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Review

Tea and fluorosis

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Abstract

Tea trees (*Camellia sinensis*), abundant in fluoride, selectively absorb F from the soil and air in the surrounding, and accumulate mainly in the tea leaves in the form of Al and F complex. Long-term consumption of high fluoride tea could result in chronic fluoride intoxication. This review summarized those data of the fluoride content in various tea commodities, and estimated the risk of fluorosis caused by high fluoride tea commodities. We also introduced fluorosis caused by tea from case reports, epidemiology observations and animal models. Fluorosis was easily misdiagnosed and over-looked and people made little acquaintance in the high fluoride in some tea commodities. So it is the time to pay more attentions on the over-looked safety problem of tea and tea products and some measures should be taken to the fluorosis caused by tea. Large-scale epidemiological investigations and further studies on tea-type fluorosis are in need for those tea-drinking areas. In addition, it is urgent that governmental and international agencies adopt safe standards of fluoride content in tea commodities.

Keywords: Tea; Fluoride; Fluorosis; Food safety

Contents

1. Introduction

Tea tree, rich in fluoride, selectively absorbs F^- from the soil and air around and accumulate them. The longer the absorbing time is, the higher the fluoride content in tea trees is [1]. Tea is also a hyper-accumulator of aluminum. Free Al^{3+} and F^- in acidic soil absorbed by tea tree transformed into AlFx complex in tea tree [2]. So there are high F and Al level in tea leaves which increase with the age of tea leaves [3]. Brick tea is fermented and baled tea, the fluoride content of which is over 680 mg/kg, some even up to 1175 mg/kg. The fluoride in other tea commodities is also very high.

Large epidemiology observations showed that fluorosis could be caused by long-term consumption a large quantities of brick tea by some inhabitants of Tibet, Mongolia and some minorities in the west of China [4]. Bileissi et al. found that the dental fluorosis in children was positively associated with the daily mean of tea drinking in a chronic dental fluorsis area in Jordan [5]. Some similar cases were reported. And recently, an American patient with osteosclerosis and cortical elephantiasis

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in her spine was confirmed to have skeletal fluorosis due to her long-term intake of large quantities of instant tea [6].

The authors attempt to review the attributes of tea plants and the types of tea products, with emphasis on the evaluations of fluoride content in various tea productions, and the fluorosis resulting from tea drinking. The author analyzed reasons for the problem and also suggested some indications for further researches.

2. Fluoride level in various tea commodities and tea beverages

Many studies measured the fluoride content in tea commodities from USA, UK, Germany, Japan, China, Hong Kong and Taiwan in China, Poland, Turkey and so on, including most kinds of tea commodities in international markets [3,6–17]. The range of the fluoride content in tea commodities were summarized in the following table, in which tea commodities were classified by their different manufactured methods and various appearances [3,6–17].

From Table 1, we can find that there are marked differences in the commodity teas of the same type. Brick tea, which is fermented and pressed into brick-shape and made by old and coarse tea leaves, is the greatest in F level. We should point out is that the brick-tea sample with the lower limited of 52.5 mg/kg reported by Zhou (1988) is produced by Yunnan of China, and is named ''Pu'er tea'' [7]. This kind tea is sold in China Mainland, Taiwan and Hong Kong, and even parts of southeastern Asia regions. Pu'er tea is different from the ''bianxiao tea'' which is special for the minority ethnic groups in the west of China and inner Mongolia regions. It is indicated that there is a significant difference between the ''Pu'er tea'' produced by Yunnan and ''bianxiao tea'' produced by Hunan, the F level of which is 76.7 ± 3.2 and 440.3 ± 3.9 mg/kg, respectively [18].

Those reports regarding the F level in black tea do not explain the type of observed black tea samples. Actually, black tea in the international market can be classified into three types: stick-shaped black tea, granular black tea and black tea bags according to different manufactured forms. It is known that the granular black tea occupied 90% of the black tea amount on market. Little of the F level in granular black tea had been recognized, however. Recently, Cao et al. [19] also evaluated the fluoride levels of black tea, including 6 products of stick black tea, 6 products of granular black tea and 25 famous-brand black tea bags collected from the market in UK, USA, Japan, Sri Lanka, China, Hong Kong special administration zone. The results showed wide differences in fluoride levels among three types black teas and the fluoride content was 0.95–1.41 mg/L in black tea sticks, 0.70–2.44 mg/L in black tea granules and 1.15–6.01 mg/L in black tea bags, respectively, the fluoride content was greatest in black tea bags because black tea bags are made of low cost, and older tea leaves.

It is known that the taste and style of tea is markedly associated with the chemical ingredient of polyphenol, caffeine and amino acid in tea [20,21]. One study determined F and polyphenol or amino acid in 14 kind of tea samples collected from China, and the results showed that the F level in tea increased with the maturity of the leaves, conversely, the tea polyphenol and aminophenol declined as the tea leaves grow. Lu et al. also found that it was a significantly negative relationship between the fluoride content and the quality grade of tea. It was indicated that F content in tea could be used as a marker for tea quality evaluation [8].

In order to avoid from the adverse effects of the caffeine, decaffeinated tea was invented and accepted widely soon. It was reported the F content in the decaffeinated tea was significantly higher than that in caffeinated tea due to the use of mineral water containing a naturally high fluoride level during the process of decaffeination [22]. The effect of brewing

Table 1
F content in various commodity teas and tea beverages (ppm)

Tea types	F content range (ppm)
Tea Leaves	
Green tea	2.1–550.0 mg/kg (China) [3,7,8]; 217 ± 26 to 336 ± 27 mg/kg (Hong Kong) [9]; 71.11–180.16 mg/kg (Japan) [10]
Oolong tea	170 ± 6–224 ± 13 mg/kg (Hong Kong) [9]; 87–176 mg/kg (China) [8]
Black tea	23.6–385 mg/kg (China) [7,8]; 322 ± 5 to 423 ± 27 mg/kg (Hong Kong) [9]; 30–340 mg/kg (Poland) [11]; 87.6–289.2 mg/kg (Turkey) [12]; 35 ± 6 to 182 ± 20 mg/kg (Iran) [13]
Flower tea	31.5–636.4 mg/kg (China) [3,7,8]
Brick tea	52.5–1175.0 mg/kg (China) [7]; 680 ± 30 to 878 ± 60 mg/kg (Hong Kong) [9]
Instant tea	
Green tea	260.3 ± 0.02–597.5 ± 0.01 mg/kg (China) [14]
Oolong tea	248.2 ± 0.02 mg/kg (China) [14]
Black tea	0.49 ± 0.03 to 3.35 ± 0.02 mg/L (Germany) [15]; 1.0–6.5 mg/L (USA) [6]; 151.0 ± 0.02 to 631.3 ± 0.02 mg/kg (China) [14]
Flower tea	196.3 ± 0.01 mg/kg (China) [14]
Tea beverage	
Green tea	0.53–0.90 mg/L (Japan) [10]; 0.21–1.474 mg/L (China) [14,17]; 22.6 ± 0.59 to 25.3 ± 0.54 mg/L (Taiwan) [16]
Black tea	0.47–2.19 mg/L (Japan) [10]; 0.25 ± 0.00 to 1.79 ± 0.01 (Germany) [15]; 0.1763 ± 0.0060 to 2.1954 ± 0.0554 (China) [14,17]; 19.9 ± 0.94 to 33.40 ± 0.97 mg/L (Taiwan) [16]
Oolong tea	0.75–1.50 mg/L (Japan) [10]; 0.0565 ± 0.0055 to 4.1068 ± 0.0644 mg/L (China) [14,17]; 21.8 ± 1.02 to 28.5 ± 0.34 mg/L (Taiwan) [16]
Flower tea	0.19 ± 0.01 to 2.2777 ± 0.0312 mg/L (China) [14,17]
Fruit tea	0.03 ± 0.01 to 0.09 ± 0.01 (Germany) [15]; 0.6708 ± 0.0130 to 4.4522 ± 0.1152 (China) [17]

method on the F content of the brewed black tea was determined; it was shown that F in traditional brew tea was higher than micro-waved tea [23].

It is well known that proper fluoride could prevent dental caries and osteoporosis. However, exposition to excessive fluoride long term could lead to chronic fluoride intoxication.

However, it is being argued that the fluoride intake tolerance recommended by international organization and governments had been largely drawn up by pro-F dental researchers under the mistaken notion that controlled ingestion of F has significant anti-caries dental benefit. So, concerns about fluorosis caused by the high F content in tea commodities should be raised.

3. Fluorosis caused by tea drinking

3.1. Case reports

Before the middle age of 20th century, the fact remained that physicians knew little about fluorosis caused by fluoride in tea. A case in point was that a 57-year-old Englishman was originally diagnosed as Paget's disease with osteoarthritis for legs paralysis, flexor spasm and low back pain in 1959 [24]. His symptom went worse after surgical therapy. In 1963 he was ultimately confirmed as fluorosis based on radiography and historic review indicated that he had only exposed to high fluoride due to his long-term heavy tea-drinking habit.

Another definite case of fluorosis caused by tea might be reported by Cook in 1968 that a Pakistan woman had got dental fluorosis, who had drank tea from age two and never lived where water contained fluoride [25].

A case of brick-tea fluorosis was that a 36-year-old woman from Hasake ethnic group in China was presented with pains from the upper limbs to the lumber, both knees and even the joints, also limitation of activity of elbow for more than 10 years, who was diagnosed as hypoparathyroidismrenal osteodystrophy. After treatment according to the hypoparathyroidismrenal osteodystrophy, however, her symptoms in bone and articulus still existed after treatment. Examinations showed that the urine F content reached up to 5.04 mg because she had a history of long-term brick-tea drinking and her daily intake of brick tea amounted to 33.33 g. After 3 months treatment by F inhibitors like Mg, her pain in joint disappeared. When stopping drinking tea, she became normal with the urine F content except slight limitation of right elbow. The course of this case is very similar with the fluorosis patient caused by black tea [26].

A team of French doctors identified 5 patients in their practice who developed skeletal fluorosis as a consequence of drinking tea (primarily Darjeeling tea) over a course of 10–25 years. The skeletal fluorosis in these patients was the osteomalacic variety of the disease, in which the bones become softened and weak [27].

Recently, an American patient with osteosclerosis and cortical elephantiasis in her spine was confirmed to have skeletal fluorosis due to her long-term intake of large quantities of instant tea, after stopping drinking instant tea, her symptom in bone was disappeared and her F level in blood and urine became normal soon [6].

An ongoing study in India has shown that black salt and black tea except unsafe fluoridated water may cause fluoride poisoning, and consuming them might lead to not only bone and joint pain, anemia, fatigue and blockage of blood vessels but also destruction of the stomach and intestinal lining. The study has also shown that drinking black tea and eating ready-made spices, chat-papri, namkeens and pickles that contain black salt for pregnant women could mean giving birth to a low-weight child who would attract diseases fast and the baby's organs may not be properly developed [28].

The problem is that some studies did not describe the detail about black tea consuming traditionally in some countries, such as Pakistan, Jordan, Poland, Britain. It is clear that some cases experienced the process of misdiagnosis. So it is certain that some heavy-tea drinkers suffering from fluorosis caused by tea drinking might not be diagnosed.

3.2. Epidemiology observation

It was determined that mean fluoride concentration on their respective infusion was2.36 mg/L in 12 most popular brand of tea obtained in Valparaiso, Chile. It was about 22.1% had risk of dental fluorosis after determining the amount of ingestion in children in a fluoridated area [29].

Cook carried out a pilot survey on the fluid intake, drinking-habits, tea consumption, and fluoride ingestion of 662 children aged 5–15 in 9 schools in Pakistan. They found that tea was the main sources of the F exposure for children there and there was 1.26 mg from tea drinking among the total daily F intake of children 2.24 mg [30].

In 1988, Blleblasl et al. indicated that the severity of dental fluorosis in Jordanian children might be associated with the excessive tea drinking after conducted an epidemiological study for dental fluorosis from 3 schools in Jordan. They analyzed the F content in water, and recorded the cups of tea taken daily by each child [5].

It was reported that brick-tea fluorosis was only seen in China. Since 1993, a series of epidemiological investigations have been conducted for the Tibetans, Mongols, Hasake and Yugu ethnic groups in the western and northern parts of China. The data covered the prevalence of dental fluorosis and skeletal fluorosis by Horowitz's TSIF Standard, total F intake per person, urine and blood F level, the F content in brick tea, the other sources of F exposure and so. The results were listed in Tables 2–4.

In addition, it is high prevalent on dental fluorosis among children in those brick-tea endemic area, but skeletal fluorosis does not happen frequently until it become apparent in adulthood. Cao et al. observed that among 111 children who had dental fluorosis in Naqu of Tibet, 96 (86.5%) were found developmental skeletal abnormalities in the wrist by radiological examination because of the F daily intake in a child amount to 9.435 mg [34]. These findings as warning signs of early-stage skeletal fluorosis indicated that dental fluorosis in

Table 2
Relation between F daily intake and the prevalence of dental fluorosis in children [4,31]

Subject (ethnic group and area)	The total F daily intake (mg/person/day)			The prevalence of dental fluorosis (%)
	From tea	From others	Total	
Yugu in Gansu	5.30	0.31	5.61	75.75
Kazak in Gansu	5.54	0.51	6.05	84.42
Mongol in Gansu	2.58	0.83	3.41	51.75
Tibetan in Qaqu	8.92	0.025	9.42	82.66
Tibetan in Lasa	7.87	0.104	7.97	52.89
Tibetan in Dangxiong	8.92	0.036	8.96	75.93

Table 3
Relation between F daily intake and the prevalence of skeletal fluorosis in adult [31,32]

Subject (ethnic group and area)	The total F daily intake (mg/person/day)			The prevalence of skeletal fluorosis (%)
	From tea	From others	Total	
Tibetans in Sichuan	13.0	1.21	14.21	97.14
Tibetans in Tibet	8.05	3.94	11.99	89.00

children should be considered more than a matter of cosmetic concern.

3.3. Animal studies

As early as 1936, Reid E had established that rats fed with tea and infusions had produced dental fluorosis [35].

Cao et al. demonstrated that measurements of fluoride intake, fluoride content in urine, blood, bone and dental were made in those rats fed with brick-tea infusions (F^- 245 mg/L) during 30, 60 and 90 days, and severe fluorosis occurred successfully after 30 days [36]. In another laboratory observation, rats were treated with the brick-tea infusions (F^- 5.04 mg/L) for 1 year, which was less than the actual F concentration in tea consumed by the Tibetans. The group developed dental fluorosis characterized as brown and white horizontal marks at the end of 8 months and white chalky dental fluorosis developed at the end of 12 months, and the total incidence reached up to 75%. Compared with the control group, the bone F level in treated group increased 2.17 times. The F metabolism observation suggested that most parts of F were accumulated in skeletal system except those F excreted outside from the urine and excrement [37].

The data were listed in Table 5: So far, little animal experiments on fluorosis caused by tea drinking, further investigations were required badly.

4. Control and prevention

Research thus far indicates that the manifestations of fluorosis are irreversible. However, Gupta et al. observed that the ingestion of a combination of calcium, vitamin D, and ascorbic acid supplementation is effective in protection from fluoride toxicity to a certain extent at lest in children [38]. Some animal experiments showed that protein supplementation in diet is conducive to overcoming fluoride induced oxidative stress in mice testis and has a beneficial effect on liver function [39,40]. Recently, Verma et al. reported that black tea extract significantly ameliorated F-induced hemolysis in RBC suspensions, however, the effect of hemolysis was overcome partly when beyond 40 μg/ml of black tea extract because of the presence of F in tea [41]. But usually, the F content in tea was negative associated with the content of tea polyphenols [8].

In order to prevent and control the fluorosis caused by brick tea in Tibet, Cao et al. developed the low-fluoride brick tea with F content of about 210 mg/kg. It was confirmed by 1-year animal investigations that the low-fluoride brick tea could controlled the prevalence of fluorosis effectively [37]. Intervention trails are currently underway in some brick-tea-type fluorosis endemic areas. The daily F intake of Tibetan children in school decreased from 6.23 mg/day/person to 1.65 mg/day/person after consuming the low-fluoride brick tea

Table 4
Main radiographic features and typing of 35 cases of brick-tea-type skeletal fluorosis [33]

	Trabecular changes Interosseous membrane	Ossification and tendon attachment calcification	Articular degeneration	Thick bone syndrome	Increasing (sclerosis type)	Decreasing (porosis type)	Alternating (mixed type)	Total
Cases	30	22	11	12	26	3	6	35
Constitutive (%)	86	63	31	34	74	9	17	100

Table 5
F daily intake,F excretion, F in serum and bone and fluorosis prevalence in rats after 4, 8, and 12months treatment with brick tea [37]

Group		F daily intake (mg)	F Excretion (mg)	Serum F (mg/L)	Bone F (mg/kg)	Fluorosis prevalence (%)
C	Month 4	0.30 ± 0.07	0.14 ± 0.06	0.07 ± 0.009	922.5 ± 69.12	0
	Month 8	0.26 ± 0.05	0.16 ± 0.08	0.05 ± 0.007	1179.3 ± 101.56	50
	Month 12	0.32 ± 0.04	0.16 ± 0.09	0.05 ± 0.008	1535.5 ± 98.12	75
N	Month 4	0.11 ± 0.02	0.04 ± 0.01	0.04 ± 0.007	288.2 ± 7.81	0
	Month 8	0.11 ± 0.02	0.04 ± 0.009	0.04 ± 0.006	342.6 ± 20.12	0
	Month 12	0.12 ± 0.03	0.05 ± 0.009	0.04 ± 0.006	367.6 ± 20.31	0

C: brick tea treated group; N: negative control group.

instead of the high fluoride one for 4 weeks [42]. A 3-year observation in Dangxiong of Tibet showed the low-fluoride brick-tea consumption decreased the total daily F intake from risk level, 7.55 mg/day/person, to safety level, 2.43 mg/day/person, among the children. The urine F content was not altered essentially and the reason might be that the accumulated F in bone was still excreted outside from the urine and excrement after the patient escaped from exposure to F and the process remained constantly and slowly [43]. The 5-year project for prevention and control the endemic fluorosis caused by brick tea has been in progress in the western and northern areas of China.

5. Perspectives

1. Previous studies showed that there were significant differences among those tea commodities in F level and confused conclusions would be made by those experiments with less samples which could not evaluated the F level of tea commodities comprehensively [14]. Therefore, it is very necessary to conduct further large-scale experiments with sufficient samples for F measurement and evaluation in tea commodities.
2. From those previous cases of fluorosis caused by black tea or brick tea, we can find that both of them experienced misdiagnosis. The main reason lies on that fluorosis is a chronic disease with the similar symptoms with articular disease, rheumatism and so on. In addition, the prevalence of brick-tea fluorosis was partly ascribed to lack of knowledge in fluorosis in those remote areas except those with high F water traditionally.
3. Further epidemiological investigations should do since there were some case-reports of fluorosis led by black tea from Europe and America. It is also doubted that if there are any threats exist in some green tea-drinking areas. Apart from dental and bone, fluorosis also has bad effects on some soft tissues. Human studies showed that high levels of fluoride exposure could lead to gastrointestinal disorders and reduced cognitive ability in children [44,45]. Pathological changes in kidney, liver, brain and even spermatids and epididymal spermatozoa were seem significantly in those animals exposed to excessive fluoride [46–49]. So, further investigations are required in epidemiology observation and animal models.
4. A review had described the present statues of the fluorosis in the European Union and concluded the sources of the F exposure including fluoridated drinking water, toothpaste, F supplements, salt and milk. However, tea as an F exposure source was ignored [50]. The author suggested that lack of use of a standardized method for measurement of fluorosis made comparison between studies difficult and assessment of trends in fluorosis prevalence unreliable. It was concluded that co-ordinate studies are necessary for measuring fluorosis throughout Europe and that development of a standardized photographic method would be useful. What the author posed is just the problem confronted with in the study of tea fluorosis. International organization and the governments should combine and establish uniform methods for F measurement and fluorosis examination.
5. Lack of the F standard in tea commodities contributes to so significant variance in F content in tea commodities. It is the time that governments and international agencies adopted safe standards of fluoride content in tea commodities.

More and more health effects on tea drinking have been studied. Tea drinking seems to be more fashionable. Therefore, the overlooked safety problem should be highlighted now.

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