Dietary sources of fluoride for infants and children in Hong Kong

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Abstract

The purpose of this study was to investigate the dietary sources of fluoride (F) for infants and children residing in Hong Kong. Analyses were made on commercially available foods and beverages as well as hospital infant diets. Food items from different countries of origin also were analyzed in order to assess the variation in sources of F for children living in different communities.

The findings indicate that whole cow's milk contained as low as 0.01 ppm F. The F content in powdered infant formulas (undiluted) varied from 0.06 to 1.08 ppm, with an average of 0.39 ppm. Hospital infant foods were found to range from 0.27 to 0.93 ppm F with an average of 0.59 ppm. Relatively high sources of F were found in soups prepared from seafoods. Mineral/spring water, in general, contained low F concentrations of 0.01-0.10 ppm and therefore does not significantly contribute to F intake if used in reconstitution of milk powderor concentrated milk formulas. The average F contents in canned soft drinks and fruit-flavored drinks were 0.42 and 0.56 ppm, respectively, while for prepackaged fruit juices, they averaged 0.65 ppm F. The average F intake of 6- to 11month-old infants from whole milk, infant formula and beverages was estimated to be 0.23 mg/day or 0.027 mg/kg of body weight which is about half of the optimum F intake of 0.05-0.07 mg/kg of body weight.

Recent studies have shown that the decline in the prevalence of dental caries has occurred not only in optimally fluoridated communities where the drinking water contains fluoride (F) concentrations of 0.7-1.2 ppm, but also in F-deficient communities.¹ It has been suggested that the general decline in dental caries prevalence may be due to an increase in the use of F dentifrices and mouthrinses, F supplements, and foods and beverages processed with fluoridated water² and other reasons. Because of the probable changes in die-

tary patterns in recent years and the current multiple sources of F ingestion, it is important to assess the daily total F intake of different age groups of children residing in fluoridated and F-deficient communities. There is concurrent evidence to indicate that there is an increase in the occurrence of mild enamel fluorosis among children in fluoridated and some nonfluoridated communities.³ In this context, a special issue of the *Journal of Dental Research* (1987) was devoted to measures in optimizing the safety and efficacy of F therapy.

Analysis of the F content in individual foods is the first step in monitoring the total F intake of infants and children. The concentrations of F and other trace elements in tea infusions and their contribution to the daily F intake recently have been reported (Wei et al. 1987). Beverages and milk formulas are the main sources of F when they are processed or prepared with fluoridated water.4 The consumption of commercially prepared soft drinks and other beverages is increasing in Hong Kong as well as in most parts of the world. For example, the daily intake of beverages (including fruit-flavored juices, carbonated and noncarbonated soft drinks, juice concentrate and mineral/spring water) in Hong Kong during 1983 was estimated to be 0.15 liters/person. Of this amount, 76% is from carbonated soft drinks (Hong Kong Department of Census and Statistics 1984). In the United States the average per capita consumption of soft drinks is estimated to be 419.5 (355 ml, 12-oz) cans during 1982 (Ismail et al. 1984). Four of 10 American toddlers consumed soft drinks in 1977 (Ritzek and Jackson 1980). The sale of one brand of spring water (Perrier) reached 23 million bottles per annum in the United Kingdom in 1979, while sales in the United States reached an average of 400 million liters per annum between 1978 and 1982 (MacFadyen et al. 1982). With increased industrial pollution of water supplies,

¹Glass 1982; Thylstrup et al. 1982; Rao 1984; Marthaler 1984.

²Ericsson and Wei 1979; Singer and Ophaug 1979; Messer and Walton 1980; Leverett 1982.

³Rozier and Dudney 1981; Driscoll et al. 1982; King and Wei 1986.

^{*}San Filippo and Battistone 1971; Adair and Wei 1979; Taves 1983; Ophaug et al. 1985.

mineral/spring waters are increasingly used in the reconstitution powder or concentrated milk formulas for babies and infants.

The aim of the present study was to determine the F content of commerically available infant formulas, beverages, whole milk, as well as typical infant foods provided by hospitals and used in the home. It is common for southern Chinese to make soups with ingredients such as meat with bones, dried seafoods, and herbs. Therefore, it also was important to include the F content of commonly available soups served in the home and purchased from local resturants.

Materials and Methods

The items tested in the present study include 20 powdered infant formulas, 15 infant hospital foods, 14 typical Chinese soups, 10 whole milk and dairy products, and 69 commercially available soft drinks, juices, and other beverages. The powdered formulas, whole milk products, and beverages were purchased in 1986 from supermarkets across Hong Kong. Soups were collected from homes and restaurants in Hong Kong. The beverages and whole milk products were analyzed for their ionic F concentrations (μ g F/ml), while the F concentrations in powdered formulas, foods, and soups represent the total F content μ g F/g). Analyses were carried out in duplicate or triplicate.

Prior to ionic F determination, carbonated beverages were decarbonated by stirring magnetically for 30 min at room temperature. Aliquots of decarbonated beverages were mixed with 10% by volume of TISAB III (Orion #940911) adjusted to pH 5.5 with NaOH. Samples of whole milk also were mixed with TISAB III (pH 5.2). The TISAB contained 2% of CDTA (1,2-diaminocyclohexane N,N,N', N'-tetraacetic acid), a metal-chelating agent which decomplexes F and therefore makes it available for measurement. The F concentrations were determined using a bank of five combination F electrodes (Orion #960900) coupled with a microprocessor ionalyzer (Orion #901). The reproducibility of electrode measurements were checked on NaF standard solutions buffered in the same manner as samples. The F concentrations in the standard solutions ranged from 0.01 to 0.1 ppm and from 0.025 to 1.0 ppm. The electrodes were preconditioned by soaking in a buffer containing F in a concentration corresponding to the lower limit of the F concentrations measured. In a separate experiment, the effect of polyvalent ions in complexing F was determined in 1 ppm F (as NaF) solutions containing 2.5 ppm Al³⁺, 10 ppm Fe³⁺, or 100 ppm Mg²⁺ and then buffered with 10% volume TISAB III.

The total F concentrations in powdered formulas, foods, and soups were assayed using the HMDS-microdiffusion method described by Taves (1968) with minor modifications (Hattab 1985). The method involved separation and concentration of F followed by measurement with the F- electrode.

Since acidulated soft drinks can contribute to erosion of dental hard tissues, the pH and the amount of acid in these drinks may be important factors which influence the erosion process. Hence, the titration values of these beverages also were determined as follows: to 10 ml of decarbonated and noncarbonated beverages, 0.1 M NaOH was added, under stirring, using an automatic .micropipette. The volume of alkaline required to raise the pH of each sample to 7-7.5 was recorded. Measurements were made using a pH-glass electrode (Radiometer pHM84, Denmark).

Results

The F concentrations in whole cow's milk and milk products, infant formulas, and hospital infant foods are shown in Table 1. The ionic F concentrations in whole milk were around 0.01 ppm. The pH of the various brands of fresh and long-life milk was approximately 6.5. The pH of acidophilus milk drink was 3.7. Addition of chocolate to milk increased the F concentration by 7to 22-fold. The total F contents ($\mu g/g$) of infant formulas were found to range between 0.06 and 1.08 ppm, with an average of 0.39 ppm F. If the infant formulas were diluted in a ratio of 1:1 with water containing 0.7 or 1 ppm F, the dosage of F would be about 55- and 70-fold more than that obtained from cow's milk or breast milk. Hospital infant foods were found to range from 0.27 to $0.93 \text{ ppm F}\mu\text{g/g}$ with an average F content of 0.59 ppm. There was less variation in F content of hospital infant foods than in infant formulas. The F content in hospital foods likely reflects the F content of the drinking water of 0.7 ppm.

The total F content in the typical soups ranged between 0.32 and 1.66 μ g/ml with an average of 0.68. The relatively high F concentrations in some of the soups are due mainly to the inclusion of seafoods and bones in the preparation. Thus, the dosage of F in a bowl of soup (approximately or equal to 250 ml) may contain as much as 0.42 mg F, assuming the F content was 1.66 μ g/ml.

The ionic F concentrations and pH of the beverages are presented in Table 2 and in the Figure (page 16). For the seven different commercially marketed mineral/ spring waters only trace amounts of F (0.01-0.10 ppm) were detected; however, Ramlosa[®] was an exception and it contained the highest F concentration of all tested beverages. The pH of the different waters ranged from 5.7 to 8.1. The F concentrations of canned soft drinks (carbonated) ranged from 0.02 to 0.78 ppm (µg/ml).

The average F contents in canned soft drinks, fruitflavored drinks, and soda water were 0.42, 0.56, and 0.42 ppm, respectively (Table 2). The corresponding pH values were 3.0, 3.5, and 3.6, respectively. The pH values of carbonated soft drinks were significantly lower than the pH of noncarbonated fruit-flavored drinks (P < 0.025). The F contents in prepackaged fruit juices are presented in the Figure. The average F content and pH were 0.65 and 4.7, respectively (N = 22). The items which contained the higher F content (1.28-1.66 ppm) are lemon teas. The mean F content in prepackaged fruit jucies and soft drinks produced in Hong Kong was 0.51 \pm 0.14 (\pm SD) ppm. This means that the F content in the beverages was 27% less than that of drinking water (0.7 ppm) used for their manufacture. With the exception of

TABLE 1. Fluoride Concentration in Whole Milk and Dairy Products (μ g F/ml) and in Powdered Infant Formulas and Hospital Infant Foods (μ g F/g)

	Mean
	Fluoride
Brand and Country of Origin	Concentration
Whole milk and dairy products	
Yakult® acidophilus drinks, HK	0.52
Pure-pak [®] chocolate milk drink, HK	0.22
Vita [®] chocolate milk beverage. China	0.07
Dairy Farm [®] whole milk, Belgium	0.01
Dutch Lady [®] whole milk, Belgium	0.01
Country Life [®] whole milk. Belgium	0.01
Devondale Long Life® milk, Australia	0.01
Paul's Long Life [®] , Australia	0.01
Paul's Shake Dairy [®] drink. Australia	0.01
Dairy Farm Long Life [®] Australia	0.01
Duily fullit Dong Dire , Hustituliu	0.01
Powdered infant formulas	
Enfamil® with iron	1.08
Alacta-NF®	0.91
Lactogen®	0.87
SMA®	0.58
Isomil [®]	0.51
Cow and Gate Plus®	0.45
Enfalac®	0.42
Premium®	0.39
Nan 1®	0.35
Follow-on®	0.32
Enfamil®	0.30
Lactogen FP®	0.29
Aptamil [®]	0.27
S26®	0.25
Mulumil®	0.23
Frisolac [®]	0.16
Nan 2®	0.15
Frisomil®	0.14
Similac [®]	0.12
Similac [®] with iron	0.06
Hospital infant foods	
II.	0.07
Porridoe	0.97
Common	0.71
Congree [®] Roblym [®] (rice coreal)	0.73
Cow and Cate® (milk correct)	0.70
Nostum [®] and coroals	0.07
Restuit * and tereals	0.04
Miluna® apple instant mills sereed	0.64
Carolac® (milk caroal)	0.01
Puree	0.56
	0.50
Soft rice	0.52
Steam and water	0.50
Minced meat	0.30
Mixed vegetable	0.20
Mineu Vegetable	0.2/

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tea drinks, there was generally no correlation between the F content and beverage type. Further analyses on some of the samples revealed that almost all F in the beverages was in ionic form and was available for measurement with the F electrode.

TABLE 2. Fluoride concentration $(\mu g/ml)$ and pH of Commercially Available Beverages

		Mean Fluoride Concen-
Brand and Country of Origin	pH	tration
Bottled mineral/spring water		
Romlosa [®] , Sweden	5.7	3.01
Gevser®, HK	7.9	0.54
No Frills® France	78	0.10
Pierval® France	77	0.10
Perrier [®] , France	5.4	0.10
Rochemaure [®] . France	7.6	0.10
Evian [®] France	7.7	0.10
Gulfa® UAE	8.1	0.02
Table water. HK	7.4	0.01
Canned soft drinks (carbonated)		
Shasta® grape, USA	3.2	0.78
Lemonade, Australia	3.0	0.76
Pashene®, Australia	2.9	0.67
Schwepps® orange, HK	3.1	0.63
Super Cola®, HK	2.5	0.63
Coca-Cola®, HK	2.4	0.58
Sprite®, HK	3.3	0.58
Sidra® apple soda, HK	2.8	0.57
Fanta® orange, HK	2.9	0.54
Orange drink, Australia	3.0	0.52
Diet Seven-Up®, HK	3.2	0.47
Sunkist® lemon-lime, HK	3.4	0.35
Sparkling orange, HK	3.2	0.33
Lemon cola, USA	2.7	0.22
Tab®, HK	2.8	0.19
Pepsi®, HK	2.4	0.12
Diet Pepsi®, HK	3.1	0.07
Seven-Up®, Holland	3.2	0.04
Lucozade® sparkling glucose, UK	3.4	0.02
Canned fruit-flavored drinks (noncarb	onated)	
Lemon tea, Pokka®, Singapore	3.5	2.05
Welchade [®] grape, USA	3.1	1.12
Lemon tea, Yeo [®] , Singapore	3.3	0.92
Welch's [®] grape (bottled), USA	3.5	0.86
Sugar cane, Singapore	5.2	0.59
Sunkist Oj Orange®, HK	3.3	0.51
Honey lemon, HK	2.6	0.21
Orange juice, USA	3.7	0.10
Pocari® electrolyte, Japan	3.6	0.06
Magic [®] orange, USA	3.2	0.05
Green Spot® orange, Holland	2.7	0.02
Canned soda water (carbonated)		
Soda water, Australia	6.2	0.81
Tonic water, Australia	2.7	0.67
Dry ginger ale, Australia	3.1	0.67
Schwepps [®] cream soda, HK	3.2	0.59
Cream soda, Australia	3.2	0.52
Tonic water, HK	2.7	0.47
Canada Dry® club soda. Holland	5.6	0.03
Canada Dry® tonic water. Holland	2.7	0.02
Canada Dry [®] ginger ale, Holland	2.8	0.02



Prepackaged fruit juice

FIGURE. The distribution of fluoride concentrations in prepackaged fruit juices.

Table 3 shows the titration values of different beverages. The titration values ranged from 1.0 to 10.0. The higher values were found in Welch's® grape (containing ascorbic acid and grape juice) and Magic® orange (containing orange pulp sac, concentration orange juice, citric acid, water, and sugar). Carbonated soft drinks which contain phosphoric acid (Coca-Cola® and Pepsi®) showed the lowest titration values.

The precision of ionic and total F measurement was evaluated on the differences between duplicate determinations. The coefficient of variation (CV = SD/mean) for F and total F measurements were 2.1 and 6.8%, respectively, indicating a high degree of precision. Analysis of standard solutions containing 0.01, 0.025, 0.05, and 0.1 ppm on 19 separate occasions gave linear calibration curves with a correlation of coefficient invariably better than 0.994. The intercept and slope of the plot of mV readings versus the logarithms of concentrations were 4.46 and 0.047, respectively. The calibration curves for 0.025, 0.05, 0.1, and 1.0 ppm F were linear with a coefficient of correlation (for 17 curves) better than 0.997. The intercept and slope for these curves were 3.19 and 0.041, respectively. When the observed mV readings of 0.01 ppm F were extrapolated and transformed to concentration unit, the predicted outcome was 0.014 \pm 0.0015 ppm and the coefficient of the variation of F concentrations was 10.7% (N = 19). The F recovered from 1 ppm F solutions spiked with 2.5 ppm Al³⁺, 10 ppm Fe³⁺, and 100 ppm $Mg^{\scriptscriptstyle 2+}$ were 89, 92, and 97%, respectively. It should be noted that the added aluminum and iron were higher than the highest levels in the drinking water; i.e., they ranged from 0.003 to 1.5 ppm for Al³⁺ and 0.002 to 1.7 ppm for Fe³⁺ (Durfor and Becker 1964).

Discussion

The notable occurrence of very mild to mild enamel fluorosis in relation to different forms of feedings highlights the need for thorough investigation of the dietary sources of F and their daily intake in young children. It has been suggested that the total dietary intake, including drinking water, should provide 0.05-0.07 mg F/kg of body weight for optimal dental health benefits (Forrester and Schulz 1974). The apparent threshold of 2.0 ppm F in drinking water at which dental fluorosis becomes evident corresponds to a daily intake of about 0.1 mg F/kg of body weight up to the age of 12 years.

In estimating the F intake for different age groups of children, attention should be paid to the three major variables that may influence the therapeutic action and safety of the ingested F. First, due to the considerable variations in the extent of F absorbed from foodstuffs, the actual F intake should be based on the biologically available F rather than solely on the F content in a given food item. The extent of F absorption is governed by many factors including the physical form of the dose, the presence of food in the stomach, the gastric pH and the concurrent oral administration of cation-rich foods such as milk and dairy products.

Second, it is important to determine whether the infant is bottle or breast fed because there is a very low F concentration (0.01-0.025 ppm) in breast milk.⁵ Infants who are bottle fed with dry-milk formulas diluted with fluoridated water ingest substantially higher F doses than breast-fed infants.

Third, as a convenient method of administration, some F tablet manufacturers have suggested dissolving the tablets in drinks. The dissolution and the release of F in beverages vary considerably depending on the physicochemical properties of the solvent (Hattab

⁵ Ericsson and Ribelius 1971; Adair and Wei 1978; Spak et al. 1983.

TABLE 3. Titration Values of Beverages Tested

Brand	Titration Value*
Fruit-flavored drinks (noncarbonated)†	
Welch's [®] grape (concentrated)	10.0
Magic [®] orange	7.3
Welchade® grape, Hi-C® mango, Vita® guava, Ribena® black currant Vita® mango, President® plum, Sunkist® orange, Vita® lemon tea, Hi-C® lemon tea,	4.2-4.7
Pocari® electrolytes	2.8-3.6
Soft drinks (carbonated)‡	
Lucozade® sparkling glucose, Sidra® apple soda Shasta® grape, Sparkling orange, Diet Seven-Up®,	4.2-4.6
(citric acid), Sunkist® lemon-lime	2.1-2.6
Sprite [®] , Seven-Up [®] , Coca-Cola [®] , Diet Pepsi [®]	1.0-1.8
t m10.1 M NoOH wood to new trailing (nH 7, 7, 5) 10 ml of	howarages

* mI 0.1 M NaOH used to neutralize (pH 7–7.5) 10 ml of beverages.

† Initial pH ranged between 2.9 and 3.6.

‡ Initial pH ranged between 2.6 and 3.4.

1985). For example, about 37% of F (as NaF tablets) added to cow's milk was not in ionic form (F) whereas essentially all added F to deionized water, orange juice, and Coca-Cola® were in ionic form.

Earlier reports on the F content in whole cow's milk showed much higher values than 0.01-0.025 ppm which was found in the present study and others.⁶ The low F concentration in cow's milk has a significant impact on the daily F intake, as Fomon (1975) found that the number of U.S. infants fed commercially prepared milkbased formulas have decreased from 64 to 29% from one month to six months of age. Conversely, the number of infants fed cow's milk has increased from 2 to 58% over the same period. By six to nine months of age, 92% of the infants are being fed cow's milk. Infants in Hong Kong are mainly fed commercial milk formulas. Breast feeding is done by around 15% of mothers for variable periods of one to six months.

There were no appreciable differences in the F concentrations of cow's milk obtained from various geographic areas (Table 1). It seems, therefore, that the F concentration in cow's milk is not altered appreciably by changes in the amount of F consumed by cows grazing in pastures of different F content. The total F content in powdered infant formulas (undiluted) ranged between 0.06 and 1.08 μ g/g, with an average of 0.39 μ g/g (Table 1).

Data on a smaller number of products of infant formulas diluted with deionized water have shown that they range from 0.10 to 0.29 ppm with an average of 0.17 ppm (Adair and Wei 1979). Recently, Johnson and Bawden (1987) measured the F content in infant formulas from seven cities across the United States. They found that the F concentrations in five powdered infant formulas diluted with deionized water ranged between 0.03 and 0.24 ppm F (mean = 0.12 ppm F). Our findings (Table 1) agree with Johnson and Bawden (1987) in that the most notable variations in F content were found between the products Enfamil® and Similac.® A variation of two to three times in F content were found in Enfamil® and Similac® with and without iron. However, measurements of F content of some infant formulas reported in this study were higher than those found in the United States.

The F content in beverages varies greatly not only between products of different sources, but also between different products of the same manufacturer. With the exception of tea drinks, the F content of the beverages produced in Hong Kong was 27% less than the F content of drinking water in Hong Kong. There is conflicting evidence regarding whether storing the beverages in glass bottles could affect the F content of the beverages. In a three-year study on the stability of NaF solutions in glass and plastic containers, it was found that the F ⁶Stamm and Kuo 1977; Ericsson and Wei 1979; Singer and Ophaug 1979. content of acidulated and neutral F solutions was not detectably changed after six-months' storage in glass containers at 37° C (Hattab 1981). It seems, therefore, that the variations in F content of a given beverage bottled in plastic or glass more likely is due to the fluctuation of the F concentration at the source of manufacture.

The differences in the titration values of the tested beverages (Table 3) seem to reflect their composition, i.e., the type and concentration of fruit juice, dietary acids (citric, ascorbic, phosphoric, and lactic), and sugar. The 100% grape juice enriched with ascorbic acid (according to the manufacturer of Welch's® grape) and the concentrated orange juice plus citric acid (Magic® orange) had the highest titration values. Lucozade® sparkling glucose which contains 22% glucose syrup, and citric and lactic acids had a titration value of 4.6. Beverages containing phosphoric acid (Coca-Cola® and Pepsi®) showed the lowest titration values. Frostell (1970) studied the effect of rinsing with different drinks on plaque pH of 18 persons. He found that lemonade and Coca-Cola® produced pH decreases comparable to those after rinsing with a 25% sucrose solution while orange, apple, and lemon juices produced initial decreases in pH exceeding those of sucrose rinses, but the pH recovered more quickly due to increased salivary stimulation.

From the F contents of foods and beverages it is possible to calculate the likely F intake from different food groups using the total diet consumption data (Pennington 1983). Presently available data indicate that infants six to 11 months of age daily consumed an average amount of: 254 g whole milk, 180 g infant formula (ready-to-feed), 150 g water, 28 g soft drinks, and 8.7 g tea beverages. Assuming the infant formulas were diluted 1:1 with tap water (0.7 ppm F) and that an infusion of tea contains 1.5 ppm F (Wei et al. 1987), the mean F intake from these dietary sources is calculated to be 0.23 mg/day or 0.027 mg/kg of body weight. Other dietary sources which contribute approximately 17% of the total dietary F intake (Ophaug et al. 1985) are derived from meat, fish, poultry, and vegetables. Dried seafoods, which constitute a significant part of the diet in Southeast Asia, are rich in F and may contain between three and 292 μ gF/g (Wei and Hattab 1987).

The present study showed a great variation in F content of commercially available infant formulas and beverages. Although the drinking water of Hong Kong is artificially fluoridated to the optimum level, the high prevalence of mild enamel fluorosis suggests that F consumption has increased. Fluoridated water often is used in processing foods and beverages. While levels of F content in infant formulas are now lower in the United States, our findings indicate that the F concentration in some formulas available in Hong Kong is still high. The

manufacturers of infant formulas are urged to control the F content in their exported product. Other nondietary sources of F which may significantly contribute to the total F intake of preschool children are derived from the unintentional ingestion of F dentifrices, mouthrinses, and topical fluorides.

Further surveys on the prevalence of enamel fluorosis and dental caries are required to validate the current trends of decline in dental caries and increase in mild fluorosis. Until the new data are available, one would like to see products with lower concentrations of F, especially dentifrices and topical gels for use in young children.

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