ELSEVIER

Environmental Pollution 104 (1999) 197–205

ENVIRONMENTAL POLLUTION

Fluoride contents in tea and soil from tea plantations and the release of fluoride into tea liquor during infusion

K.F. Fung, Z.Q. Zhang[1], J.W.C. Wong, M.H. Wong*

Institute for Natural Resources and Waste Management, and Department of Biology, Hong Kong Baptist University, Kowloon Tong, Hong Kong, People's Republic of China

Received 30 June 1998; accepted 4 October 1998

Abstract

Tea *Camellia sinensis* (L.), a perennial shrub, is cultivated in acidic soils. It has been noted that the occurrence of fluorosis in some inhabitants of pastoral and semiagricultural, semipastoral areas of Sichuan Province, People's Republic of China, is due to drinking a large quantity of tea liquor made from brick tea. Brick tea is made from fallen leaves and old leaves, and is not a considered a quality tea. The fluoride (F) contents of tea bushes and soil samples collected from five plantations from Guangdong Province, People's Republic of China, were tested. Fluorine accumulated mainly in leaves, especially in fallen leaves. The F concentration in fallen leaves (0.6–2.8 mg/g) were higher than that in young leaves (0.3–1.0 mg/g). Their concentrations were related to the total F and extractable F contents in the top soil (0–20 cm). In addition, soil pH and extractable Al concentration also affected the F contents in soils and plant tissues. Two methods (repeated and continuous) of infusing tea were employed to brew 17 brands of tea leaves in six categories: green tea, black tea, oolong tea, pureh tea, brick tea and black tea from England and Sri Lanka. In general, F contents in the infusions accounted for 24 to 83% of the total F contents of the original tea leaf samples. The cumulative F contents detected in the tea liquor prepared by repeated infusion were slightly higher than those prepared by continuous infusion. Brick tea released an extremely high F content (7.34 mg/litre), when compared with release from black tea (1.89 mg/litre) or green tea (1.60 mg/litre). In order to prevent development of fluorosis, the maximum consumption of fu-brick tea and black brick tea should be 1.7 litres and 4.8 litres/day respectively.

Keywords: Fluoride; Tea; Brick Tea; Fluorosis; Infusion

1. Introduction

Tea is the most popular beverage in the world. About 3,000,000 people drink tea in Hong Kong, and the tea consumption per capita is 2.07–2.24 kg/year (King and Tsang, 1987). The majority of tea consumed in Hong Kong is grown in mainland China, and a smaller portion from Sri Lanka, England and India (King and Tsang, 1987). There are different types of tea leaves, prepared with different infusion methods. In general, black (fermented) and green tea (without fermentation) are the two main ones, which are defined by their respective manufacturing techniques. Oolong tea produced in some regions of China, is an intermediate variant (semi-fermented) between black and green tea (Graham, 1983). All of these teas are made from young leaves. In some semi-pastoral and semi-agricultural areas of China, outer Mongolia and Russia, brick tea made from old leaves, fallen leaves and branches (Graham, 1983; Chao et al., 1995) is consumed commonly. There are different methods of infusing tea (Gulati et al., 1983). In Hong Kong, the most common method is to infuse tea leaves for a few minutes using a small volume of hot water (50 ml), and the same tea leaves are repeatedly infused until no taste or colour is produced. Tea also is infused continuously using a larger volume (>1 litre) of hot water for a longer period, up to several hours. In some areas of Sichuan, Hunnan and Xizhan Provinces, brick tea which serves as the main beverage is infused all day long (Chao et al., 1995).

Tea (*Camellia sinensis*), a perennial crop, matures after 7 years growth and, with careful field maintenance and regular pruning, can be plucked at a steady yield for up to 100 years (Carr and Stephens, 1992). The favoured conditions for tea cultivation included a suitable temperature (15–25°C), high relative humidity (80–90%), and high annual rainfall (around 1500–2000 mm;

* Corresponding author.

† Present address: School of Life Science, Zhongshan University Guangzhou, PR China.

0269-7491/99/$—see front matter
PII: S0269-7491(98)00187-0

Chan, 1978). Soil for tea cultivation should be acidic (optimum pH 4.5–6.5); and the gradient of the plantations should be about 10–20 (Chan, 1978; Carr and Stephens, 1992). Most of the tea plantations in China are found on mountains with mist, and the high humidity ensures that leaves grow slower and remain tender. With temperature ranging from 20.2–25.4°C, soil pH from 4 to 6, and an annual rainfall of 1500 mm or more, Guangdong Province in southern China is one of the popular provinces for tea production (Chan, 1978; Hseung, 1986).

A small amount of F is beneficial in the prevention of dental caries. It has also been used to treat osteoporosis. However, long-term overexposure to F may cause dental lesions and effects on bones. Rocks, soil, water, air, plants, animals and foodstuffs all contain F in widely varied concentrations. The plant (*C. sinensis*) takes up F from soil and accumulates F in its leaves, and is considered as a major source of F (WHO, 1970, 1984; Sha and Zheng, 1993a, b, 1994b; Lin, 1994; Chao et al., 1995). A substantial amount of F is released during tea infusion and nearly all (about 94.9%) of the released F is available to consumers (WHO, 1984). Recently, it has been established that the occurrence of fluorosis in some inhabitants of pastoral and semi-agricultural, semi-pastoral areas of Sichuan Province is due to drinking a large amount of tea liquor, made from brick tea (Chao et al., 1995, 1997).

The main objectives of the present study were:

1. to evaluate the F contents in tea bushes and soils from five tea plantations in southern China;
2. to compare the release of F from different brands of tea using two infusion methods; and
3. to determine the maximum daily F intake for different brands of tea by examining their F contents in the tea liquor.

2. Materials and methods

Soil and tea bushes were collected from five tea plantations in Guangdong Province namely Haiou, Lechang, Liuxihe, Puning and Yingde, to study the distribution of F in soil and different tissues of tea bushes. The soil samples were air-dried and passed through a 2-mm mesh sieve, and the plant samples were ground into powder after they had been dried at 70°C to a constant weight. The potential risk of consuming brick tea (1 black-brick tea and 1 fu-brick tea, from Hunnan Province), compared with other types of tea (3 green tea, 3 oolong tea, 3 pureh tea, 3 black tea from different provinces of People's Republic of China and 3 black tea from overseas: one from Sir Lanka and two from England) was investigated by examining the F contents in tea liquor using two infusion methods.

2.1. Total F contents in tea and soil samples

A total of 0.1 g of tea plant or soil was mixed with 3 ml of NaOH in a nickel crucible which was placed in an oven (150°C) for 2.5 h. After the NaOH was solidified, the crucible was placed in a muffle furnace (300°C and then 600°C) for 30 min. The sample was allowed to cool, before 5 ml of distilled water were added. Then 3 ml of 37% HCl was added to adjust the pH to 8–9. The sample was transferred to a 100-ml volumetric flask, diluted to the volume and filtered through Whatman No. 40 filter paper (McQuaker and Gruney, 1977). The standard reference material (tea no. GBW 07605, obtained from the Institute of Geophysical and Geochemical Exploration, Langfang, People's Republic of China) was used for quality assurance.

2.2. Water soluble F contents of soil samples

Ten grams of soil was weighed into a conical flask, and mixed with 10 ml of distilled water. The mixture was shaken for 5 min at 120 rpm, allowed to stand for 3 h and the suspension filtered through a Whatman No. 40 filter paper (Brewer, 1965).

2.3. Extractable Al contents of soil samples

A total of 5 g of the soil sample was weighed into a centrifuge tube and mixed with 25 ml of 1 M KCl. The mixture was shaken for 30 min at 120 rpm, centrifuged for 10 min at 1000 ***g***, and the suspension filtered through a Whatman No. 42 filter paper (Bloom et al., 1979).

2.4. Infusion methods

2.4.1. Repeated infusion

One gram of the tea leaves was infused with 100 ml double-distilled water (DDW) at 100°C (1%, w/v tea infusion) in a 150-ml conical flask (King and Tsang, 1987). At 5-min intervals, the tea liquor was discarded, except 10 ml saved for F determination, and the same volume of boiling water (100°C) was refilled. The same procedure was repeated five times. There were three replicates of each brand of tea leaves undergoing the same procedures.

2.4.2. Continuous infusion

One gram of the tea leaves was infused with 100 ml DDW at 100°C (1%, w/v tea infusion) in a 150-ml conical flask (King and Tsang, 1987). The tea liquor was allowed to remain in contact with the leaves for 5, 30, 60, 120 or 360 min. After the conical flasks had been filled with boiling water, they were placed in a water bath for the required time, with the temperature kept between 89 and 92°C. For each duration, 10 ml liquor

in each conical flask was used for the determination of F content. There were three replicates of each brand of tea leaves undergoing the same procedures.

2.5. Analytical procedures

Fluoride concentrations in tea infusions, soils and different tissues of tea bushes were analyzed according to the method described in Duckworth and Duckworth (1978). A total of 10 ml of standard F or samples was added to 10 ml total ionic strength adjusting buffer. After using an ISE to calibrate the standard curve, F concentrations in the samples were measured. Extractable Al concentrations in soil were detected by atomic absorption spectroscopy (AAS; Bertsch & Bloom, 1996) and pH (soil:distilled water 10:25) values were measured by a pH meter (Thomas, 1996).

2.6. Statistical analysis

Duncan's Multiple Range Test (SAS) was used to compare the difference of F contents, extractable Al contents, and pH in the top soil (0–20 cm) within five tea-plantations, the F contents in different parts of tea bushes of the five tea-plantations, and the release of F into tea liquor during infusion after conducting one-way ANOVA (Little and Hills, 1978).

3. Results

The recovery rates of F using ISE and Al using AAS, according to the certified values of the standard reference material were 97 and 95%, respectively.

3.1. Tea bushes and soil samples within five plantations from Guangdong Province, People's Republic of China

The soil types of the different plantations are: ferruginous latosols (Haiou), yellow earth (Lechang) and red earth (Liuxihe, Puning and Yingde; Chan, 1978; Hseung, 1986). According to Table 1, pH values of all the top soils were acidic, ranging from 3.8 to 4.5, total F ranging from 186 to 387 μg/g, water-soluble F ranging from 0.76 to 2.71 μg/g, and extractable-Al ranging from 87 to 353 μg/g.

Table 2 illustrates the distribution of F in different tissues of tea bushes from the five plantations. The results indicated that F was accumulated in leaves, especially in mature leaves and fallen leaves. The F contents in branches and roots ranged from 20.8 to 34.6 μg/g.

According to Table 3, there were significant relationships between total F and water-soluble F contents ($r = 0.89$, $p \leq 0.01$, df = 38), and also between water-soluble F and extractable Al contents ($r = 0.6$, $p \leq 0.01$, df = 38).

Table 1
Mean pH, fluoride and aluminium in top soil samples (0–20 cm) in five plantations from Guangdong Province, People's Republic of China

Plantation	No. of samples	pH	Total fluoride	Extractable fluoride	Extractable aluminium
			μg/g		
Haiou	6	4.37 ± 0.24b	188.87 ± 30c	1.05 ± 0.50cd	86.74 ± 46d
Lechang	3	3.81 ± 0.09e	387.87 ± 19a	2.03 ± 0.18b	352.95 ± 60a
Liuxihe	3	4.03 ± 0.04d	383.23 ± 42a	2.71 ± 0.29a	189.54 ± 25b
Puning	3	4.23 ± 0.15c	255.14 ± 34b	1.34 ± 0.51c	121.03 ± 43c
Yingde	4	4.49 ± 0.22a	186.21 ± 33c	0.76 ± 0.08e	100.46 ± 51c

Values represent mean ± standard deviation. Same letters within each vertical column indicate no significant difference at $p \leq 0.05$, according to Duncan's Multiple Range Test.

Table 2
Correlation coefficients between different parameters of five tea plantations at Guangdong Province, People's Republic of China

	Soil pH	Soil extractable Al	F					
			Soil total	Soil soluble	Roots	Branches	Young leaves	Mature leaves
Soil pH								
Soil extractable Al								
Soil total F	−0.76****							
Soil soluble F	−0.58****	0.60****	0.85****					
F in roots	0.00		0.02	0.05				
F in branches	0.06		−0.06	0.05	0.57****			
F in young leaves	−0.19		0.50****	0.69****	0.33*	0.23		
F in mature leaves	−0.64****		0.82****	0.73****	0.14	0.10	0.43****	
F in fallen leaves	−0.66****		0.78****	0.79****	0.30**	0.20	0.46****	0.85****

*$p \leq 0.05$; **$p \leq 0.01$; ***$p \leq 0.005$; ****$p \leq 0.001$.

Table 3
Total fluoride contents of tea bushes from five plantations of Guangdong Province, People's Republic of China

Plantation	No. of samples	Roots	Branches	Young leaves	Mature leaves	Fallen leaves
				μg/g		
Haiou	6	37.3 ± 4.4a	34.6 ± 4.7a	543 ± 71 b	598 ± 76 c	1212 ± 413c
Lechang	3	31.5 ± 9.1b	27.8 ± 8.1b	508 ± 54 b	1228 ± 281a	2315 ± 401a
Liuxihe	3	34.3 ± 7.7ab	25.9 ± 4.9bc	808 ± 173a	1040 ± 187b	1845 ± 247b
Puning	3	23.5 ± 2.4c	22.4 ± 2.1c	340 ± 54 c	711 ± 98 c	976 ± 158cd
Yingde	4	25.7 ± 3.2c	20.8 ± 7.4c	406 ± 77 c	463 ± 76 d	752 ± 176d

Values represent mean ± standard deviation. Same letters within each vertical column indicate no significant difference at $p \leq 0.05$, according to Duncan's Multiple Range Test.

3.2. Total F contents in 17 brands of tea leaves

Table 4 shows that the total F contents (determined by alkaline extraction) in 17 brands of tea leaves ranged from 170 to 878 mg/kg. If brick tea samples were not included, the total F contents ranged from 170 to 423 mg/kg. Brick tea, due to the use of old leaves, fallen leaves and even branches, contained the highest total F contents which were 2 to 4 times higher than other brands made from young leaves (one shoot with two leaves). The lowest F content was in oolong tea leaves (170 to 224 mg/kg).

Table 4
Total fluoride contents in 17 brands of tea leaves

Type of tea	Place	F content (μg/g)
Green	Zhejiang	217 ± 26gh
Green	Guangdong	336 ± 27e
Green	Guangdong	344 ± 22d
Oolong	Guangdong	224 ± 13fgh
Oolong	Yunnan	170 ± 6i
Oolong	Yunnan	195 ± 6hi
Pureh	Yunnan	257 ± 11f
Pureh	Yunnan	346 ± 13d
Puerh	Yunnan	297 ± 7e
Black	Guangdong	423 ± 27c
Black	Yunnan	348 ± 17d
Black	Yunnan	322 ± 5de
Twinings	Sir Lanka	296 ± 14e
Lipton	England	248 ± 9fg
Rickshaw	England	204 ± 10hi
Brick	Hunan	680 ± 30b
Brick	Hunan	878 ± 60a

Values represent mean ± standard deviation. Same letters within each vertical column indicate no significant difference at $p \leq 0.05$, according to Duncan's Multiple Range Test.

3.3. Effects of infusion method on F content of tea liquor

3.3.1. Repeated infusion

Table 5 illustrates water-soluble F contents in tea liquor prepared by repeated infusion. There were similar patterns among different brands of the same types of tea leaves. The highest total water-soluble F was obtained in tea liquor of brick tea (4.24–7.05 mg/litre), which was

Table 5
Water-soluble F concentration in 1% (w/v) tea liquor prepared by repeated infusion

Type of tea	Place	Dissolvable F in mg/litre tea liquor No. of 5 min infusion						% of F infused (total)
		1	2	3	4	5	total	
Green	Zhejiang	1.02	0.17	0.16	0.16	0.14	1.65	75.5
Green	Guangdong	1.02	0.26	0.19	0.15	0.13	1.75	52.5
Green	Guangdong	1.08	0.27	0.17	0.16	0.15	1.83	52.8
Oolong	Guangdong	0.82	0.19	0.17	0.17	0.16	1.51	67.8
Oolong	Yunnan	0.60	0.16	0.16	0.15	0.15	1.22	71.6
Oolong	Yunnan	0.54	0.23	0.15	0.15	0.15	1.22	62.7
Pureh	Yunnan	0.90	0.27	0.23	0.16	0.15	1.71	64.9
Pureh	Yunnan	1.11	0.26	0.22	0.16	0.14	1.89	54.5
Puerh	Yunnan	1.04	0.27	0.22	0.16	0.15	1.84	61.7
Black	Guangdong	1.30	0.50	0.26	0.18	0.17	2.41	56.8
Black	Yunnan	1.24	0.24	0.16	0.15	0.15	1.94	56.0
Black	Yunnan	1.08	0.27	0.23	0.16	0.16	1.90	59.1
Twinings	Sri Lanka	0.94	0.42	0.16	0.14	0.14	1.80	64.0
Lipton	England	0.97	0.23	0.17	0.14	0.09	1.60	64.6
Rickshaw	England	0.89	0.22	0.11	0.07	0.02	1.31	73.9
Brick	Hunan	2.01	1.28	0.47	0.29	0.19	4.24	62.5
Brick	Hunan	2.21	2.01	1.72	0.68	0.43	7.05	80.3

4 to 5 times higher than in the other types of tea. The lowest one was in tea liquor of oolong tea (1.22–1.51 mg/litre). In general, about 53% (green tea) to 80%(brick tea) of the total F contents contained in the tea leaves were released into the tea liquor.

Fig. 1 illustrates the water-soluble F contents in the tea liquor of different types of tea under repeated infusion. In general, the highest F contents released into tea liquor were observed in the first infusion, among the five time-intervals. In the second infusion, the F contents were rapidly diminishing. From the third to the fifth infusions, the contents gradually decreased. The F contents obtained in the first infusions ranged from 0.54 to 2.01 mg/litre (0.54 to 1.3 mg/litre when not including brick tea), and the percentages of F released into tea liquor were about 25–47% (28–47% when not including brick tea). For the second to fifth infusions, their F contents were 2.01–0.02 mg/litre (0.50–0.024 mg/litre, not counting brick tea), and the percentages of F released into tea liquor were 1.2–23% (1.2–14%, not counting brick tea).

The F contents released into tea liquor from tea leaves were related closely to the number of times of tea infusion. There were six sets of formula for six types

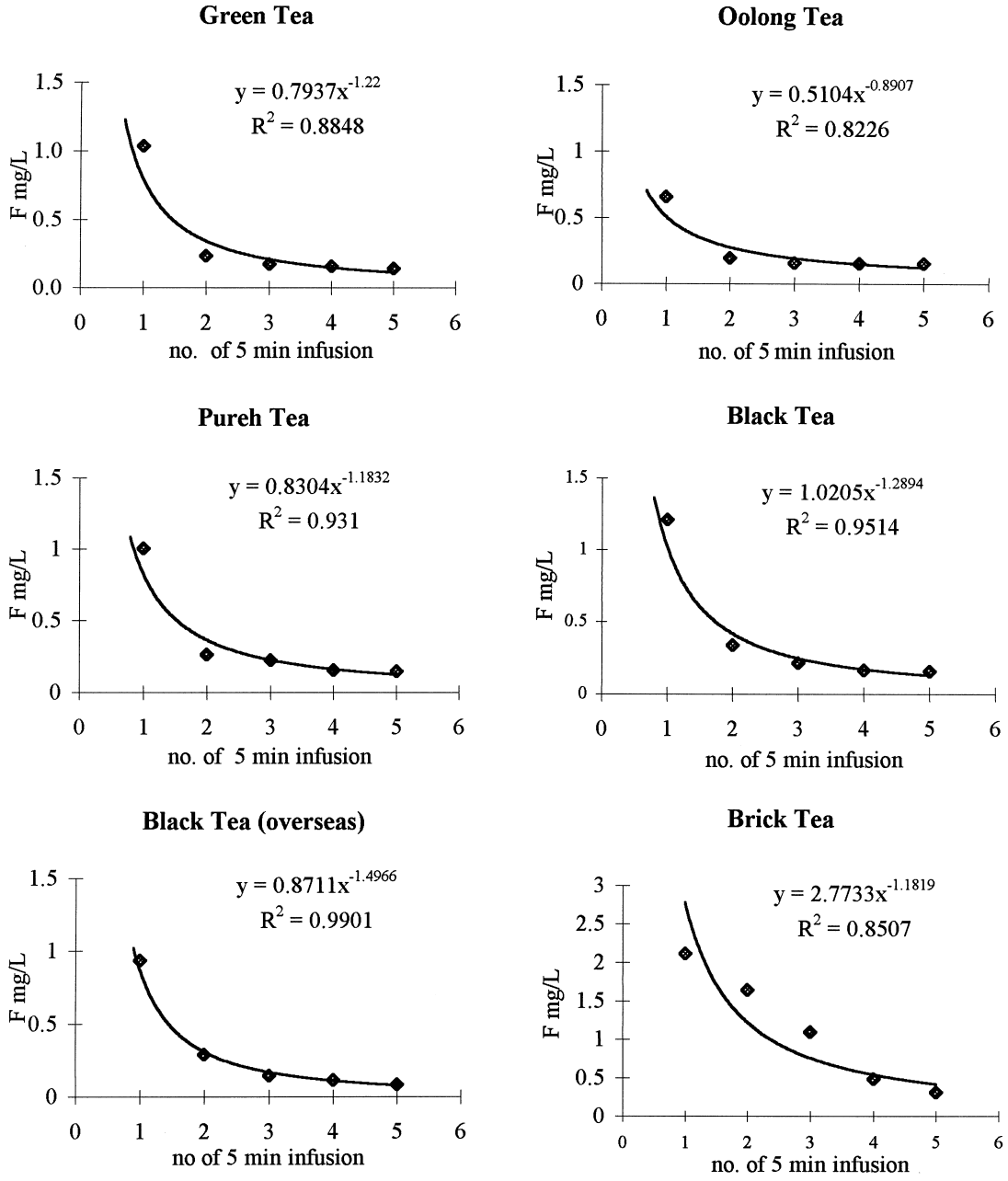

Fig. 1. Water-soluble fluoride concentrations in 1% (w/v) tea liquor prepared by repeated infusion (the types of tea and their respective origins have been shown in Table 5).

of tea leaves (Fig. 1); for example, the formula for oolong tea was $y = 0.5104x^{-0.8907}$and $R^2 = 0.8226$, with x being the number of times of infusion and y the fluoride content released during the time of infusion when 1 g of tea leaves was infused in 100 ml DDW at 100°C (1%, w/v).

The release of F into tea liquor from brick tea was different from other brands. Apart from the first infusion, the second and third infusions also released a rather high F content into the tea liquor, especially true for fu-brick tea. The difference of F contents released from the first three infusions was less than 6% (25.18−19.63%). The F contents in the first three infusions in tea liquor were 2.21–1.72 mg/litre. Its fifth infusion had the highest F content (0.43 mg/litre) when compared with those of other brands.

3.3.2. Continuous infusion

Table 6 shows the water-soluble F contents in tea liquor using continuous infusion. In general, there were similar patterns for different brands of the same type of tea leaves. The water-soluble F contents were directly proportional to the duration of tea infusion. Tea leaves produced from the same place released similar F contents into the tea liquor; for example, water-soluble F contents of oolong tea from Yunnan (0.70–0.77 mg/litre) were lower than those from Guangdong (0.97 mg/litre).

There were significant differences in F contents released among all the time intervals for brick tea, especially fu-brick tea. This was especially true for fu-brick tea, the F contents released from tea leaves during 5, 30, 60, 120 and 360 min were 2.18, 3.29, 4.75, 5.88 and 7.34 mg/litre respectively. The amount of F released from the total F content of tea leaves reached 84%.

Fig. 2 shows the release of F into tea liquor under continuous infusion. In general, a large amount of F was released into tea liquor during the first 30 min (with 77–85% released during the first 5 min), and the contents leveled off afterwards. No significant difference was obtained between the 5-min and 6-h infusions for oolong tea, whereas, for all other brands, the amount of F released from tea leaves was directly proportional to the duration of infusion, especially in brick tea.

The F contents released into tea liquor from tea leaves were related closely to the duration of tea infusion (Fig. 2). There were six sets of formula for the six types of tea leaves; for example, the formula for oolong tea was $y = 0.0383\mathrm{Ln}(x) + 0.5893$ and $R^2 = 0.99$ with x being the number of times of infusion and y the fluoride content released in the time of infusion when 1 g of tea leaves was infused in 100 ml DDW at 100°C (1%, w/v).

4. Discussion

Soil pH is a major factor that affects solubility of different compounds in soil. The availability of soluble F and Al decreased from pH <4 to pH 8 (Street and Elwaly, 1983; Wenzel and Blum, 1992). Fluoride in soil occurs as a variety of minerals and complexed with Al, iron and calcium. At a low pH, Al is the dominant partner to F to form AlF^{2+} or $Al\ F_2{}^{+}$ but the most dominant one is AlF_3 (Bower & Hatcher, 1967; Omueti & Jones, 1977; Davison, 1983). The stability of Al–F complex increases under a lower pH, and the release of F is always from a complex of Al (Ding and Huang, 1991; Wenzel and Blum, 1992). This principle explains why the more acidic top soil from Lechang (pH 3.81) and Liuxihe (pH 4.03) resulted in higher water-soluble F

Table 6
Water-soluble F in 1% (w/v) tea liquor prepared by continuous infusion

Type of tea	Place	Dissolvable F in mg/litre tea liquor					% of F in 30-min infusion	% of F in 360-min infusion
		5 min	30 min	60 min	120 min	360 min		
Green	Zhejiang	1.22	1.43	1.47	1.58	1.67	66.0	76.7
Green	Guangdong	1.20	1.32	1.53	1.52	1.61	39.4	48.0
Green	Guangdong	1.18	1.34	1.41	1.45	1.51	38.8	44.0
Oolong	Guangdong	0.82	0.87	0.91	0.95	0.97	39.1	43.2
Oolong	Yunnan	0.59	0.59	0.70	0.72	0.77	34.6	45.1
Oolong	Yunnan	0.58	0.59	0.65	0.69	0.70	30.3	35.9
Pureh	Yunnan	0.90	1.35	1.40	1.47	1.54	52.3	59.8
Pureh	Yunnan	1.13	1.35	1.41	1.48	1.56	39.1	45.0
Puerh	Yunnan	0.94	1.37	1.43	1.51	1.59	46.1	53.5
Black	Guangdong	1.02	1.59	1.63	1.67	1.77	37.6	42.0
Black	Yunnan	1.25	1.62	1.74	1.80	1.88	46.7	54.0
Black	Yunnan	1.35	1.66	1.85	1.90	1.89	51.5	58.8
Twinings	Sri Lanka	0.91	1.38	1.57	1.63	1.65	46.6	55.8
Lipton	England	0.94	1.25	1.32	1.39	1.44	50.5	58.3
Rickshaw	England	0.93	1.25	1.28	1.33	1.37	61.0	66.8
Brick	Hunan	2.28	3.01	3.17	3.78	4.73	44.3	69.6
Brick	Hunan	2.18	3.29	4.75	5.88	7.34	37.4	83.6

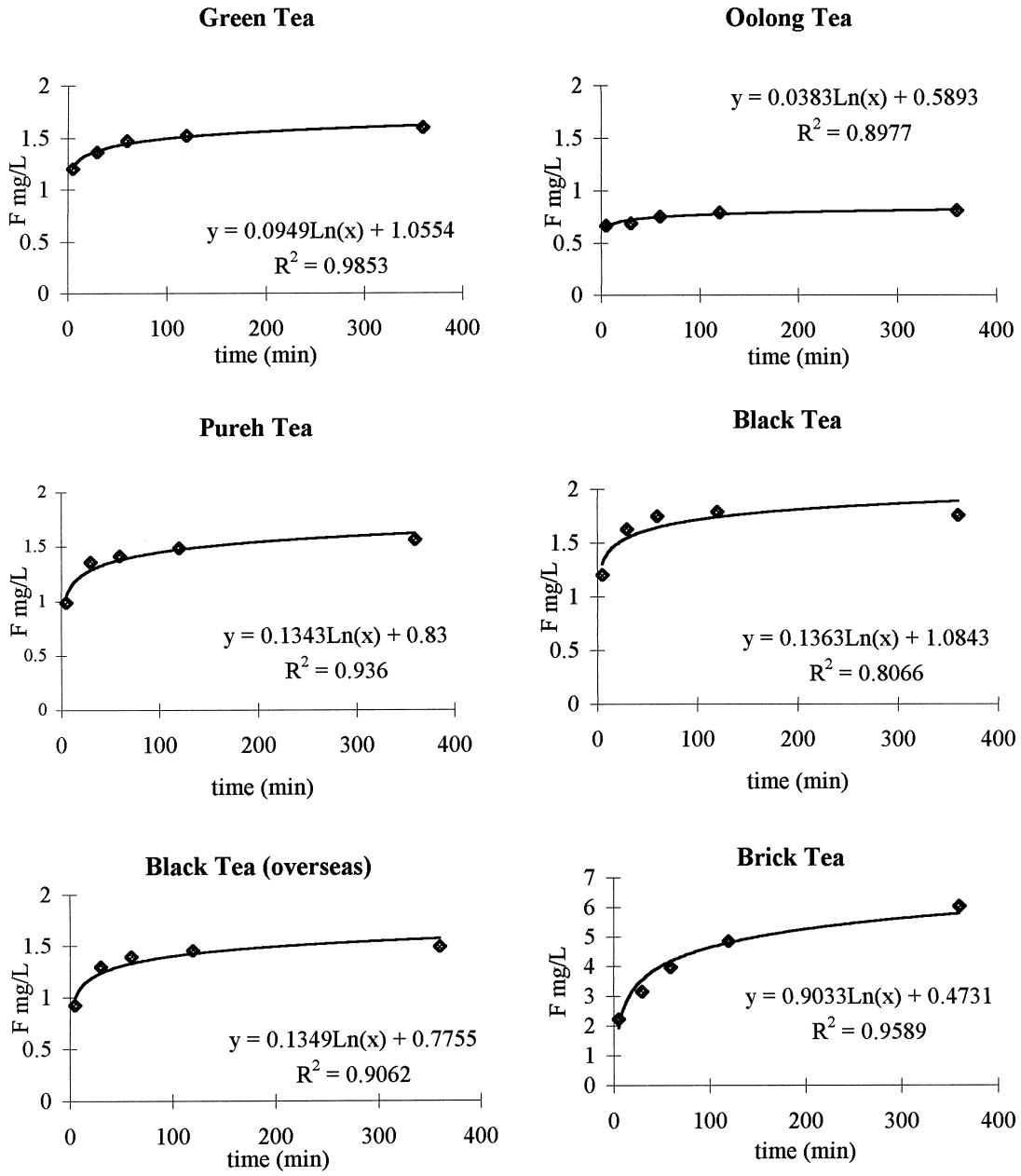

Fig. 2. Water-soluble fluoride concentrations in 1% (w/v) tea liquor prepared by continuous infusion (the types of tea and their respective origins have been shown in Table 6).

and extractable Al contents. For the contents of total F, water-soluble F and extractable Al obtained in the present study fell in the ranges reported in China soils (Sha and Zheng, 1993a, 1994a).

The bioaccumulation coefficients of leaves were very high since F contents in leaves were a 1000 times the soil water-soluble F and 2–7 times to total F contents. It has been observed that 97% F accumulated in leaves but accumulated only 3% in other parts of tea plant (Sha and Zheng, 1993a). Fluoride contents accumulated in leaves were proportionally related to the age of leaves (young leaves, 0.3–1.0 mg/g; fallen leaves, 0.6–2.7 mg/g). Higher F contents were noted in leaves of tea bushes from Lechang and Liuxihe, with lower pH and higher water-soluble F contents than in soil from the other plantations.

For the total F contents of tea products (170–423 mg/kg, which were in line with other reports: 199–422 mg/kg (King and Tsang, 1987), and 3.2–400 mg/kg (Canadian Public Heath Association, 1979). The results indicated that tea leaves from the same province seemed to have similar total F contents. The two green tea samples from Guangdong Province (336 and 344 mg/kg) had higher total F contents than those produced from Zhehiang Province (217 mg/kg); and the black tea from Guangdong Province (423 mg/kg) also contained higher

total F contents than those produced from Yunnan (302 and 325 mg/kg).

For tea infusion, higher F contents were released from black tea produced in China than those from overseas, due to the original higher total F contents in the tea leaves from China. However, higher percentages of F were released from the overseas black tea (42–59% in black tea from China, and 47–67% in black tea from overseas), due to the fact that the overseas black tea leaves had a higher surface area due to their smaller size ($<1\times1$ mm) and therefore more efficient in releasing F.

The results of repeated infusion experiment were similar to a previous study using oolong tea for the preparation of tea liquor by repeated infusion (2-min intervals, and repeated 10 times), with the formula of $y=0.7008x^{-1}$ and $R^2=0.9020$ (Sha and Zheng, 1993b). In addition, in a previous study using oolong tea for preparation of tea liquor by continuous infusion (2, 5, 10 and 30 min, 1, 6, 12 and 24 h), the formula was $y=25.1 \log x+27.0$ (equivalent to $y=0.109\mathrm{Ln}(x)+0.270$) (Sha and Zheng, 1993b). Both studies indicated the trend that a large portion of water-soluble F was released after 30 min.

The two brands of black tea (same company) from England showed a similar pattern of F release under continuous infusion, which was different from the black tea from Sir Lanka. The F contents released into tea liquor at 360 min from the two brands of tea from England were 1.44 and 1.37 mg/litre, respectively, whereas the one from Sri Lanka was 1.65 mg/litre.

According to the infusion experiments, the cumulative F contents released into tea liquor under repeated infusion were higher than those under continuous infusion. The results were in line with a similar study conducted by Sha and Zheng, (1993b) that the total water-soluble F contents from the total 10-times repeated infusion (106 mg/kg) were slightly higher than that from the 24-h infusion (94.4 mg/kg). Using black tea leaves as a typical example (Tables 5 and 6), the cumulative F contents released into tea liquor were 1.90–2.40 mg/litre (56–59% of the total F released from tea leaves) under repeated infusion, compared with 1.77–1.89 mg/litre (39–59%) under continuous infusion. However, an opposite pattern was observed for brick tea, possibly due to the fact that most of the cells of the old leaves were lignified and the F contained was released slowly during infusion.

Brick tea leaves released the highest F content among the six types of tea leaves investigated using two infusion methods. When not taking brick tea into consideration, black tea released the highest F content into tea liquor, with oolong being the lowest. There were no significant differences in F contents released from green tea leaves, pureh tea leaves and black tea leaves from overseas.

Liquids are the main source of F intake for human beings (WHO, 1984). Suggestions have been made with respect to the optimum F intake which ranged from 2 to 4 mg/day (WHO, 1970, 1984; Sha and Zheng, 1993b; Gajewska and Nabrzyski, 1994; Chao et al., 1995). The total intake of F per day, that may cause fluorosis (chronic uptake) is 13 to 14.48 mg/day (Sha and Zheng, 1993b; Chao et al., 1995). The patients who suffered from fluorosis drunk a kind of brick tea similar to the two types of brick tea used in the present study, which containing a high level of F tea (Editorial Community of Endemic Diseases and Their Environments, People's Republic of China, 1989; Chao et al., 1995, 1997).

Based on the above guidelines (safety limit of F intake, 4 mg/day; the amount of F intake that may cause fluorosis, 13 mg/day; the amount of F contained in tap water, 0.5 mg/litre), Table 7 lists the safety levels of the amounts of tea liquor for different tea brands. Since the average tea consumption per capita is about 1 litre/day, all the tea liquors are safe for consumption, except brick tea. The safety limit for drinking brick tea is 1.7 litres. People in Sichuan Province drink about 3.0–3.9 litres/day (Chao et al., 1995). The F concentration found in brick tea liquor of the present study was 7.34 mg/litre after 360 min of infusion, and did not include 0.5 mg F contained in 1 litre of tap water. In Sichuan Province, the F concentration in brick tea liquor was 7.76 mg/litre, which caused 62.5–73.5% of the population to suffer from fluorosis (Chao et al.,

Table 7
Suggested safety levels of different brands of tea with respect to F intake

Type of tea	Fluoride contents released from tea infusion (mg/litre)	Suggested amount of tea intake per day (litre) within the safety level	Suggested amount of tea intake per day (litre) within the limits that may cause fluorosis
Green tea	1.2–1.7	2.4–1.8	7.6–5.9
Oolong tea	0.6–1.0	3.6–2.7	11.8–8.7
Pureh tea	0.9–1.6	2.9–1.9	9.3–6.2
Black tea	1.0–1.9	2.7–1.7	8.7–5.4
Black tea (overseas)	0.9–1.7	2.9–1.8	9.3–5.9
Brick tea	2.2–7.3	1.5–0.5	4.8–1.7

1995). In addition, in view of the higher release of F in tea liquor for all tea brands infused for a prolonged period (e.g. 6 h), the habit of drinking tea liquor which contacted tea leaves overnight should be avoided (Sha and Zheng, 1993b).

5. Conclusion

Soil extractable F contents were closely related to soil types, soil pH, total F contents and F-containing complexes (e.g. Al). The F contents in tea bushes depended on the soil extractable F contents. Most of the F taken up by tea bushes is accumulated in leaves, especially in mature or fallen leaves (up to 2000 μg/g).

During tea infusion, 25–84% of the total F contained in tea leaves was released into the liquor. The cumulative F released under repeated infusion was slightly higher than that from continuous infusion, except for brick tea. Brick tea, released 4.73–7.34 mg F/litre after infusion for 360 min. The chronic intake of brick tea liquor therefore may cause fluorosis due to the fact that it is made from old leaves, fallen leaves and even branches which contain higher F concentrations.

Acknowledgements

Financial support from the Faculty Research Grant of Hong Kong Baptist University and the Research Grants Committee of the University Grants Council, Hong Kong, is gratefully acknowledged.

References

Bertsch, P.M., Bloom, P.R., 1996. Aluminium. In: Sparks D.L., et al. (Eds.), Methods of Soil Analysis. Part 3. Chemical Methods. 2nd Edition. Soil Sci. Am., Madison, WI.

Bloom, P.R., McBride, M.B., Weaver, R.M., 1979. Aluminium organic matter in acid soils: buffering and solution aluminium activity. Soil Sci. Soc. Am. J. 43, 488–493.

Bower, C.A., Hatcher, J.T., 1967. Adsorption of fluoride by soil and minerals. Soil Sci. 103, 151–154.

Brewer, R.F., 1965. Fluorine. In: Chapman, H.D. (Ed.), Diagnostic Criteria for Plants and Soil. Qual. Print. Co., Inc., Abilene, TX, pp. 180–196.

Canadian Public Health Association, 1979. Criteria Document in Support of a Drinking-water Standard for Fluoride. Canadian Public Health Association, Ottawa.

Carr, M.K.V., Stephens, W., 1992. Climate, weather and the yield of tea. In: Wilson, K.C., Clifford, M.N. (Eds.), Tea, Cultivation to Consumption. Chapman & Hall, London, pp. 87–135.

Chan, C.C., 1978. Detailed Record of Guangdong Province. Tin De Press, Hong Kong.

Chao, J., Zhao, Y., Liu, J.W., Bai, X.S., Zhou, D.Y., Fang, S.L., Jie, M., Wu, J.S., 1995. Research on fluorosis caused by drinking brick tea in Sichuan Province. Chinese Journal of Tea Science 1995(3), 35–38.

Chao, J., Zhao, Y., Liu, J.W., Zhan, S.Y., Wan, E.Q., Liu, X.L., Ma, W.X., Ha, B.S., 1997. Correlation analysis of fluorosis and drinking tea among Kazak people in Gansu Province. Acta Sci. Circum. 17(4), 493–497.

Davison, A.W., 1983. Uptake, transport and accumulation of soil and airborne fluoride by vegetation. In: Shupe, J.L., Peterson, H.B., Leone, N.C. (Eds.), Fluoride Toxicity. Paragon Press, Salt Lake City, UT, pp. 61–82.

Ding, R.X., Huang, X., 1991. Biogeochemical cycle of aluminium and fluorine in tea garden soil system and its relationship to soil acidification. Acta Pedologica Sinica 28(3), 229–236.

Duckworth, S.C., Duckworth, R., 1978. The ingestion of fluoride in tea. British Dent. J. 153, 64–66.

Editorial Community of Endemic Diseases and Their Environments in the People's Republic of China, 1989. The Atlas of Endemic Diseases and Their Environments in the People's Republic of China. Science Press, Hubei Province, People's Republic of China.

Gajewska, R., Nabrzyski, M., 1994. Fluoride content of tea and full day's hospital food rations. Bromat. Chem. Toksykol, XXVII(2), 153–156.

Graham, H.N., 1983. Tea. In: Mark, H.F., Othmer, D.R., Overberger, C.G., Seaborg, G.T., Grayson, M. (Eds.), Kirk-Othmer Encyclopedia of Chemical Technology, 3rd Edition. John Wiley & Sons, New York, pp. 628–644.

Gulati, P., Singh, V., Gupta, M.K., Vaidya, V., Dass, S., Prakash, S., 1983. Studies on the leaching of fluoride in tea infusions. Sci. Environ. 138, 213–222.

Hseung, Y., Li, J., 1986. The Soil Atlas of China. Cartographic Publishing House, Beijing.

King, M.N., Tsang, M.C.K., 1987. The fluoride content of Chinese and black teas available in Hong Kong. Fluoride 20(1), 18–23.

Lin, N.H., 1994. Analysis on the fluoride content in tea leaves in different ecological environment. Chinese Journal of Tea Leaves of Fujian Province 1994(2), 21–23.

Little, T.M., Hills, J.J., 1978. Agricultural Experimentation: Design and Analysis. John Wiley, New York.

McQuaker, N.R., Gurney, M., 1977. Determination of total fluoride in soil and vegetation using an alkali-fusion-selective-ion electrode. Anal. Chem. 49, 53–56.

Omueti, J.A.I., Jones, R.L., 1977. Fluoride adsorption by Illinois soils. J. Soil Sci. 28, 564–572.

Sha, J.Q., Zheng, D.X., 1993a. The characteristics of bioaccumulation of fluoride in tea plant. Chinese Journal of Tea Leaves of Fujian Province 1993(3), 25–28.

Sha, J.Q., Zheng, D.X., 1993b. Relation between the amount of fluoride infused from tea leaves and fluoride intake by human. Chinese Journal of Tea Leaves of Fujian Province 1993(4), 15–19.

Sha, J.Q., Zheng, D.X., 1994a. Fluoride in the soil of plantations of Fujian Province. Chinese Journal of Soil Science 25, 230–233.

Sha, J.Q., Zheng, D.X., 1994. Study on the fluoride content in fresh leaves of tea plant planted in Fujian Province. Chinese Journal of Tea Science 14, 37–42.

Street, J.J., Elwaly, A.M.O., 1983. Fluoride solubility in limed acid sandy soils. Soil Sci. Soc. Am. J. 47, 483–485.

Thomas, G.W., 1996. Soil pH and soil acidity. In: Sparks D.L., et al. (Eds.), Methods of Soil Analysis. Part 3. Chemical Methods. 2nd Edition. Soil Sci. Am., Madison, WI. pp. 475–490.

Wenzel, W.W., Blum, W.E.H., 1992. Fluoride speciation and mobility in F-contaminated soils. J. Soil Sci. 153, 357–364.

WHO, 1970. Fluorides and Human Health, Geneva, World Health Organization (Monograph Series No. 59).

WHO, 1984. Fluorine and Fluoride, Geneva, World Health Organization (Environmental Health Criteria 36).