of routine fluoride intake. In Western countries, DFIs from foods and drinking water

This paper is based on a thesis submitted to the Graduate School, Tokyo Dental College, published in Journal of Dental Health (in Japanese) by T. Tomori.

are evaluated for many years and monitored regularly in terms of fluoride exposure, especially in considering the occurrence of dental fluorosis^{5,9,15,16,19-24,29,30,34,35,45)}.

The fluoride intake of the Japanese population has been investigated by several groups since the 1950's, but the studies have been focused primarily on adults^{13,32,33,36,43,44)}. Additionally, the measured values have been obtained by different analytical methods and are not appropriate for direct comparisons. Because imported foods and factory-processed foods are increasingly available, and the patterns of food intake and dietary habits have been changing, reevaluation of fluoride content in foods and daily fluoride intake is needed with current data⁵⁰⁾.

The standards for daily fluoride intake in infancy and early childhood are determined using indices of an increase in the resistance of teeth against caries due to pre-eruptive maturation of the enamel of permanent teeth and improvements in its crystalline properties as well as those of prevention of the occurrence of dental fluorosis caused by an excess fluoride intake, which is suspected to be due to the intake of high fluoride in drinking water and various fluorides^{2,22,30,31)}.

Based on these considerations, Recommended Dietary Allowances (RDAs)³⁷⁾ have been issued in the United States, and tolerable intakes of major nutritional elements, minerals, and trace elements are discussed therein. The tenth edition of RDAs (1989) lists intake levels of copper, manganese, fluorides, chromium, and molybdenum that are estimated as safe and adequate daily dietary intakes in addition to such major minerals as calcium, phosphorus, magnesium, iron, zinc, iodides, and selenium. Recent publications have urged that the fluoride intake should not substantially exceed the tolerable upper intake level (UL) of the appropriate range³⁷⁾ and have requested evaluation of the establishment of new criteria^{17,50)}.

Therefore, we believe that the fluoride intake from food is a factor closely related to the use of fluoride for prevention of dental caries and that estimation of the daily fluoride

intake in infants from foods for infants is therefore needed data for planning a prevention program. Early childhood is the most important period for monitoring the appropriate use of fluoride in Japan.

MATERIALS AND METHODS

1. Foods for infants

In this study, commercial powdered milk and baby foods were analyzed as samples of foods for infants. Ten powdered milk products, 2 products each from 5 manufacturers, were analyzed. As baby foods, 48 products in Groups I (grains), II (meat, fish, and dairy), III (vegetables), and IV (fruits and juices) were selected as samples according to Imamura's classification of baby foods¹⁴⁾. They consisted of 11 grain products (5 freeze-dried and 6 retort-packed foods), 20 meat, fish or dairy products (13 freeze-dried and 7 retort-packed foods), 11 vegetable products (5 freeze-dried and 6 retort-packed foods), and juice or fruit 6 products. Food collections were carried out from 1994 to 1996. The same lot numbers of 3 to 5 commercial powdered milk and baby foods were collected. Before analysis, each sample (same lot) was mixed as follows: the freeze-dried foods were pulverized in a cartridge mill (Ikeda Kagaku, Tokyo), and the retort-packed foods containing water were homogenized in a mortar.

2. Experimental apparatuses, reagents, and solutions

1) Micro-diffusion apparatus

Fluoride was isolated from the food samples by using the micro-diffusion apparatus of Hinoide *et al.*¹⁰⁾. This micro-diffusion apparatus is made of Teflon and is heat- and chemical-proof. The apparatus is air-tight with a screw-top lid. A thin Teflon vessel (capacity 1.5 ml) is placed in the internal chamber of the diffuser. By placing the sample and isolation-diffusion fluid in the outer chamber and the fluoride capturing fluid in the vessel of the inner chamber, the isolation, diffusion, and capturing of the fluorides from the sample

Fig. 1 Fluoride analysis for infant foods

are performed in a closed system.

2) Apparatus for fluoride ion assay

A combination fluoride ion electrode (96-09 BN, Orion Research, Ltd., Boston, USA) with an ion analyzer (EA940, Orion Research, Ltd., Boston, USA) was used to determine the fluoride concentration in the trapping solution.

3) Reagents and solutions

(1) Isolation-diffusion solution

The isolation-diffusion solution for fluorides from the foods was an HMDS/saturated 5 M perchloric acid prepared by diluting a 60% perchloric acid (for precise analysis, Wako Pure Chemical Industries) with distilled water to 5 M and mixing 200 ml of this 5 M perchloric acid with 10 ml of hexamethyldisiloxane (HMDS, Shinetsu Kagaku, Tokyo) according to Taves^{38,39}.

(2) Fluoride capturing solution

The isolated and diffused fluorides were captured with 1 ml of 0.1 M NaOH solution.

(3) Standard solutions of fluoride ion concentration

Standard solution of fluoride (1,000 ppmF) was prepared by dissolving 221 mg of NaF (Special grade, Wako Chemical Industries, Ltd.) in distilled water and diluting to 100 ml. Standard samples of NaF solution containing 0.1, 1.0, 10, and 100 µg/ml of fluoride ions were used as references for the samples.

3. Analytical procedures for fluorides in foods by the micro-diffusion method

The sample was weighed (0.5 g of a freeze-dried food, 2 g of retort-packed food) and placed in the outer chamber of the micro-diffusion apparatus. Then the capturing vessel was placed in the inner chamber of the micro-diffusion apparatus, and 1 ml of the fluoride capturing fluid (0.1 M NaOH) was dripped in. After 4 ml of the isolation-diffusion solution (HMDS/saturated 5 M perchloric acid solution) was poured into the outer chamber, the lid was immediately screwed on. The apparatus was held horizontally and gently rotated to allow the isolation-diffusion solution to permeate evenly into the sample. The contents were allowed to react for a predetermined period at 60°C in an incubator (DS-43, Yamato Kagaku, Tokyo). After the reaction, the contents were removed from the incubator and allowed to cool at room temperature. The capturing vessel was removed from the micro-diffusion apparatus, and the weight loss was complemented with distilled water. To the capturing solution, 0.1 ml of TISAB III (Total Ionic Strength Adjusted Buffer III, Orion Research, Ltd., Boston, USA) was added, and fluoride ion was assayed by the fluoride ion electrode method. Blank assays were performed by two diffusion vessels containing the isolation-diffusion fluid without a sample in the outer chamber. At each assay, a blank value was obtained as the mean fluoride ion concentration of the above two diffusion

Table 1 Fluoride recovery by HMDS-microdiffusion method

F added (μg)	F determined (μg)	F recovery (%)	C.V. (%)
0.2	$0.19 \pm 0.003^*$	$95.0 \pm 1.5^*$	1.6
2.0	1.96 ± 0.068	98.2 ± 3.4	3.5

*: mean $\pm$ S.D., n = 5Table 2 Effect of time on fluoride diffusion in baby foods ($\mu\text{g/g}$)

Baby foods	Sample (g)	Diffusion condition		
		60°C•2 hr	60°C•6 hr	60°C•12 hr
Rice gruel*	2	0.53 (0.01)	0.72 (0.04)	0.82 (0.04)
Beef with vegetable**	0.5	0.54 (0.06)	0.57 (0.04)	0.88 (0.14)
Rice gruel**	0.5	0.14 (0.01)	0.18 (0.02)	0.21 (0.01)
Rice gruel with young sardine**	0.5	2.67 (0.10)	2.81 (0.21)	2.91 (0.06)

*: Freeze-dried food, **: Retort food, mean $\pm$ S.D., n = 3

Table 3 Comparison of F concentration of infant foods by distillation method (ashed) and microdiffusion method (unashed)

Infant food		Unashed	Ashed	Unashed/ashed $\times$ 100 (%)
		Microdiffusion method	Steam distillation method	
Rice gruel with young sardine	E	2.91 ± 0.06	3.21 ± 0.24	90.6
Rice gruel	F	5.22 ± 0.07	5.38 ± 0.13	97.0

mean $\pm$ S.D., n = 5

vessels, and assays were repeated three times with each sample (Fig. 1).

4. Fluoride analysis by the distillation method

Each food sample was weighed (4–5 g) and placed in a porcelain crucible. Five ml of CaOH solution was added as a fluoride fixing agent, and the contents were dried at 80°C and incinerated at 550°C for at least 12 hours. The distillation was performed using 100 ml of 60% perchloric acid as the wet incineration solution at a distillation temperature of $140 \pm 2^\circ\text{C}$ and distillation rate of 30 ml/10 min. Distillation was ended when 200 ml of distillate had been obtained²⁶⁾. Fluorides in the distillate were assayed by the fluoride ion electrode method.

5. Analysis of Ca, Mg, and P in foods for infants

For analysis of Ca, Mg, and P in foods for infants, the sample (0.1 g of freeze-dried food, 1 g of retort-packed food) was wet-incinerated with 1 ml of 70% HNO₃ and 0.5 ml of 60% HClO₄. Ca and Mg were then assayed by atomic absorption spectrophotometry (508, Hitachi), and P by the spectrophotometric method of Chen *et al.*³⁾ (MPS-2000, Shimadzu).

6. Calculation of mean daily fluoride intake in infants

The freeze-dried foods for infants were prescribed to be prepared with drinking water at 5 times the volume of the dried food. As the fluoride ion concentration in tap water in Japan is less than 0.2 ppm, the test

foods were prepared by assuming the fluoride ion concentration in drinking water was 0.1 ppm. Because the retort-packed foods are eaten directly from the package, the assay results were regarded as their fluoride concentrations.

The age specific body weights of infants, total energy intakes, and intakes of various foods were calculated on the basis of the data of Nishimura *et al.*²⁸⁾. The age specific mean daily intakes of various food groups were calculated by dividing the total energy (kcal) of each food group and the mean energy of each food group for infants (kcal/g). Therefore, the mean daily fluoride intake at a given age was calculated as follows;

The mean daily fluoride intake at a given age of the month (mg) = Σ mean fluoride concentrations in various food groups at the time of intake $\times$ intake of various food groups.

The mean daily fluoride intake per kg of body weight was calculated by dividing the mean daily fluoride intake obtained above by the body weight at the age in months²⁸⁾.

RESULTS

1. Fluoride analysis of foods for infants by the micro-diffusion method: Evaluation of diffusion time and recovery rate

The fluoride ion standard addition method by micro-diffusion (60°C, 12-hour) was performed by repeating the assay procedure 5 times each at fluoride ion concentrations of 0.2 and 2.0 μ g. The fluoride ion addition-recovery rate (%) was calculated as the measured value of fluoride ion concentration – blank value/quantity of fluoride ion added $\times 100$. The recovery rates at the added quantities of fluoride ion of 0.2 μ g and 2.0 μ g were $95.0 \pm 1.5\%$ and $98.2 \pm 3.4\%$, respectively (Table 1). The blank value obtained in five repeated assays was $0.026 \pm 0.008 \mu$ g, a trace level.

Before analyzing fluorides in foods for infants by a combination of the micro-diffusion method and the fluoride ion electrode

method, the diffusion time was evaluated using samples of 4 food items (Table 2). The diffusion temperature was fixed at 60°C, and the assays were repeated 3 times, except in the case of “porridge containing dried young sardines”, which was assayed 5 times. The measured fluoride concentrations in “porridge containing dried young sardines” were 2.67 ± 0.10 , 2.81 ± 0.21 , and $2.91 \pm 0.06 \mu$ g/kg after diffusion for 2, 6, and 12 hours, respectively, showing increases with the diffusion time. The value after the 2 hours diffusion was greater than 90% of the value after the 12 hours diffusion. The measured values of fluoride after 12 hours diffusion were $0.21 \pm 0.01 \mu$ g/g in “rice porridge”, $0.82 \pm 0.04 \mu$ g/g in “porridge”, and $0.88 \pm 0.14 \mu$ g/g in “beef and vegetable”. The recovery rate after 6 hours of diffusion compared with that after 12 hours diffusion was 64.7% in “beef and vegetables” but 85% or higher in the other foods.

The fluoride concentrations in 2 baby foods were compared by the micro-diffusion at 60°C, 12 hour-fluoride ion electrode method and the incineration-vapor distillation-fluoride ion electrode method. The recovery rates of fluorides on 5 repeated assays were 90.6% for “porridge containing dried young sardines” and 97.0% for “rice porridge” (Table 3).

The recovery rate of fluorides was 90% or above and the blank value was very low with the micro-diffusion method, so that the fluoride assay was shown to be possible even in a small quantity of a sample containing a low level of fluoride by this method. Therefore, a combination of the micro-diffusion method (60°C, 12-hour or longer)-fluoride ion electrode method was selected as the method for fluoride analysis of foods for infants.

2. Fluoride concentrations in processed powdered milk

The fluoride concentrations in 10 commercial processed powdered milk products were 0.30–1.00 μ g/g (mean 0.50 μ g/g), and, when they were prepared according to the manufacturers’ instructions with distilled water (13–14 g/100 ml), the fluoride concentrations

Table 4 F, Ca, Mg, and P concentration in powdered milk

No.	Powdered milk		F	Ca	Mg	P
	Trade name	Manufacturer	μg/g			
1	F & P	A	0.84	4,090	472	1,900
2	Step	A	1.00	6,520	625	2,940
3	Neomilk Ai	B	0.43	3,950	465	1,910
4	Neomilk Tyuyoiko origo	B	0.48	5,240	502	3,110
5	LF Tirumiru	C	0.52	3,240	480	1,800
6	New-LF Tirumiru	C	0.62	3,210	494	2,830
7	SMA S-26	D	0.37	3,490	461	2,240
8	SMA follow 6	D	0.50	6,590	703	3,990
9	Lebense follow	E	0.30	3,530	391	2,110
10	Lebense f	E	0.35	3,500	395	3,300

at the time of intake by infants were 0.04–0.12 $\mu\text{g/ml}$. The processed powdered milk manufactured by A (No. 2), in which the fluoride concentration was the highest (1.0 $\mu\text{g/g}$) among the samples examined in this study, characteristically had a slightly higher calcium content (6,520 $\mu\text{g/g}$) compared with the other products (Table 4).

3. Fluoride concentrations in foods for infants

Table 5 shows the concentrations of F, Ca, Mg, and P contained in “porridge”, which is classified in the grain group among baby foods. The fluoride concentration was high in “rice porridge” (No. 6, 5.22 $\mu\text{g/g}$) and in two of the products labeled “porridge containing dried young sardines” (No. 2, 2.91 $\mu\text{g/g}$; No. 7, 2.35 $\mu\text{g/g}$) among freeze-dried grains. In “rice porridge” (No. 6), which showed the highest fluoride concentration, the Ca (6,990 $\mu\text{g/g}$) and P (9,150 $\mu\text{g/g}$) concentrations were markedly higher than in the other foods. The fluoride concentration was 0.6 $\mu\text{g/g}$ or less in the other grain foods. Among retort-packed grain foods, the fluoride concentration was high at 0.44 $\mu\text{g/g}$ in “porridge containing dried young sardines and vegetables” but was very low at 0.07 $\mu\text{g/g}$ or below in other grain foods not containing dried young sardines. Among retort-packed foods, the fluoride concentration tended to be low in the products

with a low Ca concentration or not containing seafood such as dried young sardines.

Group II includes infant foods in which protein is the primary component; meat, fish, and dairy products are classified into this group (Table 6). The fluoride concentrations in freeze-dried products were in the range of 0.19–0.88 $\mu\text{g/g}$, and no high-fluoride food was included in this group, but the Ca content (405–15,900 $\mu\text{g/g}$) was not necessarily low, and it was considerably higher in some products (No. 5, 12) than in the grain products. Among retort-packed products, the fluoride concentration was highest in “beef” at 0.18 $\mu\text{g/g}$ but was generally low at 0.02–0.08 $\mu\text{g/g}$ in the other products.

Table 7 shows the F, Ca, Mg, and P concentrations in the baby foods classified as Group III, in which vegetables were the primary components. The fluoride concentrations in freeze-dried vegetables were 0.22–0.62 $\mu\text{g/g}$ ($n = 5$). Many of the retort-packed vegetables were characteristically made in the USA, and their fluoride concentrations were 0.04–0.56 $\mu\text{g/g}$ ($n = 6$). However, the fluoride concentration in “mixed vegetables” made in the USA (0.56 $\mu\text{g/g}$) was about ten times higher than in “mixed vegetables” of the same maker made in Japan (0.05 $\mu\text{g/g}$).

Among the commercial juices and fruits, the fluoride concentrations in fruits were low at 0.03–0.05 $\mu\text{g/g}$ ($n = 3$), and those in juices

Table 5 F, Ca, Mg, and P concentration in gruel available

No.	Gruel		F	Ca	Mg	P
	Trade name	Manufacturer	μg/g			
1	Rice gruel*	E	0.21	92	155	884
2	Rice gruel with young sardine*	E	2.91	1,300	406	2,070
3	Rice gruel*	A	0.50	1,490	221	1,210
4	Rice gruel boiled with miso*	A	0.60	4,520	334	1,350
5	Cereal*	A	0.22	227	817	2,420
6	Rice gruel*	F	5.22	6,990	554	9,150
7	Rice gruel with young sardine*	F	2.35	937	469	2,980
8	Pilaf with shrimp*	F	0.45	1,050	326	3,570
9	Rice gruel with liver**	F	0.07	181	47	866
10	Rice gruel with egg**	F	0.06	87	36	712
11	Rice gruel with sardine**	F	0.44	147	50	466

*: Freeze-dried food, **: Retort food

Table 6 F, Ca, Mg, and P concentration in baby food (meat and fish)

No.	Meat and fish		F	Ca	Mg	P
	Trade name	Manufacturer	μg/g			
1	Beef with vegetable*	A	0.88	4,460	650	2,430
2	Liver with vegetable*	A	0.54	4,790	909	3,150
3	Fish with vegetable*	A	0.60	4,750	639	6,710
4	Beef stew*	E	0.54	405	550	1,700
5	Beef stew*	F	0.19	8,970	710	5,060
6	Tuna stew*	E	0.29	1,410	413	2,480
7	Samon doria*	E	0.37	1,860	326	2,370
8	Chicken stew*	E	0.19	1,490	446	3,200
9	Chicken risotto*	F	0.34	310	539	5,700
10	Macaroni gratan*	E	0.54	2,480	465	2,780
11	Samon stew*	F	0.27	1,900	715	7,700
12	Flat fish & spinach stew*	F	0.26	15,900	1,370	8,030
13	Liver with vegetable*	E	0.75	1,090	610	5,050
14	Beef**	F	0.18	43	126	2,240
15	Pork (ham)**	F	0.02	36	165	3,040
16	Cheese doria**	F	0.07	301	43	656
17	Beef & liver product**	F	0.05	66	88	2,300
18	Chicken product**	F	0.08	547	139	2,220
19	Beef product**	F	0.06	43	83	902
20	Fruits yogurt**	F	0.07	499	75	954

*: Freeze-dried food, **: Retort food

Table 7 F, Ca, Mg, and P concentration in baby food (vegetable group)

No.	Vegetable		F	Ca	Mg	P
	Trade name	Manufacturer	$\mu\text{g/g}$			
1	Pumpkin*	E	0.42	1,150	302	1,826
2	Vegetable mix*	E	0.22	443	616	1,400
3	Vegetable mash*	A	0.55	465	671	1,300
4	Vegetable soup*	E	0.62	642	233	703
5	Spinash & green pea*	E	0.33	1,940	853	1,850
6	Carot (U.S.A.)**	F	0.11	130	133	386
7	Green pea (U.S.A.)**	F	0.47	122	103	658
8	Pumpkin (U.S.A.)**	F	0.04	270	133	346
9	Spinash (U.S.A.)**	F	0.12	993	384	1,280
10	Mix vegetable (U.S.A.)**	F	0.56	138	89	666
11	Mix vegetable (JPN)**	F	0.05	108	112	746

*: Freeze-dried food, **: Retort food

Table 8 F concentration in fruit and juice for infant

No.	Fruits and Juice	Manufacturer	F ($\mu\text{g/g}$)
1	Peach*	F	0.04
2	Apple*	F	0.05
3	Apricot*	F	0.03
4	Apple juice**	F	0.14
5	Apple & fruits juice**	F	0.18
6	Mix fruits juice**	F	0.15

*: Retort food **: Fresh

were 0.14–0.18 $\mu\text{g/g}$ ($n = 3$) (Table 8).

4. Fluoride concentrations in foods for infants according to food groups

Table 9 shows the fluoride concentrations in various food groups at intake calculated on the assumption that the fluoride concentration in drinking water is 0.1 ppm. The fluoride concentration and energy in freeze-dried foods at intake were calculated by assuming that they were prepared with five volumes of drinking water.

The fluoride concentrations in processed powdered milk at intake were 0.14–0.2 $\mu\text{g/g}$ (mean 0.17 $\mu\text{g/g}$). The fluoride concentrations at intake were 0.06–0.88 $\mu\text{g/g}$ (mean 0.30 $\mu\text{g/g}$) in grains and 0.02–0.23 $\mu\text{g/g}$ (mean 0.13 $\mu\text{g/g}$) in meat and fish. It was 0.04–

0.60 $\mu\text{g/g}$ (mean 0.23 $\mu\text{g/g}$) in vegetables and 0.03–0.18 $\mu\text{g/g}$ (mean 0.10 $\mu\text{g/g}$) in juices and fruits. Among the food groups, the fluoride concentration was highest in grains (mean 0.30 $\mu\text{g/g}$).

5. Estimation of mean daily fluoride intake based on the intakes of commercial foods for infants

The mean daily fluoride intakes were estimated from the fluoride concentrations in various food groups by calculating the intakes of foods from the age and body weight of the infants and the energy intakes from various food groups. The age specific mean daily fluoride intakes were estimated from the intakes of various food groups and their fluoride concentrations (Table 10). The mean

Table 9 Fluoride concentration in infant foods ($\mu\text{g/g}$)*

	Infant food	Ready-to-feed (F $\mu\text{g/g}$)			Energy kcal/g
		Range	Mean	N	
	Infant formula	0.14–0.22	0.17	10	0.67
I	Cereal	0.06–0.88	0.30	11	0.62
II	Meat & fish	0.02–0.23	0.13	20	0.76
III	Vegetable	0.04–0.60	0.23	11	0.53
IV	Fruits & juice	0.03–0.18	0.10	6	0.47

*: 0.1 ppm fluoride drinking water

Table 10 Average daily fluoride intake based on intake of baby foods*

Age month	Foods	Energy kcal	Intake g	Average of fluoride intake		
				mg/day	%	mg/kg body wt.
3–4 months	Powdered milk	578	863	0.147	88.0	
7.31 kg	Gruel	13	21	0.006		
	Meat & fish	12	16	0.003	6.6	
	Vegetable	2	4	0.001		
	Juice	43	91	0.009	5.4	
	Total	647		0.166	100	0.023
5–6 months	Powdered milk	481	718	0.122	55.4	
8.38 kg	Gruel	109	178	0.053		
	Meat & fish	44	58	0.007	32.2	
	Vegetable	12	23	0.005		
	Juice	33	70	0.010	7.4	
	Water	...	50	0.005		
	Total	678		0.202	100	0.024
7–8 months	Powdered milk	467	697	0.118	44.4	
9.17 kg	Gruel	210	339	0.102		
	Meat & fish	166	218	0.028	51.1	
	Vegetable	15	28	0.006		
	Juice	24	51	0.005	4.5	
	Water	...	70	0.007		
	Total	882		0.266	100	0.029

*: 0.1 ppm fluoride drinking water

daily fluoride intake of infants was estimated to be 0.166 mg/day at the age of 3–4 months, 0.202 mg/day at 5–6 months, and 0.266 mg/day at 7–8 months. Of the mean daily fluoride intake at the age of 3–4 months, 88% was accounted for by processed powdered milk,

but the percentage of fluoride intake from processed powdered milk decreased gradually with the advancing age; it was reduced to 44.4% at the age of 7–8 months. The intakes of baby foods increased as babies got older; these increases were mostly due to the

increases in grains, which accounted for 51.1% of the fluoride intake at the age of 7–8 months, exceeding the 44.4% from processed powdered milk.

The fluoride intake per kg of body weight was estimated to be 0.023 mg/kg at the age of 3–4 months, 0.024 mg/kg at 5–6 months, and 0.029 mg/kg at 7–8 months.

DISCUSSION

The effectiveness and safety of fluoride application for the prevention of dental caries have been evaluated for nearly 50 years^{2,11,49}. WHO issued recommendations for the use of fluorides for caries prevention in 1969⁴⁷, 1970⁴⁸, and 1994⁴⁹, and about 360 million people in 60 countries are using fluoridated water today¹. However, as the use of fluorides has become routine, confirmation of the standards for daily fluoride intake through each life-stage has become a global issue¹⁹. The Food and Nutrition Board Commission on Life Sciences of the National Research Council announced Recommended Dietary Allowances (RDAs) of vitamins A, D, E, K, and B's as well as Ca, P, Mg, Fe, Zn, I, and Se. Fluorides, the contents of which in the adult body are the next highest only to Fe, are mentioned in the Estimated Safe and Adequate Daily Dietary Intakes of Selected Vitamins and Minerals³⁷. In Japan, the Resources Council of the Science and Technology Agency has published "Standard Tables of Food Composition in Japan" in cooperation with related administrative organizations, but the daily intake of fluorides is not mentioned even in the 6th edition of the Nutritional Requirements⁸. In its latest revision, fluorides appear to have been evaluated mainly at the Community Health, Health Promotion, and Nutrition Division of the Health Service Bureau, Ministry of Health and Welfare.

Today, fluoride application is evaluated as a lifetime means for caries prevention, and estimation of the daily fluoride intake is indispensable and fundamental assessment required to assure its effectiveness and safety.

Fluorine as an essential trace element for the body has an important role in promoting the generation and maturation of apatite crystals in the odontogenic period, and there is a vast literature on this issue. Fluorides make major contributions not only to dental apatite crystal maturation but also to the stability of bone apatite crystals, and, as their usefulness throughout the human lifetime is being clarified, the establishment of adequate intake (AI) standards for fluoride^{8,50} is a crucial task in life science.

There have been several reports of the analysis and estimation of fluoride contents of foods in Japan since the 1950's^{13,32,33,36,43,44}. Samejima³³ estimated the daily fluoride intake from drinking water and foods in 12-year-old children to be 1.3–2.2 mg (fluoride concentration in drinking water: 0.05–0.2 ppm) from the menu. Saito³² reported the mean daily fluoride intake from the daily diet in Japanese adult males to be 1.52–2.1 mg. Iizuka¹³ estimated the daily fluoride intake in adults at 0.48–2.64 mg. Tsunoda *et al.*⁴⁴ calculated the daily fluoride intake in an average Japanese to be 1.01 mg on the basis of the national mean intakes of various foods according to the Nutritional Survey of Japan. Tomomatsu *et al.*⁴³ reported that the mean fluoride intake in adults calculated by the market basket method was 1.75 mg. However, with improvements in the analytical method for fluorides in foods as well as the increased diversity of foods and changes in the dietary environment, reevaluation of dietary fluoride analysis and daily fluoride intake has become necessary.

The daily fluoride intake from foods in infancy and early childhood is considered to be an important indicator for setting fluoride levels appropriately for both promotion of pre-eruptive maturation of the enamel in the formation of teeth and prevention of dental fluorosis, which is suspected to be caused by excessive fluoride intake from highly fluoridated drinking water and other sources. For this reason, we evaluated the daily fluoride intake in infants by analyzing the fluoride content in foods for infants. The daily fluoride intake from foods for

infants and children has been compared with literature reported internationally in Japan, Brazil, and Thailand by Nishijima *et al.*²⁷⁾ and Chitaisong *et al.*⁴⁾.

In examining methods for fluoride analysis of foods, Taves⁴⁰⁾ compared the results of the micro-diffusion method with incinerated and non-incinerated samples and reported no difference. In this study, we also employed fluoride analysis by the HMDS/micro-diffusion method/fluoride ion electrode method without incineration and ascertained its validity.

Ten commercial processed powdered milk products available in Japan, 2 each from 5 domestic makers, and 48 items of baby foods were analyzed. In this study, the fluoride concentrations in the 10 processed powdered milk products were 0.30–1.00 $\mu\text{g/g}$ (mean 0.50 $\mu\text{g/g}$). In Japan, Watanabe *et al.*⁴⁶⁾ and Tomita *et al.*⁴²⁾ previously reported the fluoride concentrations in powdered milk. According to Watanabe *et al.*⁴⁶⁾, the total fluoride concentrations in powdered milk of 5 domestic manufacturers analyzed by the incineration/micro-diffusion/fluoride ion electrode method were 0.22–0.40 $\mu\text{g/g}$. Tomita *et al.*⁴²⁾ selected twenty powdered milk products (6 were soybean-based) from 5 domestic manufacturers and determined their fluoride concentrations by (1) detection of fluoride ion by gas chromatography, (2) diffusion method, (3) pyrohydrolysis method, and (4) direct measurement by the ion electrode method, showing that there was no significant difference in the fluoride concentration between methods (2) and (3). They suggested that the diffusion method is acceptable for the measurement of the total fluoride concentration. They also reported that the total fluoride concentrations in milk prepared with distilled water were 0.033–0.210 $\mu\text{g/ml}$ ($n = 20$).

In other countries, Howat *et al.*¹²⁾ compared the fluoride concentrations in 6 processed powdered milk products after preparation with deionized water, and reported that they fell in the range of 0.02–0.08 $\mu\text{g/ml}$. McKnight-Hanes *et al.*²⁵⁾ similarly reported that the fluoride concentrations in processed powdered

milk prepared with deionized water were 0.055–0.121 $\mu\text{g/ml}$. Dabeka *et al.*⁶⁾ showed that the fluoride concentrations were 0.11–0.42 $\mu\text{g/g}$ in defatted powdered milk, but Singer *et al.*³⁵⁾ reported that the fluoride concentrations in prepared processed powdered milk were 0.08–0.31 $\mu\text{g/ml}$ in an area where tap water was not fluoridated (0.3 ppmF or below).

In this study, the fluoride concentrations in processed powdered milk prepared with distilled water were 0.04–0.12 $\mu\text{g/ml}$ (mean 0.07 $\mu\text{g/ml}$), which were within the ranges reported by Watanabe *et al.*⁴⁶⁾ and Tomita *et al.*⁴²⁾ and were close to the values reported by Howat *et al.*¹²⁾ and McKnight-Hanes *et al.*²⁵⁾. From the results of fluoride analysis of processed powdered milk in this study, the fluoride concentrations of prepared powdered milk drunk by infants are considered to be generally 0.1–0.2 $\mu\text{g/ml}$ in Japan, where tap water is not fluoridated, and they are estimated to be 0.3 $\mu\text{g/ml}$ or less all over Japan.

Concerning cow's milk, Hattab *et al.*⁷⁾ reported the fluoride concentrations in imported products, with the exception of those made in Hong Kong, to be 0.01–0.07 $\mu\text{g/ml}$ ($n = 7$). Their usual concentrations are also reported to be 0.1 $\mu\text{g/ml}$ or less^{16,41)}. In earlier reports in Japan^{13,32,33,43,44)}, the fluoride concentrations were 0.14–0.22 $\mu\text{g/ml}$, but Soejima³⁶⁾ reported a high concentration of 1.52 $\mu\text{g/ml}$. According to our preliminary investigation, the fluoride concentrations in commercial cow's milk were 0.04–0.06 $\mu\text{g/ml}$ ($n = 4$). Although direct comparison is impossible because of the differences in the samples, the 1.52 $\mu\text{g/ml}$ reported by Soejima³⁶⁾ is extremely high for common cow's milk. If this value is directly used for estimation of the daily fluoride intake in 3-year-old children, it is likely to be overestimated.

Fluoride analysis was performed in 48 baby foods from 3 manufacturers. The "rice porridge" that showed a high fluoride concentration (5.22 $\mu\text{g/g}$) among freeze-dried foods was a product made in the USA, and its Ca concentration (6,990 $\mu\text{g/g}$) and P con-

centration ($9,150\mu\text{g/g}$) were also markedly higher than in the other products, probably because of the addition of calcium phosphate. The fluoride concentration was higher in the "porridge containing dried young sardines" from two companies ($2.91\mu\text{g/g}$ and $2.35\mu\text{g/g}$) than in plain "porridge" due to the addition of "dried young sardine". In the Japanese diet, small fish such as "dried young sardines" are recommended for children in the growth period from infancy to school age as a source of calcium, but they also contribute to fluoride intake.

The mean daily fluoride intake in Japanese infants was reported by Nishijima *et al.*²⁷⁾ and Chitaisong *et al.*⁴⁾ to be $0.087\text{--}0.238\text{ mg/day}$ at the age of six months and was suggested by Kimura *et al.*¹⁸⁾ to be $0.10\text{--}0.57\text{ mg/day}$ (mean 0.23 mg/day) in one-year-old children by dietary fluoride analysis by the Duplicate-portion sampling method. According to the data in our study, the daily fluoride intake at the age of 5–8 months was $0.202\text{--}0.266\text{ mg/day}$, which is similar to the values in the above reports. These values were also similar to or lower than the values reported overseas¹⁹⁾.

Issues raised by this study were: (1) Clarification of the standard fluoride concentrations in mother's milk and home-cooked baby foods, (2) clarification of the fluoride intakes in wide regional blocks, mother's milk, and processed milk based on the food intakes of Japanese infants, and (3) comparison of the food intake pattern between children aged 0–3 years and those aged 4 years and above in relation to the standard nutritional requirements. Concerning problem (2), in particular, there are data concerning the daily intakes of various foods from infants to adults in Western countries, where the daily intakes of nutritional elements are well documented. In Japan, nationwide investigations using a unified format are strongly required.

CONCLUSIONS

This study was designed to provide data useful for the assessment of fluoride appli-

cation for dental caries prevention at each life-stage and was carried out with particular emphasis on analysis of the daily fluoride intake of infants and evaluation of the measured values. Fluoride analysis was performed on commercial baby foods, the measured values were evaluated, and the daily fluoride intakes of infants at various ages were estimated. The results were as follows.

1. Fluoride analysis of baby foods was performed using a Teflon micro-diffusion apparatus without incineration of the samples under the diffusion conditions of 60°C and 12-hour with HMDS-saturated 5 M perchloric acid solution. In this study, the recovery rates of fluorides from the samples were 90.6% or above. This fluoride assay method, developed and evaluated at our department, allows rapid and simple analysis of low-concentration fluorides in small quantities of samples.
2. The fluoride content in 10 processed powdered milk products commercially available in Japan analyzed in this study were $0.30\text{--}1.00\mu\text{g/g}$ (mean $0.50\mu\text{g/g}$). With these fluoride contents, when the fluoride ion concentration in routinely used tap water is assumed to be $0.1\mu\text{g/ml}$, the fluoride concentration in the milk drunk by infants is estimated to be $0.14\text{--}0.22\mu\text{g/g}$ (mean $0.17\mu\text{g/g}$).
3. The mean fluoride contents of commercial foods for infants at the intake as baby foods were $0.30\mu\text{g/g}$ in grains, $0.13\mu\text{g/g}$ in meat, fish, and dairy, $0.23\mu\text{g/g}$ in vegetables, and $0.10\mu\text{g/g}$ in juices (fruits). Among the baby foods, the fluoride concentration was high in "rice porridge" made in the USA to which calcium phosphate was added ($5.22\mu\text{g/g}$) and commercial "porridge containing dried young sardines" ($2.91\mu\text{g/g}$), which is prepared by adding small fish to porridge.
4. On the basis of the food intake of infants, the mean daily fluoride intake was estimated to be 0.166 mg in infants aged 3–4 months, 0.202 mg in those aged 5–6 months, and 0.266 mg in those aged 7–8 months. The mean daily fluoride intake

per kg of body weight at these ages was 0.023–0.029 mg/kg, which is considerably lower than 0.05–0.07 mg/kg, the allowance advocated by Ophaug *et al.*³⁰⁾.

This study provided basic data for estimated daily fluoride intake based on the results of fluoride analysis in foods for infants in Japan. These data are considered to be useful as part of the reference data for promoting water fluoridation in Japan.

ACKNOWLEDGEMENTS

The authors extend their sincere gratitude to the members of the Department of Epidemiology and Public Health, Tokyo Dental College, for kind advice.

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